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RADIOECOLOGICAL STUDIES
ON THE
COLUMBIA RIVER
Part I

by
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Part II of this report, Appendices A and B, contain the tabular data of all analyses and is available from the authors on request.
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ERRATA

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RADIOECOLOGICAL STUDIES

IN THE

COLUMBIA RIVER

INTRODUCTION

The use of Columbia River water as a reactor coolant and the subsequent discharge of this cooling water into the River introduces a number of radioactive elements into the river environment. These radio-nuclides are produced by the neutron activation of stable elements in solution in the cooling water and the sloughing off of radioactive corrosion products from the surfaces of the reactor cooling tubes. The relationships of these radioactive materials to river organisms have been studied since reactor operations began in 1944 (1-10). Radioanalyses in the earlier studies (3, 4), dealing mainly with seasonal variations, species differences, and geographical distribution of radioisotopes by the river biota, were confined to the measurement of total beta emitter activity and to the estimation of relative amounts of phosphorus-32 through decay curve analysis.

The purpose of this study is (1) to define the interspecies and seasonal variations in the concentration of several of the more biologically important radionuclides, and (2) to update the findings of some of the earlier investigations.
period.

The establishment of hydroelectric dams upstream from Hanford has modified the river environment. Between 1956 and 1967 three low-head dams were built at distances from 20 to 147 km upstream from the reactors. Priest Rapids Dam, immediately upstream from the Hanford Reservation, exerts the greatest influence on the local river conditions. River plankton species composition and quantity have changed markedly (13). Dominant species are lentic rather than lotic. The retention and release of water at Priest Rapids, to satisfy varying daily electrical power demands, produces average diurnal river elevation fluctuations of 1.8 m, with maximum as high as 3.8 m immediately downstream (Fig. 2). Daily fluctuations of 2 m were common near our sampling sites.

Diurnal changes in river flow were correspondingly great, with the maximum often more than double the minimum. A low-head dam, such as that at Priest Rapids, exerts its greatest influence during average or low flow periods (fall and winter), and least during the fresher (spring and early summer).

These changes in hydrographic conditions have destroyed much of the littoral zone productivity. Near-shore production of benthic forms, particularly sessile algae, has been reduced due to restrictions in light penetration resulting from higher turbidity and greater mean water depth because of diurnal water level fluctuations. The stability of shoreline habitat used as feeding and resting areas by immature fish is destroyed. The rapid variations in river elevation have nearly eliminated organisms such as crayfish and water boatmen (Corixidae) from littoral zone areas of former abundance.
FIGURE 2. CONCENTRATION OF RADIONUCLIDES IN COLUMBIA RIVER WATER
METHODS

All samples, except large fish, were collected near 100-F area (Fig. 1), about 13.5 km downstream from the nearest reactor effluent outfall in a zone of nearly complete effluent mixing. The sampling site was characterized by a gently sloping bottom, substrate ranging in size from fine sediment to rubble \(1\) dm in diameter (average approximately \(1\) dm), and water depth fluctuating daily from 0.5 to more than 2 m.

Samples were obtained by wading during morning low water periods. Plankton was collected with 70 \(\mu\) mesh nets suspended in the river currents, periphyton and invertebrates were picked or scraped from stones from the bottom, and small fish were collected with a hand seine in small inlets along shore. All samples were usually processed and submitted for radioactivity measurement within 2 hr after collection.

Large fish were collected near White Bluffs in an area about 11 km downstream from the nearest reactor where low current velocities permitted the use of gill nets. They were refrigerated until processed in the laboratory, usually on the day of collection.

Sample processing was as follows:

1. Periphyton, sponge, caddisfly larvae - washed in river water, debris removed, excess moisture blotted with paper towels.

2. Plankton - water decanted from settled sample, excess moisture blotted with paper towels.

3. To standardize the removal of surface moisture, the above samples were placed in 20 ml plastic tubes with a water
reservoir in the bottom and centrifuged for 5 min; caddis fly larvae at 700 rpm, remainder at 2000 rpm.

(4) Limpets - sediment and algae washed from shell, steamed over a water bath for about 3 min, shell and soft parts separated.

(5) Small fish - surface moisture blotted, gut contents removed, macerated to form a homogenate.

(6) Large fish (at least 6 individuals of each species sampled when available) - gut contents removed, sample from anterior gut retained for radioanalysis, fish separated into muscle and "carcass" (remainder) fractions, ground to a homogenate.

Gamma emitter analyses:

(1) Plankton, periphyton, sponge, caddisfly larvae, limpets, small fish, and large fish gut contents - three aliquots of about 4 ml equivalent volume placed in the bottom of 20 ml glass test tubes, 1 ml 10% formalin added to inhibit decay.

(2) Large fish muscle and "carcass" - a 125 to 500 ml equivalent volume of each tissue from each fish put in 500 ml polyethylene bottle.

(3) Gamma measurement done with either a 3" x 3", or 9" x 11" NaI well crystal with readout on a 400 channel analyzer. Measurements were made for the following radionuclides: sodium-24, scandium-46, chromium-51, manganese-54, iron-59, cobalt-60, zinc-65, zirconium-niobium-95, ruthenium-rhodium-106, iodine-131, cesium-137, barium-140, lanthanum-140, cerium-praseodymium-144,
and neptunium-239. Data transferred to punch cards for computer analyses.

(4) Initial computation of concentrations of gamma emitters was made by use of the GEM program (15); further analyses were made on output from a FORTRAN program.

Beta emitter analyses:

(1) Total beta - a single aliquot of 1 to 1.5 g wet wt spread evenly and dried on a 1" stainless steel planchett, counted with a wide beta, thin window, gas flow proportional analyzer.

(2) Phosphorus-32 - measured by a differential absorber technique (14), on the same sample and counting system used for total beta. A decay period of 7 to 10 days allowed before counting for phosphorus-32 for decay of interfering short-lived radio-nuclides.

(3) Results were calculated on a special computer program.

Standard weights:

(1) One to five grams wet sample taken on each series of samples, dried at 60 C and 15" of mercury vacuum for 24 hr for standard dry weight determination.

(2) Sample then ashed at 425 C for a minimum of 24 hr for determination of ash weight.

(3) All samples cooled to room temperature in a dessicator before weighing.
Stable element analyses:
Measured by flame spectroscopy (Na, K), colorimetric (P), and atomic absorption (Ca, Mg, Cu, Zn, Fe, Mn, Cr, and Co) techniques:

RESULTS AND DISCUSSION

Water

A number of radionuclides are introduced into the river in the reactor effluents but only a few are of biological significance. Some with short half-lives are quickly lost from the ecosystem through radioactive decay, while others are not accumulated to any extent by the river biota. Zinc-65 and phosphorus-32 are the radionuclides of greatest biological importance and chromium-51 is the most abundant. At Richland, about 45 km downstream from the reactors, chromium-51 accounts for approximately 50% of the total radioactivity in the water, and zinc-65 and phosphorus-32 contribute 2% and 1%, respectively (16).

The concentrations of chromium-51, zinc-65, and phosphorus-32 in water, collected with a continuous sampler near the organism sampling sites, are given in Fig. 2. The 1967 values for July through September replace the measurements for the same period in 1966 when the normal seasonal patterns were disrupted by the previously mentioned reactor closures. This replacement of one year's values for those of another is made with the realization that the precise pattern and quantity of radionuclides in the water may vary in consecutive years, but this approach is considered reasonable for the general illustration of seasonal trends. The effluent discharge rate is relatively constant so seasonal
variations in water radioactivity are inversely related to river flow. This is apparent by comparison of seasonal trends in Fig. 2 with variations in river flow shown in Fig. 3.

Factors other than dilution may also influence the radionuclides in solution in river water. Zinc-65, manganese-54, zirconium-niobium-95, iron-59, and scandium-46 are associated mainly with particulates (17) and the concentrations of these isotopes in solution would be reduced by suspended silt in the River. These nuclides are depleted more rapidly with transport downstream than is chromium-51, for instance, which remains mainly in solution.

The concentrations of some radionuclides in water were slightly higher in 1967 than previously reported measurements. In September 1957, the zinc-65 and chromium-51 values were 89 pCi/l and 2000 pCi/l, respectively (7); in September 1967, they were 200 pCi/l and 4000 pCi/l, respectively. The 1957 measurements were made on a single grab sample and may be less representative than the analyses of integrated samples in 1967.

River organisms

Some of the 15 radionuclides for which analyses were made in the river biota were not present consistently and in sufficient amounts to be useful in demonstrating seasonal trends. Concentrations of phosphorus-32, zinc-65, chromium-51, manganese-54, iron-59, zirconium-niobium-95, and, in some instances, scandium-46 were measurable in most organisms throughout the year. To permit comparison with results of earlier
studies (3, 4), analyses were also made for total beta activity which will be discussed later. Sodium-24 was present in most biological materials, but its relatively short half-life (15 hrs), the lapse of several days between collection and lab processing and subsequent radioactivity counting, and the associated uncertainty of decay corrections, made the analytical results for this isotope questionable. Consequently they are not discussed in this report. The mean values of radionuclide concentrations in all samples are presented in Appendices A and B.

The seasonal variations in concentrations of phosphorus-32, zinc-65, chromium-51, manganese-54, iron-59, and zirconium-niobium-95 in representative river organisms are shown in Figs. 4 to 21. Similar to the treatment of the water data, the 1967 July through September biota values are substituted for the same values in 1966 when the reactor shutdown altered normal seasonal patterns. Values plotted on the graphs are the means of replicate analyses, when available. Values less than twice the associated counting and computation error were rejected and treated as zero in the calculation of the mean values. This tended to produce many zero values, shown as baseline points in the Figures, during seasons of the year when concentrations of certain radionuclides fluctuated around this cut-off point. This is particularly evident for the manganese-54 and zirconium-niobium-95 levels in net plankton (Figs. 5 and 6).

Variation in river flow, which produces changes in the dilution of the reactor effluents, and river temperature, which affects metabolic rate, are two of the most influential factors in the control of radionuclide content of the river biota. Comparison of river flow and
FIGURE 4. CONCENTRATIONS OF PHORPHORUS-32 AND ZINC-65 IN NET PLANKTON

FIGURE 5. CONCENTRATIONS OF CHROMIUM-51 AND MANGANESE-54 IN NET PLANKTON
FIGURE 6. CONCENTRATIONS OF IRON-59 AND ZIRCONIUM-NIOBIUM-95 IN NET PLANKTON

FIGURE 7. CONCENTRATIONS OF PHOSPHORUS-32 AND ZINC-65 IN PERiphyton
FIGURE 8. CONCENTRATIONS OF CHROMIUM-51 AND MANGANESE-54 IN PERIPHYTON

FIGURE 9. CONCENTRATIONS OF IRON-59 AND ZIRCONIUM-NIOBIUM-95 IN PERIPHYTON
FIGURE 10. CONCENTRATIONS OF PHOSPHORUS-32 AND ZINC-65 IN CADDISFLY LARVAE, HYDROPSYCHIDAE

FIGURE 11. CONCENTRATIONS OF CHROMIUM-51 AND MANGANESE-54 IN CADDISFLY LARVAE, HYDROPSYCHIDAE
FIGURE 12. CONCENTRATIONS OF IRON-59 AND ZIRCONIUM-NIOBIUM-95 IN CADDISFLY LARVAE, HYDROPSYCHIDAE

FIGURE 13. CONCENTRATIONS OF PHOSPHORUS-32 AND ZINC-65 IN LIMPET SOFT PARTS AND SHELL, _Fisherola nuttalli_

FIGURE 15. CONCENTRATIONS OF IRON-59 AND ZIRCONIUM-NIOBIUM-95 IN LIMPET SOFT PARTS AND SHELL, *Fisherola nuttalli*. 

FIGURE 17. CONCENTRATIONS OF PHOSPHORUS-32 AND ZINC-65 IN MUSCLE OF LARGESCALE SUCKER, Catostomus macrourus, CHISELMOUTH, Acrocheilus alutaceus, AND NORTHERN SQUAWFISH, Ptychocheilus oregonensis.
FIGURE 18. CONCENTRATIONS OF PHOSPHORUS-32, ZINC-65, CHROMIUM-51, AND MANGANESE-54 IN LARGESCALE SUCKER CARCASS, *Catostomus macrocheilus*

FIGURE 19. CONCENTRATIONS OF PHOSPHORUS-32, ZINC-65, AND CHROMIUM-51 IN NORTHERN SQUAWFISH CARCASS, *Ptychocheilus oregonensis*
FIGURE 20. CONCENTRATIONS OF PHOSPHORUS-32, ZINC-65, IRON-55 AND ZIRCONIUM-NIOBIUM-95 IN LARGE SCALE Sucker Gut CONTENTS, *Catostomus macrocheilus*

FIGURE 21. CONCENTRATIONS OF CHROMIUM-51, MANGANESE-54, AND SCANDIUM-46 IN LARGE SCALE Sucker Gut CONTENTS, *Catostomus macrocheilus*
temperature (Fig. 3) with the seasonal fluctuation in radionuclide burden in the organisms shows this relationship. In general, minimum levels for most radionuclides in both plant and animal forms occurred during the late spring and early summer, and were primarily the result of maximum dilution of reactor effluents by the annual freshet. High turbidity, which limits autotrophic metabolism through the restriction of light, and increased quantity of suspended sediments offering more sites for isotope adsorption also contribute to the lower concentrations of radionuclides during periods of high flow.

The season of maximum radionuclide burden was different in plant and animal forms. Highest levels in animals usually occurred in late summer and early fall when river flows were decreasing and water temperatures were at the yearly maximum. The slight increase in radionuclide concentrations accompanying the spring rise in river temperatures was reversed by dilution of the annual runoff. Concentrations in the autotrophic organisms, plankton and periphyton, showed maximums in late fall and winter when both flow and temperature were at a minimum, and radionuclide concentrations in the water at a maximum.

Phosphorus-32 was generally the most abundant nuclide in the biota and showed a relatively large seasonal variation. Differences between the annual minimums and maximums ranged from 10 to 100 fold, and greatest seasonal changes occurred in large fish tissues. Concentrations of 200,000 pCi phosphorus-32/g dry wt were observed in periphyton and there was a general decrease in concentration with progressively higher trophic levels. The comparatively high levels of phosphorus-32 in river organisms are due to the low concentrations of stable phosphorus in the river water.
(ca 0.01 ppm \( \text{PO}_4 \)) and to its high biological concentration (18), thus enhancing the relative amounts of radioactive phosphorus.

Zinc-65 was generally the second most abundant radionuclide in the biota. Its concentrations and seasonal variations were almost identical to those of phosphorus-32 in net plankton. Its concentrations in periphyton, composed mainly of sessile plant forms, was appreciably lower than that of phosphorus-32. The net plankton, primarily phytoplankton, is derived to a large extent from the river upstream from the reactor outfalls, although the sloughing of radioactive periphyton downstream from the reactors contributes to the radionuclide burden of the plankton that was collected in this study. The exposure of the plankton to the reactor effluents is relatively brief and equal to the few hours necessary for the river to travel from the points of effluent discharge to the collection site. Plankton exposure to radionuclides is confined to those contained in the water mass in which it is traveling and therefore reflects short term fluctuations in the radioactivity of the water. Much of its radioactivity is adsorbed to the outer surface and not assimilated by the algal cells. Although both the plankton and periphyton have large surface to volume ratios making surface adsorption an important route of uptake, the periphyton, because of its fixed position acts as an integrator and reflects a smoother seasonal pattern of accumulation.

A definite summer decrease in zinc-65 in periphyton was observed in 1963-64 (10), but is not as evident in the few samples available during the high water period of the present study.

Seasonal variations in zinc-65 concentrations decrease with increase
in trophic level, with very little seasonal fluctuation evident in fish. This is partly due to the long effective half-life of this element in fish. In rainbow trout, a half-life of 134 days has been reported (19), and is probably similar in other fishes.

Chromium-51 was the most abundant radionuclide in plankton and periphyton, was nearly as plentiful as phosphorus-32 and zinc-65 in the invertebrates, but was absent or infrequently observed in fish muscle. It was consistently found in fish "carcass," the portion that included the outer surface of the animals. Radiochromium is of little biological importance as a metabolite, but because of its relative abundance in the water (Fig. 2), much is adsorbed on the outer surfaces of river organisms. Its seasonal variation is much less than that of phosphorus-32 with the maximum concentration usually less than 20 times the minimum. The gut contents of the invertebrates, caddisfly larvae and limpets, are composed largely of algae and detritus, and contribute significantly to the chromium-51 burden in these animals.

The seasonal pattern of concentrations of manganese-54, iron-59, and zirconium-niobium-95 were similar with the maximum usually less than 20 times the minimum. Both iron-59 and manganese-54 showed little seasonal change in fish and zirconium-niobium-95 was not found often enough to show seasonal trends.

**Total Beta Comparisons**

The usefulness of total beta activity measurements is of limited value because of the varying influence of contributing radionuclides.
Total beta analyses do offer a basis, however, for comparing measurements obtained prior to 1956 with those of this study. The direct comparison of the amounts of beta radioactivity found in the present study with that of earlier investigations is hampered by (1) the number of operating reactors, and hence effluent output, changed with time, (2) modifications in cooling water treatment that altered the character of the effluent in terms of amounts and relative proportions of certain radionuclides, and (3) changes and refinement of beta measurement in the biological materials.

Figure 22 presents the seasonal concentrations of total beta activity in four organisms during 1952, 1955, and 1966-67.

In general, seasonal fluctuations and levels were similar for the plankton, caddisfly larvae, and largescale sucker muscle. This is especially remarkable when one considers the changes which occurred in the River during this period as mentioned in the Introduction.

Net plankton concentrations were generally higher in 1952 and the time of the lowest concentrations was in June, a month earlier than in 1955 and 1966-67. Data for caddisfly larvae and largescale sucker are not complete enough for comparison of seasonal trends. Similar patterns, however, and levels of activity are evident where comparison is possible.

Trends in the concentrations of total beta activity in the redside shiners, however, appeared to be different during the January to June period of the three years, but similar in late summer and fall. Seasonal low values occurred at successively later dates; in February in 1952, March and April in 1955, and mid-summer in 1966-67.

Figure 23 shows the total beta concentrations in several repre-
FIGURE 23. INTERSPECIES COMPARISON OF BETA ACTIVITY CONCENTRATION IN COLUMBIA RIVER ORGANISMS, 1955* AND 1966-67
sentative species at four seasons for the years 1955 and 1966-67. Differences between species is greatest during the first quarter of the year when water temperature and flow are near the minimum; least differences occur during the second and third quarters of the year when higher temperatures enhance the metabolism of many of the animal species, and river flows are declining and approaching the annual minimum. A decrease in activity with progression up the trophic levels is very apparent in the first half of the year. This relationship changes in the third quarter when the activities in many of the animals approach or exceed the concentrations in net plankton.

In spite of the changes in reactor operation and sample analyses that limit the direct comparison of quantities of beta activity found in this study with that of earlier investigations, one would expect a greater change with time than is indicated in Fig. 23. With most organisms and at all seasons of the year, the concentrations of beta activity in 1966-67 are much the same as that of 1955. In 1955, eight reactors were operating, and only five in 1966-67. This would seem to indicate that the accumulation of radioactive materials by the river biota is not directly related to the number of operating reactors, but to the total radioactivity in the effluents.

**Gamma Comparisons**

Analytical capabilities for measuring concentrations of gamma radionuclides have been perfected since results of many of the early River studies were completed. Hence, relatively few gamma measurements
are available prior to 1960. Analysis of these early data are also subject to the same qualifications mentioned in the previous section comparing total beta activity.

Comparison of the concentrations of gamma emitting radionuclides are given in Table 1. All data presented in this table were collected during approximately the same time of year (August-October) and from the same general area of the river immediately downstream from 100-F area. Similar to the total beta measurements, no consistent time related trends in the concentrations of specific radionuclides is apparent.

**Effects of Reactor Shutdown**

The shutdown of the reactors in July and August of 1966 produced marked results in the concentrations of radionuclides in the biota. Concentrations of radionuclides decreased rapidly to varying levels. Chromium-51 and phosphorus-32 decreased two to three orders of magnitude in the lower trophic levels; phosphorus-32 levels were not measurable within about seven days after shutdown and chromium-51 levels were below detection limits after about five weeks. The declines in concentrations of zinc-65, manganese-54, and iron-59 were much less and measurable amounts remained present at all times. Decreases of an order of magnitude, however, were common in most organisms. In fish, phosphorus-32 was lost rapidly and zinc-65 more slowly.

The fact that some elements remained present in measurable quantities suggests that quantities of these isotopes were still being contributed
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<td>--</td>
<td>1790</td>
<td>--</td>
<td>66.3</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>140 Ba</td>
<td>--</td>
<td>901</td>
<td>--</td>
<td>42.2</td>
<td>--</td>
<td>--</td>
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<td></td>
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</table>

**Table 1 (continued)**
TABLE 1. (continued)

<table>
<thead>
<tr>
<th></th>
<th>Sessile Green Algae</th>
<th>Sessile Sponge</th>
<th>Caddisfly Larvae</th>
<th>Limpet Soft Parts</th>
<th>Limpet Shell</th>
<th>Minnows</th>
</tr>
</thead>
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<tr>
<td></td>
<td>Plankton</td>
<td></td>
<td></td>
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<tr>
<td>$^{140}$La</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>1957</td>
<td>--</td>
<td>3270</td>
<td>1230</td>
<td>347</td>
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<td>1610</td>
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<td>223</td>
<td>73</td>
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<td>1965</td>
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<td>1330</td>
<td>322</td>
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<td>1967</td>
<td>4630</td>
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<td>2400</td>
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<td>379</td>
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<td>$^{239}$Np</td>
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<td>401</td>
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<td>3010</td>
<td>1750</td>
<td>1080</td>
<td>384</td>
<td>79</td>
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<tr>
<td>$^{32}$P</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1957</td>
<td>--</td>
<td>66000</td>
<td>4460</td>
<td>24300</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>1966</td>
<td></td>
<td>3270</td>
<td>6560</td>
<td>3790</td>
<td>988</td>
<td>7110</td>
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<tr>
<td>1967</td>
<td>12800</td>
<td>15100</td>
<td>28200</td>
<td>19000</td>
<td>2310</td>
<td></td>
</tr>
</tbody>
</table>


(2) Present study September 1967 data.

(3) Present study September 1966 data.

to the river. Two sources contributed radionuclides to the water during shutdown: (1) a small amount of certain nuclides from water pumped through the reactor cooling systems, and (2) release of radionuclides sorbed to the river sediments. Estimates of the percentage decrease of several radionuclides from McNary Reservoir sediments due to leaching and scouring during the shutdown period have been measured (20). These range from 20% for chromium-51 and scandium-46 to 50% for zinc-65 and 75% for manganese-54 and cobalt-60. Cycling of these elements probably provided the radionuclides which were present in the biota during shutdown. All organisms rapidly accumulated the radionuclides following resumption of reactor operation and near equilibrium concentrations in most organisms were approached within two or three weeks. Figures 24 and 25 present typical curves showing the influence of the reactor shutdown on radionuclide concentrations in net plankton and shiners. A fuller description has been published (11).

The changes in zinc-65 concentration, due to reactor shutdown, were measured in estuarine juvenile starry flounders (Platichthys stellatus) near the mouth of the Columbia River (21). After correction of about 19 days for river flow time between Hanford and the river mouth, a lapse of nearly a month occurred before a significant decline in zinc-65 concentration was noted in the starry flounder. This is comparable to the time lapse before a marked reduction in zinc-65 was evident in shiners taken at near the reactors (Fig. 25). After the reactors resumed operations, the concentrations at both sites approached a "normal" zinc-65 concentration within two weeks.
FIGURE 25. RADIONUCLIDE CONCENTRATION IN SHINERS (Richardsonius balteatus) DURING AND AFTER REACTOR SHUTDOWN
Concentration Factors

The concentration factor (CF) is an expression used to estimate the degree to which an organism concentrates a particular radionuclide or element from the ambient water. It is calculated by dividing the pCi/g wet weight of the organism by the pCi/ml of the same radionuclide in the water. There has been a good deal of discussion as to the validity of this measurement and a discussion of its present status has been published (22).

Figure 26 presents the seasonal variations in the concentration factors of zinc-65, chromium-51, and phosphorus-32 in net plankton, periphyton, caddisfly larvae, and redside shiner.

Analysis by Species

Zinc-65 and phosphorus-32 were concentrated to a higher degree by net plankton than chromium-51. Zinc-65 CFs ranged from 3500 to 40,000 with low values occurring in summer and high values in winter. Phosphorus-32 values ranged from 9000 to 68,000, and chromium-51 from 440 to 9400. Zinc-65 and phosphorus-32 CFs ranging from 300 to 19,000 and from 5000 to 118,000, respectively, have been reported for net plankton in the Columbia River in 1963-64 (23). These values include those of the present study for phosphorus-32, but are lower for zinc-65.

The seasonal variations observed in the plankton were also apparent in the other organisms sampled. It is probably related more to the seasonal dilution of radionuclides in the water by high summer flows than to seasonal changes in metabolic activity. The latter would be expected
FIGURE 26. CONCENTRATION FACTORS FOR COLUMBIA RIVER ORGANISMS, 1966-67 (NOTE SCALE DIFFERENCE)
to produce higher CFs during warm weather in summer and low CFs in
colder months. This pertains to that radioactivity which is incor-
porated by metabolic processes governed by temperature and not to that
radioactivity which is adsorbed and is a function of surface to volume
ratios. Chromium-51, for instance, is not biologically important and
is accumulated by adsorption. Zinc-65 is accumulated mainly by
adsorption by plankton and periphyton but by food-chain absorption
by caddisflies.

Zinc-65 CFs in periphyton were again higher than those of chromium-51. Values for zinc-65 ranged from 5000 to 24,000; for chromium-51 from 500 to 4300, approximately an order of magnitude difference.

Phosphorus-32 CFs in Hydropsychidae were about an order of magnitude higher than those for zinc-65, which, in turn, were an order of magnitude higher than CFs for chromium-51. Values ranged from 8400 to 100,000 for phosphorus-32, 22,000 to 97,000 for zinc-65, and from 800 to 9900 for chromium-51. Concentration factors of 100,000 for phosphorus, 2,000 for chromium, and 40,000 for zinc have been reported for fresh water invertebrates (24). These are within agreement with the present results.

Concentration factors for both zinc-65 and phosphorus-32 were considerably lower in shiners than in the net plankton, periphyton, or
caddisfly larvae. Concentration factors for zinc-65 ranged from 900 to
2800; for phosphorus-32 from 1900 to 4200. Concentration factors of
100,000 for phosphorus, 200 for chromium, and 1000 for zinc have been
reported for freshwater fish (24). Zinc-65 CFs for the shiners agree with
these data, but phosphorus-32 CFs are much different. Chromium-51 levels
were not measurable in shiners.
Analysis by Radionuclide

Zinc-65 CFs were highest in the autotrophic plankton and periphyton and decreased through the caddisfly larvae and shiners.

Phosphorus-32 CFs were higher in caddisfly larvae, though only slightly lower in net plankton; lowest values were found in the shiners. No values are available for periphyton since it was unavailable during the period for which we have phosphorus-32 water data.

Concentration factors for chromium-51, a biologically unimportant element, were of a similar order of magnitude for net plankton, periphyton, and caddisfly larvae; levels were not measurable in shiners. Lower values would be expected for this adsorbed element by caddisfly larvae because of their smaller surface to volume ratio as compared to those of the algae.

These data conflict somewhat with the usually observed increase in CFs as one moves up the trophic level, i.e., from autotrophs to herbivores.

Comparison with Total Beta CFs (1955)

Comparison of CFs for total beta, the only data available from the earlier report (4), with those of phosphorus-32 from the present study are tenuous because of the unknown contribution of other beta emitters to the earlier data. Nevertheless, the majority of the activity was probably due to phosphorus-32, so comparative data are given in Table 2.
TABLE 2. Concentration Factors - Total Beta (1955), Phosphorus-32 (1966-67)

<table>
<thead>
<tr>
<th></th>
<th>Total Beta</th>
<th>Phosphorus-32</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Average</td>
<td>Maximum</td>
</tr>
<tr>
<td>Net plankton</td>
<td>1600</td>
<td>2600</td>
</tr>
<tr>
<td>Caddisfly</td>
<td>1300</td>
<td>3000</td>
</tr>
<tr>
<td>Shiners</td>
<td>470</td>
<td>900</td>
</tr>
</tbody>
</table>

In all cases, data from this study showed marked increases over results from the earlier studies. This may be partially related to improved radioanalytical techniques.

Specific Activity

The specific activity of an element in an organism (ratio of radioactive to stable atoms) is perhaps the best measurement of the ecological demand for that element, assuming that certain limitations are met (25). Use of specific activities allows one to predict expected contamination levels in organisms if the stable concentrations are known for the organism and the media and also the expected environmental levels of contamination.

Table 3 presents the specific activities for eight elements in representative Columbia River biota sampled in November and December 1966. Since the stable element analyses for these data were done, we have become aware that the techniques used for analyzing certain elements,
particularly Na and K, have provided questionable results, so that the values in Table 3 should be regarded as approximations.

**TABLE 3. Specific Activities, uCi/g**

<table>
<thead>
<tr>
<th></th>
<th>P</th>
<th>Na</th>
<th>K</th>
<th>Zn</th>
<th>Fe</th>
<th>Mn</th>
<th>Cr</th>
<th>Co</th>
</tr>
</thead>
<tbody>
<tr>
<td>Net plankton</td>
<td>6.03</td>
<td>2.24</td>
<td></td>
<td>24.6</td>
<td>0.624</td>
<td>3.28</td>
<td>1830</td>
<td></td>
</tr>
<tr>
<td>Periphyton</td>
<td>4.93</td>
<td></td>
<td></td>
<td>20.3</td>
<td>0.764</td>
<td>4.85</td>
<td>1570</td>
<td></td>
</tr>
<tr>
<td>Sponge</td>
<td>1.17</td>
<td></td>
<td>14.5</td>
<td>0.417</td>
<td>2.44</td>
<td>972</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Caddis larvae</td>
<td>2.76</td>
<td>0.185</td>
<td></td>
<td>13.3</td>
<td>0.701</td>
<td>3.35</td>
<td>459</td>
<td></td>
</tr>
<tr>
<td>Limpet</td>
<td></td>
<td></td>
<td></td>
<td>8.50</td>
<td>1.59</td>
<td>2.53</td>
<td>35.7</td>
<td></td>
</tr>
<tr>
<td>shell</td>
<td>1.47</td>
<td>0.0969</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>soft parts</td>
<td>0.757</td>
<td>0.540</td>
<td>18.5</td>
<td>0.107</td>
<td>3.28</td>
<td>137</td>
<td>5.90</td>
<td></td>
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<tr>
<td>Shiner</td>
<td>0.810</td>
<td>0.162</td>
<td>16.8</td>
<td>4.56</td>
<td></td>
<td>&gt; 48.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>C.S. sucker</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>muscle</td>
<td>0.134</td>
<td>0.130</td>
<td>0.00158</td>
<td>5.75</td>
<td>0.827</td>
<td>&gt; 0.340</td>
<td></td>
<td></td>
</tr>
<tr>
<td>carcass</td>
<td>0.107</td>
<td>0.176</td>
<td>0.00307</td>
<td>6.71</td>
<td>0.624</td>
<td>60</td>
<td>0.375</td>
<td></td>
</tr>
<tr>
<td>Gut cont.</td>
<td>2.96</td>
<td></td>
<td>11.1</td>
<td>0.0872</td>
<td>1.83</td>
<td>520</td>
<td>2.10</td>
<td></td>
</tr>
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</table>

Table 4 lists some specific activities reported in the literature for comparison with those in Table 3.

**TABLE 4. Specific Activities**

<table>
<thead>
<tr>
<th></th>
<th>Zn</th>
<th>P</th>
<th>Mn</th>
<th>Reference</th>
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<tbody>
<tr>
<td>Clam, soft parts</td>
<td>250</td>
<td>25</td>
<td>26</td>
<td></td>
</tr>
<tr>
<td>Clam, soft parts</td>
<td>3000</td>
<td>27</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fish, whole</td>
<td>0.5</td>
<td>8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chironomid larvae</td>
<td>4</td>
<td>8</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The closest agreement is between the specific activities for phosphorus in the chironomid larvae (4) and that for the caddis larvae (2.76). For comparison, the specific activity of Columbia River water in January 1964, was 3.8 for phosphorus and 225 for zinc.

CONCLUSIONS

A study of radionuclide concentration levels in Columbia River biota was undertaken to (1) define the interspecies and seasonal variations in the concentration of several of the more biologically important radionuclides, and (2) update the findings of some of the earlier investigations.

1. Concentrations of radionuclides in the Columbia River water are generally inversely proportional to flow levels, with highest values in winter and lowest in summer. This is related to dilution by seasonal runoff.

2. Concentrations of most radionuclides in the biota followed a pattern of high levels in winter followed by low concentrations in summer. There were often indications of an increase in concentration just after levels started to decline in spring. This may be attributed to improved light and temperature regimes which favored metabolic uptake; this increase, however, was soon overwhelmed by the dilution of radionuclides by the spring runoff.

3. A remarkable similarity in total beta concentration was observed between the present data and that collected prior to 1956. In
view of the environmental and technological changes which have occurred, this was quite unexpected.

4. No consistent time related trends were apparent in the gamma activity measurements between the present data and that collected prior to 1956.

5. The shutdown of all reactors for several weeks during this study produced marked changes in the radionuclide concentrations in the biota. Levels of most nuclides dropped several orders of magnitude and many were undetectable. Measurable amounts of certain isotopes were present from radioactivity recycled from the sediments or small amounts coming from the reactor effluents. After the reactors resumed operation, levels increased rapidly to pre-shutdown levels or higher.

6. Concentration factors were highest for the biologically important radionuclides phosphorus-32 and zinc-65 and were highest in the primary producers, net plankton and periphyton, and decreased in higher trophic levels.

Selected specific activities are presented and compared with values from the literature.

ACKNOWLEDGEMENTS

We would like to express our appreciation to the following people for their assistance in the various aspects of this study: G. Johnson and R. Steele for their help in the field and laboratory; D. Wade for her help in laboratory processing and data compilation; R. Olsen for statistical assistance; A Case and G. Powers for sample analyses, and J. Corley for providing data on water chemistry.
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PART I: ATMOSPHERIC SCIENCES
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ANNUAL REPORT FOR 1971 TO THE
PACIFIC NORTHWEST LABORATORY

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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1971
to the
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VOLUME II: PHYSICAL SCIENCES
Part 1. Atmospheric Sciences

By

C. L. Simpson, Manager
and Staff of
Atmospheric Sciences Program

December 1972

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Volume I of this report to the USAEC Division of Biology and Medicine, covering work in the Biological and Ecological Sciences, is issued as BNWL-1650, Parts 1 and 2, respectively. Volume II of the report covers work in the physical sciences and is issued in this present part, Atmospheric Sciences, and in Part 2, Radiological Sciences, BNWL-1651.
FOREWORD

The results of the studies reported in this 1971 Annual Report are essential to evaluations for siting nuclear facilities, to assessment of exposures from routine low level releases during operations, and to describing hazards resulting from accidental release of significant quantities of radioactive materials to the atmosphere. Such studies are also important in evaluating consequences of releasing other pollutants routinely or accidentally to the atmosphere in connection with conventional power generation, fuel storage, shipment, and processing. In all such cases, an understanding of the processes in the atmosphere which transport, diffuse and deposit pollutants in order to estimate the dose that a given distribution of population will receive and the amount of material left on the surface which would enter into ecological processes of concern or be resuspended into the atmosphere at some later time where it could once more become a direct exposure hazard, is necessary.

The distance and time scale over which the processes of contaminant transport, diffusion, deposition and resuspension must be considered is very important. The nature and relative importance of these processes can change when considering the impact of pollutant releases over various distance and time scales. For example, models for atmospheric dispersion currently in general use are primarily empirical and based on data collected over short distances from near surface releases. These have been extrapolated to distances where larger scale motions are significant to the diffusion and extrapolated to heights where entirely different turbulence structure controls the diffusion than that present in the experiments from which models were developed. In addition, the experimental data on which most models have been based contain the effects of deposition in varying degrees, but the models fail to separate those effects from the turbulent diffusion in producing a given concentration and diffusion is generally assumed to be solely responsible for the resultant concentration.

The ongoing dispersion research and associated turbulence and deposition studies are aimed at independently accounting for the turbulent diffusion and the turbulent transfer and deposition of pollutants on the surface which jointly causes the reduction in plume concentration. The diffusion is being related to theoretical considerations of its dependence on turbulence structure and to models of turbulence under development from experimental programs, rather than depending on strict empiricism. Evaluations of deposition in field experiments together with laboratory and theoretical studies are leading to models of deposition describing dependence on turbulence, particle characteristics, and surface conditions. Ultimately, the model will be applicable for
areas having various surface roughness characteristics (from mountainous terrain to sea surface), for conditions of various atmospheric stability, for various heights of release, and for a variety of pollutant types (including particulates which deposit in varying degrees depending on particle size, reactive gases which deposit readily, and noble gases which should not deposit appreciably at all).

The dispersion of short term releases such as puffs is also under investigation. Present models do not adequately account for wind shear which elongates and dilutes the cloud to varying extents, depending on the stability, the distance from the source, and the depth of the layer through which the cloud has diffused. The dispersion of instantaneous and continuous releases in near calm conditions is an area where little information is available. Relationships of the dispersion to residual air motions has been difficult to establish until recent developments in sensitive anemometry have allowed detection of these motions.

The study of the instantaneous variations in continuous plume characteristics is another important research area being pursued. Until the development and application of the continuously monitoring Kr-85 tracer system, information on plume behavior dealt almost exclusively with average characteristics. Now short term variations peak-to-mean ratios are being described which have already found application to problems where instantaneous aspects must be known, such as washout of gases in diffusing plumes where nonlinear reactions preclude the use of an average plume description.

Precipitation scavenging research is also progressing vigorously. In the past, much emphasis was placed on the below-cloud "washout" of contaminants. This year some in-cloud "rainout" experiments are reported. These results represent the first successful in-cloud scavenging experiments performed in frontal storms; the few other successful in-cloud experiments performed under other AEC contracts and in other countries, were performed in convective storms. The results of these experiments are immediately and directly applicable to evaluating the fate of any pollutant dispersed within precipitating storms.

Many other important scavenging results are described in this report: continued monitoring and explorations of natural and anthropogenic radioactivity has revealed several new phenomena; significant progress has been made toward characterizing background and radioactive aerosols; new levels of understanding have been reached in the study of the washout of both aerosols and gases; new instrumentation and techniques have been developed. However, rather than elaborating on these here, the reader is referred to the specific reports that follow.
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INTRODUCTION

Precipitation scavenging is probably the dominant method by which the atmosphere rids itself of particulate pollution. For reviews, see Junge,\(^1\) and the proceedings of two recent conferences: Atmospheric Scavenging of Radioisotopes (1968)\(^2\) and Precipitation Scavenging (1970).\(^3\) Research in precipitation scavenging is directed toward measuring and understanding the rates at which precipitation of various types removes the many different forms of atmospheric contaminants, and although considerable progress toward meeting these objectives has been made for processes which occur beneath clouds, there still remains much experimental and theoretical work to be done on the in-cloud problem. As far as we know, there are only three reports of successful in-cloud scavenging experiments: Dingle, et al.\(^4\) Shopauskas, et al.\(^5\) and Burtsev, et al.\(^6\) Other experiments are in progress (Dingle, Semonin, et al., and Summers).\(^7,8,9\) However, all the above experiments were performed in convective storms. The purpose of this note is to report on in-cloud scavenging experiments recently performed in frontal storms.
The primary objective of these experiments was to determine if they were feasible. They are conceptually simple (though logistically complicated) but we were unsure of the appropriate time/distance scales. Some confidence was gained from the successful experiments of Dingle, et al., but it was clear that not much reliance could be placed on a juxtaposition of their time scale, since the processes in frontal storms are significantly different from those in convective storms. We were especially concerned about targeting the tracer material and as a result chose a sampling grid which spanned approximately an order of magnitude larger than the 8 km used by Dingle, et al. A second major objective was to compare the scavenging efficiencies for aerosol tracers of different physical and chemical properties.

The experiments were performed on the Olympic Peninsula of Washington. Figure 1. RHI and PPI radar coverage of the area was provided by the 758 U.S. Air Force Radar Squadron at Nahua Bay, on the northwestern tip of the peninsula. Hourly soundings were provided by National Weather Service personnel at Quillayute. Tracer aerosols were released from an B-23 aircraft which has been equipped for cloud physics research by members of the Cloud Physics Group of the University of Washington. Figure 2 shows the locations along various roads of the approximately 100 samplers, each of which was a polyethylene bag supported by a 17 cm diameter carton. The samples were frozen about two hours after the tracer releases were made. Chemical analysis was by neutron activation; samples and standards were irradiated with a flux of approximately $8 \times 10^{12}$ neutrons cm$^{-2}$ sec$^{-1}$, using the facilities of Washington State University.

Neg 720024-9

FIGURE 1. Schematic of the Olympic Peninsula Field Site Showing the Location of the Radar Site, Radiosonde Station and General Area of Tracer Releases. Distances shown are in statute miles.

3/11/71 Experiment

The first experiment was performed on 11 March 1971. It was decided to release the tracers at a low elevation (within the clouds) in the hope of improving the chances to detect the tracers in the samplers. 450 gms of indium were released by burning 10
FIGURE 2. Location of Samplers on Various Roads Throughout the Olympic Peninsula
pyrotechnic flares from 1157 to 1204 PST. In addition, approximately 1 kgm of lithium stearate was dispensed mechanically from 1208 to 1217 PST. Since the indium aerosol (mean particle size based on number distribution probably in the range 0.01 to 0.1 μm) is expected to be in the form of wettable $\text{I}_2\text{O}_3$, and since the lithium stearate (~10 μm) particles are non-wettable, it was hoped that a significant difference in scavenging rates would be observed.

A satellite photo, Figure 3, taken approximately at the time of the tracer release (~1200 PST) shows almost a textbook example of a frontal storm, with a well developed cloud shield. The front passed the Olympic Peninsula approximately 4 hours after the
tracer release. The RHI radar echo displayed a typical banded structure (cf. later figures) with echo tops at approximately 10,000 ft.

On the left hand side of Figure 4 are shown the temperature profiles as measured by radiosondes released from Quillayute at 852 and 1135 PST. The stable layer from approximately 900 to 750 mb was also observed after the time of the tracer release and transport. On the right hand side of Figure 4 is plotted the humidity measured by the 1135 sonde. The high wind speeds and large directional shear of the wind should be noted.

In Table 1 is given a description of the cloud particles which were replicated on Fomvar as the aircraft spiraled down to the release elevation of approximately 5,000 ft. Figures 5 and 6 summarize the information for the relative cloud particle concentrations during the times of the tracer releases. An evaluation of the collection efficiency of the instrument has not been included in this data analysis. Typical values for the average liquid water content were about 0.2 g m\(^{-3}\).

Neg 721419-12

FIGURE 4. Sounding Taken at 1135 PST 11 March 1971 at Quillayute, Washington
<table>
<thead>
<tr>
<th>Time</th>
<th>LAS</th>
<th>T1/2 (°C/°C/°C)</th>
<th>ALT</th>
<th>DROP</th>
<th>SIZE</th>
<th>DENS (g/cm³)</th>
<th>CONT (°C)</th>
<th>RATING</th>
</tr>
</thead>
<tbody>
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<td>1035</td>
<td>110</td>
<td>-11.1/-21.2</td>
<td>20,000</td>
<td>None</td>
<td>80-200</td>
<td>0.1 - 0.2</td>
<td>0.12 - 0.24</td>
<td>All Ice</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Thin Cloud</td>
<td></td>
<td>Mean 150</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1036</td>
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<td>-22.6/-26.2</td>
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<td>0.12 - 0.24</td>
<td>All Ice</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Thin Cloud</td>
<td></td>
<td>Mean 100</td>
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</tr>
<tr>
<td>1041</td>
<td>150</td>
<td>-19.8/-23.9</td>
<td>18,000</td>
<td>10-40</td>
<td>50-500</td>
<td>2 - 8</td>
<td>1 - 4</td>
<td>1 : 10</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mean 35</td>
<td></td>
<td>at first</td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td></td>
<td></td>
<td>Thicker Cloud</td>
<td></td>
<td>at first</td>
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<td></td>
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<td>Later</td>
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<tr>
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<td></td>
<td></td>
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<td>1043</td>
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<tr>
<td></td>
<td></td>
<td>Mean 40</td>
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<tr>
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<td>50-250</td>
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<td>0.25 - 3.3</td>
<td>10 : 1</td>
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<tr>
<td></td>
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<td>Mean 50</td>
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<tr>
<td>1050</td>
<td>130</td>
<td>-13.3/-17.5</td>
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<td>50-250</td>
<td>0.2 - 2</td>
<td>0.1 - 4.5</td>
<td>1 : 5</td>
</tr>
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<td>30-50</td>
<td>50-200</td>
<td>0.02 - 1.1</td>
<td>0.1 - 1.1</td>
<td>2 : 1</td>
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<td></td>
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<tr>
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<td>125</td>
<td>-10.7/-14.0</td>
<td>14,000</td>
<td></td>
<td></td>
<td>60-800</td>
<td>0.3 - 5</td>
<td>0.6 - 6</td>
</tr>
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</tr>
<tr>
<td>1114</td>
<td>155</td>
<td>-7.8/-12.1</td>
<td>15,000</td>
<td>30-45</td>
<td>60-800</td>
<td>2 - 5</td>
<td>2.3 - 6</td>
<td>1 : 25</td>
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<td>60-1000</td>
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<td>1.5 - 5.1</td>
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<td>11,000</td>
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<td>50-600</td>
<td>1 - 3</td>
<td>1.1 - 3.2</td>
<td>1 : 20</td>
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<td>1126</td>
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<td>0.6 - 3.6</td>
<td>2 : 1</td>
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<tr>
<td>1129</td>
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<td>-0.2/-4.2</td>
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<td>60-300</td>
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<td>15 : 1</td>
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<td></td>
<td>Mean 50</td>
<td></td>
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</table>

**TABLE 1**
<table>
<thead>
<tr>
<th>Time</th>
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<th>Temp.</th>
<th>Temp.</th>
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<th>Mgmt.</th>
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<tr>
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<td>175</td>
<td>-0.6</td>
<td>2.8</td>
<td>5,500</td>
<td>15-50</td>
<td>2 - 30</td>
<td>2.8 - 34</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
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<td>1155</td>
<td>175</td>
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<td>5,500</td>
<td>15-50</td>
<td>2 - 30</td>
<td>2.8 - 34</td>
<td>Irregular, Plateless &amp; Needles</td>
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<tr>
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<td>120</td>
<td>0.8</td>
<td>2.0</td>
<td>5,000</td>
<td>15-70</td>
<td>5 - 80</td>
<td>2 - 30</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
<tr>
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<td>175</td>
<td>-0.6</td>
<td>2.8</td>
<td>5,500</td>
<td>15-50</td>
<td>2 - 30</td>
<td>2.8 - 34</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
<tr>
<td></td>
<td>115</td>
<td>-0.8</td>
<td>1.0</td>
<td>3,000</td>
<td>14-70</td>
<td>1 - 20</td>
<td>1.5 - 12.0</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
<tr>
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<td>175</td>
<td>-0.6</td>
<td>2.0</td>
<td>5,000</td>
<td>15-70</td>
<td>5 - 80</td>
<td>2 - 30</td>
<td>Irregular, Plateless &amp; Needles</td>
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<tr>
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<td>0.8</td>
<td>2.0</td>
<td>5,000</td>
<td>15-70</td>
<td>5 - 80</td>
<td>2 - 30</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
<tr>
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<td>175</td>
<td>-0.6</td>
<td>2.0</td>
<td>5,000</td>
<td>15-70</td>
<td>5 - 80</td>
<td>2 - 30</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
<tr>
<td></td>
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<td>2.0</td>
<td>3,000</td>
<td>14-70</td>
<td>1 - 20</td>
<td>1.5 - 12.0</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
<tr>
<td>1150</td>
<td>175</td>
<td>-0.6</td>
<td>2.0</td>
<td>5,000</td>
<td>15-70</td>
<td>5 - 80</td>
<td>2 - 30</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
<tr>
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<td>0.8</td>
<td>2.0</td>
<td>5,000</td>
<td>15-70</td>
<td>5 - 80</td>
<td>2 - 30</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
<tr>
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<td>175</td>
<td>-0.6</td>
<td>2.0</td>
<td>5,000</td>
<td>15-70</td>
<td>5 - 80</td>
<td>2 - 30</td>
<td>Irregular, Plateless &amp; Needles</td>
</tr>
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*TABLE 1. (contd)*
### TABLE 1. (contd)

<table>
<thead>
<tr>
<th>Time</th>
<th>ALD</th>
<th>Anv.</th>
<th>ICE</th>
<th>Time</th>
<th>ALD</th>
<th>Anv.</th>
<th>ICE</th>
</tr>
</thead>
<tbody>
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<td>21</td>
<td>1</td>
<td>3.11</td>
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<td>5,100</td>
<td>20-80</td>
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<td>1.5</td>
<td>7.21</td>
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</table>

**Turbulence**

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<th>Anv.</th>
<th>ICE</th>
<th>Time</th>
<th>ALD</th>
<th>Anv.</th>
<th>ICE</th>
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<td>20-70</td>
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<td>5,000</td>
<td>20-60</td>
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<td>1.4</td>
<td>14</td>
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<td>-1.6</td>
<td>5,000</td>
<td>20-60</td>
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<td>1.4</td>
<td>14</td>
</tr>
<tr>
<td>1224</td>
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<td>5,000</td>
<td>20-60</td>
<td>1.1</td>
<td>1.4</td>
<td>14</td>
</tr>
</tbody>
</table>

**FIGURE 5. Relative Cloud Droplet and Ice Crystal Concentrations During the Burn of the Indium Flares.** To determine the concentrations, averages were taken over 15 second intervals, but an evaluation of the collection efficiency of the instrument has not been included.

**FIGURE 6. Relative Cloud Droplet and Ice Crystal Concentrations During the Release of Lithium Stearate**
Figure 7 shows the indium deposition pattern for the 3/11/71 experiment. The complete data for both experiments is given in Table 2. Background concentrations of indium are typically of the order of 1 ng l⁻¹, which is one to two orders of magnitude smaller than maximum recorded values. Because of the logistics of deploying and retrieving the samplers, useful values of the concentration of indium in the precipitation are not available. The total water collected in each sampler was of the order of 100 cm³. Thus typical indium concentrations in the samplers were about 0.1 ppb. Samples 15 through 38 and four reagent blanks were analyzed for lithium by atomic absorption spectroscopy. The concentrations were all below the detection limit of about 10 ppb.

4/16/71 Experiment

A second in-cloud scavenging experiment was performed on 16 April 1971, during which 450 g of indium and about 500 g of AgI were burned between 1320 and 1327 PST. Since it was found that the indium aerosol was inactive as an ice nucleus down to -22°C, it was hoped that the experiment would indicate differences in scavenging rates for aerosol particles that could have different interactions with the cloud particles. The release elevation was chosen to be about at the -15°C level, at which temperature AgI acts as an efficient ice nucleus.

The temperature and dewpoint from the 1310 sounding are shown in Figure 8. Winds were lighter than on 3/11 and the lapse rate near the release height was close to the wet adiabatic.

A portion of the PPI echo for 1245 PST is shown in Figure 9. A portion of the 170° RHI echo for 1329 PST is shown in Figure 10. The specific cell within the frontal storm, into which the tracers were released, is indicated by the vertical cross hair. The distorted echoes at 1350 and 1406 PST, shown in Figure 11 suggest that the AgI enhanced the growth of the cell. Figure 12 shows a convective cell growing out of the otherwise stratified cloud deck, as observed at about 1400 PST from the
<table>
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<tr>
<th>Sample Number</th>
<th>Weight (g)</th>
<th>10^-9 g Indium</th>
<th>Weight (g)</th>
<th>10^-9 g Indium</th>
<th>10^-9 g Silver</th>
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<td>4-16-71</td>
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<td>1</td>
<td>130</td>
<td>0.05</td>
<td>75.0</td>
<td>N.D.*</td>
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<td>2</td>
<td>82</td>
<td>&lt;0.001</td>
<td>64.0</td>
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<td>3 ± 3</td>
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<td>92</td>
<td>0.17</td>
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<td>0.52 ± 0.27</td>
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* N.D. = Not Detectable
** These samples highly contaminated with dirt.
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<th>Weight (g)</th>
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</table>

** These samples highly contaminated with dirt.
FIGURE 8. Sounding Taken at 1310 PST 4 April 1971 at Quillayute, Washington

FIGURE 9. A Portion of the PPI Probe Of the 4 April 1971 Storm, Taken at 2045 PST

FIGURE 10. A Portion of the 170° RHI Probe Taken at 1329 PST 4 April 1971. The cell into which the tracers were released is indicated by the vertical cross hair.
Although we do not yet consider this result to be an adequate evaluation of nucleation versus post-nucleation scavenging, nevertheless it is extremely encouraging.

**DISCUSSION**

Based on the observed indium deposition patterns the experiments were demonstrated to be feasible; however, they were only partially successful. There are a number of interesting and some surprising features of the results, most of which are being investigated during our 1972 experiments:

1. From the 3/11 deposition pattern it is apparent that the sampling grid is not of optimum scale. The amount of deposited indium appears to be still increasing with distance: on the arc at about 30 miles from the release point, approximately 1.8 mg of indium was deposited on a strip 1 meter wide, stretching across the Peninsula; on the arc at about 50 miles, approximately twice this amount fell. The 1972 sampling grid extends about twice as far as the 1971 grid.

2. The east-west spread of the deposition pattern is amazingly wide. It is highly suggestive that this cross wind spread is caused by the hydrometer trajectories through the direction wind shear.*

---

*See the report in this volume entitled "Raindrop Trajectories Through Wind Shear."
FIGURE 12. Abnormal Appearance of the Treated Cell Within the Stratus Deck, as Viewed from the Aircraft

3. It is not expected that significant dry deposition occurred, even for the 3/11 experiment, since otherwise the cross wind spread would not be so great. However, the possibility of dry deposition will be explored in future experiments by releasing SF₆ with the aerosol tracer and collecting grab samples at ground level.*

4. If a single-stage, in-cloud removal process is assumed, then the 3/11 results suggest a "rainout

* See the report in this volume entitled "An Inexpensive Infusion Sampler for Atmospheric Tracer Studies."

FIGURE 13. 16 April 1971 Indium Deposition Pattern
time constant" (for exponential decay of the airborne mass of indium) of about 1 hour. More satisfactory analytical methods are being developed.*

5. One of the most interesting aspects of the 4/16 experiment was the circumstantial evidence that the indium deposition was influenced by the simultaneous release of ice nuclei. Further such "targeting" experiments will be undertaken.

6. A preliminary study of the 4/16 results suggests that the rainout rate for aerosols which are active ice nuclei is significantly larger than the removal rate for inert material. This is being investigated during our 1972 experiments by utilizing a denser grid near the target area and by releasing more AgI, so that (hopefully) the concentration in the precipitation will be considerably above background.

ACKNOWLEDGEMENTS

The execution of these experiments depended on the cooperation of many individuals. The division of responsibilities among the authors was: D. G. Atkinson, collecting, reducing, and interpreting the cloud particle data; W. E. Davis, positioning the aircraft based on the radiosonde and radar data; L. F. Radke, in charge of aircraft operations; B. C. Scott, field coordinator and radar observer; W. G. N. Slinn, project director and writer; T. M. Tanner, sampling assistance and in charge of the radiochemical analyses; J. M. Thorp, meteorology and rapid radiosonde data reduction; and N. A. Wogman and J. A. Young, conceiving, deploying, and quality controlling the ground-based sampling.

We are especially grateful for the assistance of the 758 U.S. Air Force Radar Squadron, members of the Quillayute Weather Station, and students and staff at the University of Washington and Washington State University. Special thanks are due: M. T. Dana, A. N. Dingle, R. J. Engelmann, F. O. Gladfelder, A. M. Kahan, P. V. Hobbs, S. E. Shumway, J. W. Slocot, C. W. Thomas and F. M. Turner.

REFERENCES


* See the report in this volume entitled "Computer Results for an In-Cloud Release of Indium on 11 March 1971."
INTRODUCTION

Experimental results for the 11 March 1971 in-cloud scavenging experiments are given in a companion paper in this volume.* The purpose of this report is to apply the model recently developed by Davis¹ in an attempt to interpret the observed deposition pattern.

* See the preceding report.

MODEL

The in-cloud scavenging model was originally developed to explain the deposition of cosmogenic radionuclides. For nonradioactive tracers we take (see Davis):¹

1) \( \alpha \) (the radioactive decay coefficient) = 0,
2) \( B \) (the production term) = 0, and
3) \( t_0 \) (time in-cloud before precipitation) = 0.
With these three changes, the model was run for an \( n_o = 453 \) grams. As input for \( \psi \) (dilution coefficient, \( \text{sec}^{-1} \)) a liquid water content of 0.29 \( \text{g/m}^3 \) was assumed. Because of instrument failure, a measurement of liquid water content was not available. Further, a parcel precipitation rate was determined from both radiosonde and aircraft data. Using the mean rate of rise and assuming the liquid water content to be constant, a range of \( \psi \)'s was determined between \( 2 \times 10^{-4} \text{sec}^{-1} \) to \( \approx 1 \times 10^{-3} \text{sec}^{-1} \).

For each computer run, \( \psi \) and \( \lambda \) [the in-cloud scavenging coefficient, \( \text{(sec}^{-1}) \)] were assumed constant (Figure 1). These values were compared to the

**Figure 1.** Deposition Rate of Indium as a Function of Time After Release
observed pattern. As one can see, they seem to fit curves for \( \lambda > 10^{-3} \text{sec}^{-1} \)
and \( \phi > 4 \times 10^{-4} \text{sec}^{-1} \).

Another interesting point can be made by looking at the time of maximum deposition. From

\[
\frac{d^2n}{dt^2} = \frac{\psi \lambda n_0}{\phi - \lambda} \left[ \psi e^{\psi t} - \lambda e^{-\lambda t} \right] = 0,
\]

where \( n \) is the mass of indium deposited on the ground, then

\[
t_{max} = \frac{1}{\phi - \lambda} \ln \left( \frac{\lambda}{\psi} \right),
\]

where \( t_{max} \) is the time of maximum deposition. Figure 2 was then drawn.

The calculation of the deposition rate from the observed pattern indicated that at \( \approx 750 \) seconds the rate was still increasing. Figure 2 would indicate that a range of values for \( \lambda \) could be arrived at based on this observation. For a \( \psi \) range of \( 2 \times 10^{-4} \) to \( 10^{-3} \text{sec}^{-1} \) this would yield a range for \( \lambda \) of \( 10^{-3} \) to \( 10^{-2} \text{sec}^{-1} \). A value for \( \psi < 2 \times 10^{-4} \) would not explain the deposition pattern because, as can be seen in Figure 1, all values for deposition rates would be below those calculated from observed concentrations.

**SUMMARY**

A range of \( \lambda \) from \( 10^{-3} \) to \( 10^{-2} \text{sec}^{-1} \) was found for a range of \( \phi \) of \( 2 \times 10^{-4} \) to \( 10^{-3} \text{sec}^{-1} \). Also a comparison of times of maximum depositions for a range of \( \psi \) 's and \( \lambda \) 's was presented. Considerable care must be used in drawing conclusions from these results. The initial aim of the field experiment was to determine whether detectable quantities of indium would be seen at the ground from an in-cloud release. Data such as liquid water content, rainfall and snowfall rate through the plume were not observed. Thus what we are presenting at this time is an initial estimate which will be revised with further measurements. There are now planned experiments to determine changes in \( \psi \) by measuring the liquid water content associated with the plume throughout the period in which scavenging is taking place. With this information, the model will be rerun to determine the in-cloud scavenging coefficient for indium.
RAINDROP TRAJECTORIES THROUGH WIND SHEAR

W. G. N. Slinn and T. J. Bander

Trajectories and terminal locations were calculated for raindrops falling through the wind shear measured by the 1135 PST, 11 March 1971 radiosonde released from Quillayute on the Olympic Peninsula. Updraft velocities were parameterized. The results suggest that the spread of the deposition pattern for the indium tracer released into the storm could have been caused by the raindrops' trajectories through the wind shear.

The deposition pattern for the indium released into the 11 March 1971 coastal storm between 1157 and 1204 PST shows the indium spread over a distance of about 30 miles cross wind, after traveling only about 30 miles downwind. In an attempt to explain these observations, an analysis was made of the raindrop trajectories through the measured wind shear (see the 1135 PST 11 March 1971 sounding in the companion paper). Since the indium was released at the 0°C level, it would probably have been appropriate to include in the analysis that over a part of their trajectories, the hydrometeors were frozen. However, in this first look at the problem, this aspect was ignored.

The influence of two other parameters was explored. One was the possible influence of various magnitudes of updrafts which might have been induced by condensation on the wettable, indium aerosol particles; another was the effect of various

* "Precipitation Scavenging of Tracers Released into Frontal Storms."
"times of attachment" of the indium to precipitable hydrometeors. Both these parameters will be accounted for more completely in a developing "aerosol evolution code," which hopefully will lead to accurate predictions of the in-cloud scavenging of aerosol particles from frontal storms.

The origin of the coordinates was taken at ground elevation beneath the indium release point, which was at 1650 m elevation. The indium was then assumed to drift with the mean wind as given by the Quillayute sounding, and at various updrafts \( w = 0.05, 0.5, 1, 2, \) or \( 5.0 \) m sec\(^{-1}\). After times \( \tau = 100, 200, 500, 1000, \) or \( 2000 \) sec, the indium was assumed to be attached to precipitable cloud drops of radii \( R = 0.1, 0.2, 0.5, 1, \) and \( 2 \) mm.

The trajectory of each raindrop is dictated by

\[
\frac{d\mathbf{R}}{d\tau} = \mathbf{V} \tag{1}
\]

\[
\frac{d\mathbf{V}}{d\tau} = g(-\mathbf{k}) - \frac{\mathbf{D}}{\mathbf{M}} \tag{2}
\]

where \( \mathbf{R} \) is its position vector, \( \mathbf{V} \) its velocity, \( g \) is the acceleration of gravity and \( 1 \)

\[
\frac{\mathbf{D}}{\mathbf{M}} = \frac{3}{8} (1.1 \times 10^{-3}) \frac{\mathbf{V}^2}{R \mathbf{C}_o}
\]

\[
\left[ 1 + \frac{\xi_0}{(\text{Re})^{1/2}} \right]^2 \frac{\mathbf{V}}{\mathbf{V}} \tag{3}
\]

in which \( \xi_0 = 9.06, \mathbf{C}_o = 24/\xi_0, \)

\( \text{Re} = 2\mathbf{R}v/\nu, \nu = 0.153 \times 10^{-4} \text{ m}^2 \text{ sec}^{-1}, \)

and \( \mathbf{V} = \mathbf{V} - \mathbf{U}, \nu = |\mathbf{V}|, \) where \( \mathbf{U} \) is the mean wind as given by the sounding.

In the calculations, a numerical Simpson's integration routine was used which was accurate to the nearest meter. The wind speed and direction at any height were obtained with a quadratic interpolation between the data given by the sounding. The trajectories were found by solving the six differential equations using a fourth-order Runge-Kutta routine.

Typical results are shown in Figure 1. Along each thick solid line

\[\begin{figure}
\centering
\begin{tikzpicture}
\node at (0,0) {NEG 721170-1};
\node at (0,0) {FIGURE 1: The Terminal Location of Drops Falling Through the Wind Shear as Measured by the 1135 PST, 11 March 1971 Quillayute Sounding. The lines \( w = 5 \) cm/sec, 50, 100, 200, show the lines along which different raindrops (radii, 2, 1, 0.5 and 0.2 mm) would fall if they intercepted the indium \( 10^3 \) seconds after its release into the updrafts given by the \( w \) values. The dotted lines show the effect of different attachment times (\( \tau = 10^2 \) and \( 2 \times 10^3 \) sec) for specific updraft velocities.}
\end{tikzpicture}
\end{figure}\]
is the terminal position of those raindrops which picked up the indium 1000 seconds after its release. On each line is indicated where a 2 mm, 1 mm, 0.5 mm, and 0.2 mm drop would land. The different locations of the lines reflect the effects of different updraft velocities. The dotted line which is farthest south on the map, shows where the raindrops would have landed if \( w = 5 \text{ cm sec}^{-1} \) and \( \tau = 10^2 \) sec. The most easterly dotted line is for \( w = 100 \text{ cm sec}^{-1} \), and \( \tau = 2 \times 10^3 \) sec.

It is interesting that for specific values of \( w \) and \( \tau \), the terminal locations of raindrops of radii larger than about 0.2 mm lie on essentially straight lines. To see why this is so, consider the components of Equations (1) and (2). For drops larger than about 0.2 mm and less than 2 mm, it is adequate to use the empirical fit to the terminal velocity: \( w_t = 8000 R \text{ (sec}^{-1}) \). Thus we use \( D/M = g \tau/(8000 R) \equiv c_v \). The \( x \) (i.e., northerly) component of (2) then becomes

\[
\frac{du}{dt} + c_u = cU. \tag{4}
\]

Ignoring the initial \( x \)-component of the drop's velocity, (4) integrates to

\[
u(z) = \frac{c}{w_t} \int_z^H U(z') \exp\left\{-c(z' - z)/w_t\right\} \, dz',
\]

where \( H \) is the initial height of the drop. If \( U(z') \) is expanded in a Taylor series about its value at \( z \),

\[
U(z') = U(z) + \frac{3U}{3z} (z' - z) + \ldots \tag{6}
\]

and (5) is integrated term by term, it is seen that for the 11 March 1971 sounding, the first order correction is

\[
\frac{3U}{3z} \frac{w_t^2}{c} = \left\{ \frac{15 \text{ m sec}^{-1}}{500 \text{ m}} \right\} \left( \frac{8R}{10^6 \text{ m}} \right)^2 \tag{7}
\]

where \( R \) is the radius of the raindrop in mm. Even for a 2 mm drop, for which the "relaxation" length is approximately 25 meters, the correction, equation (7), is less than 1 m sec\(^{-1}\) which is only 3\% of the first order term. Thus

\[
u(z) = \frac{c}{w_t} (H - z) U(z). \tag{8}
\]

Substituting this into \( dx/dt = u(z) \) and similarly the \( y \) (westerly) components of equations (1) and (2) leads to a ratio of final \( x \) to final \( y \) locations which is independent of \( R, x, \) and \( y \) and therefore raindrops of different radii fall along a straight line.

In summary, it appears that the width of the indium deposition pattern from the 11 March 1971 experiment could have been caused by the spread of the raindrop trajectories falling through the shear flow. If this was the cause and if the Quillayute sounding gives an accurate picture of the winds above the entire Peninsula, then apparently some updraft velocity was either present or induced, since otherwise it is difficult to understand how the material was deposited so far to the east of the release.
point. It is also apparent from Figure 1 that the time constant for
attachment of the indium to precipitable hydrometeors must have been of
the order of $10^3$ seconds. Further experiments are in progress; in partic-
ular, measurements will be made in an
attempt to ascertain the influence of
mesoscale diffusion on the deposition
patterns.

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INITIATION OF THE ICE PHASE IN A SEEDED CLOUD*

A. J. Alkezweeny

A recently described contact nucleation model is improved to
account for variations with height of the updraft speed, liquid
water content, and droplet size distribution. The model is now
capable of calculating the concentration and size distribution
of ice crystals produced in a cloud seeded with AgI particles
below the cloud base.

INTRODUCTION

Recently Alkezweeny(1) presented
a simple model to describe the con-
tact nucleation of cloud droplets,
caused by seeding at the base of a
cumulus cloud. Previously, applica-
tions of the model were restricted
to cases with constant updraft speed,
constant liquid water content, and a
monodisperse cloud droplet size dis-
btribution. In the present report
these restrictions are removed by
postulating arbitrary updrafts and
liquid water contents and by discret-
izing the Khrgian-Mazin(2) cloud dro-
plet size distribution. In the future
it is hoped that this input data will
be supplied by coupling the model with
a dynamical model of a cumulus cloud.

INPUT DATA

Since a description of the model is
available elsewhere(1) it will not be

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Inc., and in part by the USAEC
Contract AT(45-1)-1830, with
Pacific Northwest Laboratory.
reviewed here. Special features of the present calculations include the height of the cloud base (640 mb), vertical grid step (100 m), maximum cloud life (3000 sec) cloud top height (5 km), initial concentration of AgI particles \((1.2 \times 10^{-11} \text{ m}^{-3})\), particle diffusion coefficient \((1.35 \times 10^{-4} \text{ cm}^{2} \text{sec}^{-1})\) and total number density of cloud droplets \((500 \text{ cm}^{-3})\).

The assumed vertical profiles of liquid water content and updraft speed are shown in Tables 1, 2 and 3. Values at intermediate \(z\) levels were found by interpolation.

---

**TABLE 1.** Liquid Water Content Profiles Used in the Calculation

<table>
<thead>
<tr>
<th>Height Above Cloud Base (M)</th>
<th>(W_{c1})</th>
<th>(W_{c2})</th>
<th>(W_{c3})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>1600</td>
<td>0.3</td>
<td>1.0</td>
<td>3.2</td>
</tr>
<tr>
<td>2600</td>
<td>0.5</td>
<td>1.5</td>
<td>5.6</td>
</tr>
<tr>
<td>3600</td>
<td>0.75</td>
<td>1.8</td>
<td>5.0</td>
</tr>
<tr>
<td>4600</td>
<td>1.0</td>
<td>2.0</td>
<td>4.0</td>
</tr>
<tr>
<td>5200</td>
<td>1.2</td>
<td>2.1</td>
<td>4.2</td>
</tr>
<tr>
<td>5600</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>7000</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

**TABLE 2.** Updraft Speed Profiles Used in the Calculation

<table>
<thead>
<tr>
<th>Height Above Cloud Base (M)</th>
<th>(u_{1})</th>
<th>(u_{2})</th>
<th>(u_{3})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1.5</td>
<td>2.0</td>
<td>3.0</td>
</tr>
<tr>
<td>1600</td>
<td>2.5</td>
<td>3.5</td>
<td>5.0</td>
</tr>
<tr>
<td>3600</td>
<td>5.0</td>
<td>7.0</td>
<td>10.0</td>
</tr>
<tr>
<td>4600</td>
<td>7.0</td>
<td>10.0</td>
<td>14.0</td>
</tr>
<tr>
<td>5200</td>
<td>8.5</td>
<td>12.0</td>
<td>17.0</td>
</tr>
<tr>
<td>5700</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>6000</td>
<td>-2.0</td>
<td>-3.0</td>
<td>-4.0</td>
</tr>
</tbody>
</table>

**TABLE 3.** Pairs of Liquid Water Content, \(W_c\), and Updraft Speed, \(u\), Profiles for Each Case Studied.

<table>
<thead>
<tr>
<th>Case</th>
<th>(W_{c1})</th>
<th>(u_{1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>(W_{c1})</td>
<td>(u_{1})</td>
</tr>
<tr>
<td>2</td>
<td>(W_{c2})</td>
<td>(u_{1})</td>
</tr>
<tr>
<td>3</td>
<td>(W_{c3})</td>
<td>(u_{1})</td>
</tr>
<tr>
<td>4</td>
<td>(W_{c1})</td>
<td>(u_{2})</td>
</tr>
<tr>
<td>5</td>
<td>(W_{c1})</td>
<td>(u_{3})</td>
</tr>
</tbody>
</table>

**RESULTS**

The computed results for the five cases are numerous and will not be presented here. Detailed results of the computations for Case 2 can be found elsewhere. In this report only a comparison between the five cases will be described.

The heights above cloud base, at which droplets in each class first made contact with AgI particles are shown for each case in Table 4. It can be seen from the table that increasing the liquid water content for the same updraft speed causes larger droplets to contact AgI particles at a lower height. However, no change in the heights was found with changing updraft speed. The total concentrations of droplets which contacted AgI particles at each level changed only by less than a factor of three from one case to the other.

In order to use the model output, a certain assumption concerning the activation temperature of AgI particles should be made. For example, it was assumed that only droplets
TABLE 4. The Height (M) Above Cloud Base at Which Droplets in Each Class First Made Contact with AgI Particles for Each of the Five Cases Studied

<table>
<thead>
<tr>
<th>Droplet Size (μm)</th>
<th>0-20</th>
<th>20-40</th>
<th>40-60</th>
<th>60-80</th>
<th>80-100</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>50</td>
<td>150</td>
<td>450</td>
<td>1450</td>
<td>2750</td>
</tr>
<tr>
<td>Case 2</td>
<td>50</td>
<td>150</td>
<td>450</td>
<td>1450</td>
<td>2750</td>
</tr>
<tr>
<td>Case 3</td>
<td>50</td>
<td>150</td>
<td>450</td>
<td>1450</td>
<td>2750</td>
</tr>
<tr>
<td>Case 4</td>
<td>50</td>
<td>150</td>
<td>450</td>
<td>1450</td>
<td>2750</td>
</tr>
<tr>
<td>Case 5</td>
<td>50</td>
<td>150</td>
<td>450</td>
<td>1450</td>
<td>2750</td>
</tr>
</tbody>
</table>

It seems that the heat released changes linearly with height and increases with an increase in the liquid water content. Changing the updraft speed has little effect on it. This result may be of value for input into existing dynamical models of cumulus clouds.

CONCLUSIONS

With assumptions on the activation temperature of AgI particles, the model is capable of calculating the initial concentration and size distribution of ice crystals in a seeded cloud as a function of height. Before it can be compared with observations, the model should be included in a

![Figure 1: Heat Released as a Function of Height Above Cloud Base](Neg 715708-1)
growth model where the final sizes and concentration of ice crystals can be obtained for each level.

REFERENCES


AN ANALYSIS OF DIFFUSION IN WIND SHEAR

W. G. N. Slinn**

A theoretical framework is presented to describe turbulent diffusion in an atmosphere with a nonzero gradient of the mean wind. Moments of the convective diffusion equation are taken and if these are terminated at the second moment, then the contaminant concentration is approximated to be Gaussian with specified covariance matrix. A special case is evaluated to demonstrate the resulting diffusion ellipsoid and the t, z, and z0 regimes of 2.

Although the importance of wind shear to the diffusion of atmospheric contaminants is well established,(1) nevertheless the theoretical evaluation of diffusion in wind shear has apparently made essentially no progress.(2)* We use the term "wind shear" to describe both the directional change and variation in magnitude of the mean wind. In particular, we shall consider the linear variation (with height) of the wind given by

\[ \mathbf{V} = \left( U + \frac{U_0}{x} z \right) i + \frac{V_0}{y} z j + w_0 k. \] (1)

A constant updraft velocity, \( w_0 \), is included in Equation (1), for application of the theory to the diffusion within a cloud of material upon which condensation might occur.

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** Work supported in part by Battelle Memorial Institute's Physical Sciences Program.
† Note added in proof: F. A. Gifford has called the author's attention to two significant analyses of diffusion in wind shear, listed as references (5) and (6).
The present analysis is based on a reinterpretation of the convective diffusion equation

\[
\frac{\partial \chi}{\partial t} = -\nabla \cdot \vec{F}
\]  

(2)

where \( \vec{F} \) is the flux of the contaminant. Here, rather than treating \( \chi \) as the contaminant concentration, we interpret \( \chi \) to be a probability density function. For example, let \( \chi(\vec{r}, t; \vec{r}_0, 0) \, dt \) be the probability that a single contaminant aerosol particle (or gas molecule) will be in the volume element \( dt \) about \( \vec{r} \) at time \( t \), if it were known to be at \( \vec{r}_0 \) at \( t = 0 \). Alternatively if \( \chi \) is multiplied by \( N \), the total number of contaminant particles released, then \( N \chi \) is interpreted to be the probability that \( N \chi \) contaminant particles are within \( dt \) at time \( t \).

Proceeding from this interpretation of (2), we seek to obtain various moments of \( \chi \), such as the mean position

\[
\langle \vec{r} \rangle = \int \vec{r} \chi \, dt
\]  

(3)

the mean square, etc., from which we can derive the covariance,

\[
\delta^2 = \langle \vec{r}^2 \rangle - \langle \vec{r} \rangle^2,
\]  

(4)

skewness, kurtosis, etc., of the distribution. In (3), the integration is over all space.

To obtain expressions for these moments we multiply (2) through by various powers of \( \vec{r} \) and integrate over all space. For example, multiplying through by \( \vec{r}^* = 1 \) leads to

\[
\frac{\partial}{\partial t} \int \chi \, dt = -\int \nabla \cdot \vec{F} \, dt,
\]  

(5)

Utilizing Green's theorem on the rhs of (5) and assuming that the flux is zero at infinity gives

\[
\frac{\partial}{\partial t} \int \chi \, dt = -\int \vec{F} \cdot d\vec{r} = 0.
\]  

(6)

Thus the probability that a contaminant particle is somewhere in the whole space (or the total number of particles in the whole space) is constant. Choosing the first interpretation, we take this constant to be 1, i.e., the particle is certain to be somewhere.

The mean position of the particle is given by

\[
\frac{\partial}{\partial t} \int \vec{r} \chi \, dt = -\int \nabla \cdot \vec{F} \, dt.
\]  

(7)

Utilizing Green's theorem and assuming that \( \vec{F} \) vanishes at infinity leads to

\[
\frac{\partial}{\partial t} \langle \vec{r} \rangle = \int \vec{F} \, dt.
\]  

(8)

Similarly, the mean square is given by

\[
\frac{\partial}{\partial t} \langle \vec{r}^2 \rangle = \int (\vec{r} \vec{F} + \vec{F} \vec{r}) \, dt
\]  

(9)

where \( \vec{r} \vec{F} \), etc., are second order tensors.

To proceed, a specification of the flux is needed. As is customary in the "X-theory" of turbulent diffusion, we take

\[
\vec{F} = -\vec{\kappa} \cdot \nabla \chi + \nabla \chi
\]  

(10)
where \( \hat{K} \) is the (symmetric, second order tensor) turbulent diffusivity and \( \hat{V} \) is the mean wind (plus sedimentation velocity, which here we take to be zero).

As an example of the application of this theory, consider the case when the mean wind is given by (1). Then from (8) and (10) the mean position of the particle is given by

\[
\frac{3}{\overline{t}_t} \langle \mathbf{r} \rangle = \int \left[ \mathbf{\nabla} \cdot \hat{K} \cdot \mathbf{V} \chi + \mathbf{V} \cdot \hat{K} \right] \, dt. \tag{11}
\]

Since

\[
\mathbf{\nabla} \cdot \hat{K} = \mathbf{K} \cdot \mathbf{\nabla} \chi + \chi \mathbf{\nabla} \cdot \hat{K} \tag{12}
\]

then upon a further application of Green's theorem, (11) becomes

\[
\frac{3}{\overline{t}_t} \langle \mathbf{r} \rangle = \int \left[ \mathbf{V} + \mathbf{V} \cdot \hat{K} \right] \chi \, dt. \tag{13}
\]

If \( \hat{K} \) were a constant, then using \( \hat{V} \) given by (1), we obtain

\[
\langle \mathbf{r} \rangle = \left[ \mathbf{U} + \frac{u_0}{l_x} \frac{\omega_{0z}^2}{2} \right] \langle \mathbf{t} \rangle + \left[ \frac{v_0}{l_x} \frac{\omega_{0z}^2}{2} \right] \langle \mathbf{j} \rangle + \left[ \omega_{0z} \right] \langle \mathbf{k} \rangle \tag{14}
\]

which expresses the rather obvious result that the mean position of an aerosol particle (or of the cloud of aerosol particles) is dictated by the mean wind in a straightforward manner.

It is interesting that if \( \hat{K} \) is dependent on height, for example, if

\[
\hat{K} = k_0 + \hat{k} \frac{z}{l_z}
\]

then

\[
\mathbf{V} \cdot \hat{K} = \frac{z}{l_z} \hat{k}
\]

and (14) is changed by replacing the updraft velocity by \( w_0 + (z/l_z) \). Thus a vertical gradient in the diffusivity causes an effective increase in the updraft. Usually this is small and can be ignored. However if \( \hat{K} \) were to vary as much as an order of magnitude (mks units) in a distance of 10 meters, as might occur in the lowest regions of the atmospheric boundary layer, then this effective updraft velocity could be

\[
0 \left( 10 \, \text{m}^2 \, \text{sec}^{-1} / 10 \, \text{m} \right) = 1 \, \text{m} \, \text{sec}^{-1} \tag{17}
\]

which is significant.

It is a little more tedious to determine the six components of the (symmetric) second order tensor \( \langle \mathbf{r} \mathbf{r} \rangle \). Using the mean wind as given by (1) and assuming that \( \hat{K} \) is diagonal, then from (9) we obtain, for example,

\[
\frac{3}{\overline{t}_t} \langle x^2 \rangle = 2K_{xx} + 2 \langle x \rangle U + 2 \langle xz \rangle \frac{u_0}{l_x} \tag{18}
\]

\[
\frac{3}{\overline{t}_t} \langle xz \rangle = \langle x \rangle w_o + \langle z \rangle U + \langle z^2 \rangle \frac{u_0}{l_x} \tag{19}
\]

\[
\frac{3}{\overline{t}_t} \langle z^2 \rangle = 2K_{zz} + 2 \langle z \rangle w_o. \tag{20}
\]

Substituting the results for \( \langle x \rangle, \langle y \rangle, \langle z \rangle \) as given by (14) into the above, solving them in the order (20), (19) and (18), subtracting appropriate
terms of \( \langle \vec{r} \rangle^2 \), and evaluating the other three terms of \( \langle \vec{r}^2 \rangle \) in a similar manner, gives, for the terms in the covariance matrix \( \nu = \frac{\nu^2}{\nu} = \langle \vec{r}^2 \rangle - \langle \vec{r} \rangle^2 \), the result

\[
\begin{bmatrix}
2K_{xx} + \frac{2}{3} K_{zz} \left( \frac{u_{x}^{2} - 1}{x} \right)^{2} & \frac{2}{3} K_{zz} \left( \frac{u_{y}^{2} - 1}{y} \right)^{2} & K_{zz} \left( \frac{u_{z}^{2} - 1}{z} \right) \\
\sigma_{xy}^{2} & 2K_{yy} + \frac{2}{3} K_{zz} \left( \frac{v_{y}^{2} - 1}{y} \right)^{2} & K_{zz} \left( \frac{v_{z}^{2} - 1}{z} \right) \\
\sigma_{xz}^{2} & \sigma_{yz}^{2} & 2K_{zz}
\end{bmatrix}
\]  

(21)

It is interesting that the updraft velocity, \( w_{o} \), does not appear in the covariance matrix. It becomes increasingly tedious to proceed analytically. However there appears to be no theoretical obstacle to obtaining higher order moments of the distribution. If we stop at the second moment then we have that the distribution is Gaussian with mean given by (14) and covariance, by (21). Thus, with \( |\nu| \) the determinant of the covariance matrix \( \nu \), and \( \nu^{-1} \) its inverse, we have that

\[
\chi(\vec{r},t) = \left[ (2\pi)^{3} |\nu| \right]^{-1/2} \exp \left( -\frac{1}{2} \nu^{-1} \nu^{T} \right)
\]  

(22)

and for \( u_{o} = 0 \), then the variances along the principal axes are given by

\[
\sigma_{xx}^{2} = \lambda_{p1} = 2Kt
\]

(23)

\[
\lambda_{p2}, \lambda_{p3} = 2Kt \left\{ 1 + \frac{1}{6} \left( \frac{v_{y}^{2} - 1}{y} \right)^{2} + \frac{1}{2} \frac{v_{z}^{2} - 1}{z} \right\}^{1/2}
\]

(24)

where \( \vec{x} = \vec{r} - \langle \vec{r} \rangle \) and \( \vec{x} \) and \( \vec{x}^{T} \) are appropriate row and column matrices.

Specific applications of these results should probably proceed via a numerical evaluation of \( \nu, \nu^{-1} \) and \( |\nu| \), since it is not easy to derive the characteristic values of \( \nu \) in the general case. For the specific case \( \nu = KI \), where \( I \) is the identity tensor, the principal axes are located at angles \( \Theta_{p} \) from the \( y \) axis, given by

\[
\tan 2\Theta_{p} = \frac{5}{(v_{x}^{2}/y)}
\]

(25)

At small times (i.e., \( t \ll 1_{y}/V_{o} \)) these reduce to

\[
\lambda_{p1} = 2Kt
\]

(26)
\[ \lambda_{P_2} = 2Kt \left( 1 + \frac{1}{2} \frac{v_{o}^{t}}{v_{y}} \right) \]  
(27)

\[ \lambda = 2Kt \left( 1 - \frac{1}{3} \frac{v_{o}^{t}}{v_{y}} \right) \]  
(28)

\[ \theta_{P_2, P_3} = \frac{\pi}{4}, \frac{3\pi}{4} \]  
(29)

At large times they become

\[ \sigma_{xx}^{2} = \lambda_{P_1} = 2Kt \]  
(30)

\[ \sigma_{yy}^{2} = \lambda_{P_2} = 2Kt \left[ 1 + \frac{1}{3} \left( \frac{v_{o}^{t}}{v_{y}} \right)^{2} \right] \]  
(31)

\[ \sigma_{zz}^{2} = \lambda_{P_3} = 2Kt \]  
(32)

\[ \theta_{P_2, P_3} = 0, \pi/2 \]  
(33)

Notice that at large time, the principal axes of the diffusion ellipsoid are again \( x, y, z \) and that diffusion in the crosswind direction is enhanced.

The major feature of the present result is the demonstration that for turbulent diffusion, as for Brownian diffusion in a nonuniform gas, a gradient in the mean velocity of the fluid influences the diffusion. Instead of spherical, the diffusion about the mean position is ellipsoidal, with principal axes orientation dictated by the gradient of the wind. An essential difference of the present result from that in Brownian diffusion is the appearance of the time in the correction terms. Of course we are intrigued to inquire if this time dependence [cf., e.g., the rhs of (27) or (31)] might explain the observed \( t^2 \) and \( t^3 \) dependence of \( \sigma_{y}^2 \) observed in the upper troposphere and stratosphere. (4)

In conclusion we should point out that the restriction of the analysis, to mean winds which vary at most linearly with the coordinates [cf., (1)], appears to be an essential restriction, since otherwise closure of the moment equations would not be obtained.

I wish to thank S. A. Slinn for her valiant attempt to diagonalize (21) in the general case.

REFERENCES


NATURAL PRECIPITATION WASHOUT OF SULFUR DIOXIDE

M. T. Dana, J. M. Hales and M. A. Wolf

A field investigation and a modeling effort are being conducted as components of an ongoing program for the study of sulfur-compound washout from power-plant plumes. The theoretical analysis of SO₂ washout developed in this work was shown to agree fairly well with field measurements of washout of SO₂ from controlled releases from towers.

Research described in this report was undertaken to elucidate several aspects of SO₂ washout identified in a previous study. This research consisted of three parts, including a field investigation of SO₂ washout, a laboratory study of SO₂ solubility in water, and a program to develop a means of predicting SO₂ washout from plumes on the basis of emission data and atmospheric characteristics.

The field investigation was conducted at the Quillayute site in western Washington. It consisted of releasing controlled amounts of SO₂ from two towers during rain periods and measuring the resulting concentrations in ground-level rain as a function of spatial location beneath the plumes. Release heights were variable, ranging from 7.6 m to 30.5 m. Ten separate dual tower experiments were conducted during this study.

The laboratory phase was conducted to establish experimentally the relationship between SO₂ solubility, concentration, and solution pH. It was found that the equation

\[
\text{SO}_2 = \frac{[\text{SO}_2]_g}{H} \cdot \frac{[\text{H}_3\text{O}^+]_e}{\sqrt{[\text{H}_3\text{O}^+]_e}} + 4K_1 [\text{SO}_2]_g/H
\]

provides good agreement with experiment for the concentration range studied (0.05 part per million and above), provided the results of Johnstone and

* This work was supported by the Environmental Protection Agency via a related services agreement with the U.S. Atomic Energy Commission.
Leppa(2) are used for the solubility parameters H and k. In Equation (1) the bracketed terms denote concentrations in moles/liter, c_{SO_2} is the total dissolved SO_2 concentration, and H_3O^+ denotes the concentration of acid donated by sources other than dissolved SO_2.

The modeling program has resulted in two models of SO_2 washout. The first of these is based on the assumption of linear transport and equilibrium properties, and was developed in conjunction with the tritium washout study described earlier.(3) The second model is more general and can account for nonlinear behavior. It also can correct for sloping trajectories of the rain as it encounters the plume. This model, however, requires computer solution; this contrasts with the linear model, which is amenable to hand calculation.

Comparison of the nonlinear model with results for one of the field experiments is shown in Figure 1. The

**FIGURE 1.** Comparison of Measured SO_2 Washout Concentrations with Those Predicted on the Basis of the Nonlinear Model and a Time Averaged Plume. Downwind Distance = 200 feet.
dashed and solid lines pertain, respectively, to model predictions based on gas-phase limited and stagnant drop behavior [c.f. (1)]. The vertical lines show measured SO$_2$ concentrations and the circular dots denote concentrations in rain that would be in equilibrium with the calculated ground-level concentration of SO$_2$ in the plume.

Calculations leading to Figure 1 were based upon average dispersion parameters for the experiment period. Since such averages are not totally appropriate for computing nonlinear washout from fluctuating plumes (1), an attempt was made to define the system in a more "instantaneous" fashion by computing washout for incremental time periods during the experiment. These incremental predictions were averaged to obtain the estimates of washout for the total experiment period, which are shown in Figure 2. Here it is observed that washout predictions under these circumstances do not deviate markedly from those predicted on the basis of time-averaged conditions. Predicted ground-level equilibrium concentrations, however, do exhibit large deviations. This is somewhat perplexing since the close proximity of

![Graph](image)

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**FIGURE 2.** Comparison of Measured SO$_2$ Washout Concentrations with Those Predicted Using the Nonlinear Model for Incremental Time Periods. Downwind Distance = 200 feet.
equilibrium predictions to actual measured washout shown in Figure 2 suggests that the observations can be explained in two ways -- either by a simple equilibrium situation or by the more complex behavior suggested by the general washout models. Although the data obtained from the field experiments suggest that the latter explanation is correct, further measurements are needed to confirm this completely.

The washout models formulated in this investigation will be used in the near future to examine washout of SO₂ from a commercial power plant plume.

REFERENCES


PRECIPITATION WASHOUT OF GASEOUS TRITIUM COMPOUNDS

J. M. Hales

A theoretical analysis of the washout of tritium compounds from plumes has been completed. Based upon reversible gas-washout theory, this analysis provides a means for calculating washout concentrations from plumes of various geometries, including the bivariate-normal distribution. At present no field measurements exist to check the validity of this analysis for tritium compounds; data for SO₂ washout, however, indicate that agreement between theory and measurement should be acceptable under most circumstances.

The investigation of precipitation washout of gaseous tritium compounds (HT, T₂, HTO, and T₂O) described in the 1970 annual report (1) has been completed. The equations presented in that report have been modified and solved for hypothetical example situations. This work has been completed in conjunction with a related analysis of SO₂ washout conducted for the Environmental Protection Agency. (2) The equations formulated during this study
are based upon reversible washout theory and provide expressions for the concentration of scavenged material in rain at ground level. If the material being scavenged occurs in the atmosphere as a normally distributed plume (with reflection at the ground) then the basic equations are

\[
 c_A_o (a) = \frac{v}{2 \sigma_y \sigma_z} \exp \left( - \frac{\sigma_y^2}{2} - \frac{\sigma_z^2}{2} \right) \left[ \exp(ch) \text{erfc} \left( \frac{h - \sigma_z^2}{\sqrt{2} \sigma_z} \right) + \exp(-ch) \text{erfc} \left( \frac{h - \sigma_z^2}{\sqrt{2} \sigma_z} \right) \right] ; \quad (1)
\]

\[
 z = \frac{5K_y H^*}{v_t a} ; \quad (2)
\]

and

\[
 z = \frac{3E m}{4\mu H^* \sigma_w} + \frac{5K_y}{v_t a} ; \quad (3)
\]

Nomenclature for equations (1 through 3) is given as follows:

\[
\begin{align*}
a & = \text{raindrop radius, } \text{m}.

\varepsilon & = \text{fraction of plume removed by washout upwind from point of calculation.}

h & = \text{source height, } \text{m}.

H^* & = \text{partition coefficient for pollutant, } \text{m}^2/\text{mole}.

\sigma_w & = \text{density of water, } \text{m}/\text{m}^3.

\sigma_y & = \text{plume spread parameters, } \text{m}.

\sigma_z & = \text{plume spread parameters, } \text{m}.

\end{align*}
\]

\[
\begin{align*}
 v_t & = \text{fall velocity of raindrop, } \text{m}/\text{sec}.

 y & = \text{crosswind distance, } \text{m}.

 K_y & = \text{overall mass-transfer coefficient, moles}/\text{m}^2\text{sec}.

 m & = \text{liquid-water content of cloud, } \text{m}/\text{m}^3.

 \bar{u} & = \text{mean wind velocity, } \text{m}/\text{sec}.
\end{align*}
\]

As discussed previously, these equations are based upon the assumptions of linear transport and equilibrium behavior, negligible redistribution of the plume in the atmosphere by action of the rain, vertical rainfall, and negligible change of cloud droplet size en route to the ground.

Equation (1) has been solved for the compounds HT and HTO for the following conditions:

- Temperature: 20°C
- Wind Speed: 5 m/sec
- Plume spread parameters: Pasquill type D stability (4)
- Effective release heights: 30 m, 100 m
- Downwind distances: 100, 200, 400, 800, and 1600 m.

A portion of the computed results is given in Figures 1 and 2, which show the spread of estimated washout...
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FIGURE 1. Calculated Average Concentrations of HT in Rain at Ground Level, 1600 m Downwind from Source

concentrations for a typical rainfall. These pertain to HT and HTO in the cross-plume direction for release heights of 50 m and 100 m and a downwind distance of 1600 m. A more thorough documentation of results is available elsewhere. (4)

These results predict behavior that differs from that predicted by traditional washout theory in several respects. The most important of these differences are listed as follows:

1. Source height may have a significant effect on washout rates.

2. Dispersion affects washout significantly.

3. Tritiated water vapor is washed out much more effectively than tritium gas; this occurs because of the greater "solubility" of the former in rainwater.

4. Washout rates can increase with distance from the source under some circumstances.

At present no experimental data exist to indicate how closely these equations agree with actual behavior
of tritiated compounds. Field data from the related study of SO₂ washout, however, suggest that agreement within a factor of two may be attained in a majority of cases.

REFERENCES


WASHOUT OF RHODAMINE DYE PARTICLES

M. Terry Dana

An ultrasonic nozzle aerosol dispenser was used in conducting nine additional washout experiments in the rhodamine dye series. Particle mass median radii from 0.08 to 0.61 μ were dispersed by atomization of methanol-rhodamine solution droplets of initial radii 2.5 μ. An analysis of several physical processes which might have affected the experiments in the series demonstrates that the results of many of them could have represented washout of solution droplets larger than the desired dry particles.

This report introduces results of a series of washout experiments in which submicron-sized rhodamine dye particles were dispersed by an ultrasonic nozzle. Completion of the nozzle calibration study(1) has led to a revision of the likely particle sizes generated by the nozzle. In addition, results of previously reported experiments(2) along with the current results, are analyzed in terms of the effect upon them of washout of not fully evaporated methanol-dye solution droplets.
ULTRASONIC NOZZLE CALIBRATION

The original calibration of the ultrasonic nozzle relied upon measurements of particle size from electron microscope grids exposed in an electrostatic precipitator. The reported relationship was derived without results of the concurrent sizing made by cascade impactor. The latter sizing effort has been subsequently analyzed. Calculations of effective cutoff radii for the various stages of the six-stage Anderson impactor were done using the theory of Ranz and Wong (5) for the operating conditions of the runs. The average mass median radii of the particles for runs made with various tracer solution concentrations are listed in Table 1, along with similar results from electrostatic precipitator samples.

The satisfactory agreement between the mass distributions computed two ways for C = 1 samples, and the consistency of the values A computed from the cascade impactor runs led to a choice of A = 2.5 \( \mu \) for the effective initial droplet diameter of the ultrasonic nozzle.

RECENT EXPERIMENTAL RESULTS

A series of washout experiments using the ultrasonic nozzle was conducted at Quillayute Airport in Washington. Table 2 lists experimental details for the nine successful experiments of the series. Of two others, one was aborted because of an unacceptable change in weather, and one resulted in too-low tracer concentrations in the rain because of insufficient mass dispersed.

<table>
<thead>
<tr>
<th>Tracer Solution Concentration C (g( \text{cm}^{-3} ))</th>
<th>Number of Samples</th>
<th>Average Mass Median Radius ( a ) (( \mu ))</th>
<th>Average Initial Droplet Radius ( A ) (( \mu ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>5</td>
<td>0.11 (e) (e)</td>
<td>2.7</td>
</tr>
<tr>
<td>1.0</td>
<td>7</td>
<td>0.21 0.21 0.11 (e)</td>
<td>2.4</td>
</tr>
<tr>
<td>4.0</td>
<td>2</td>
<td>0.35 (e) (e)</td>
<td>2.5</td>
</tr>
</tbody>
</table>

- a. Cascade impactor samples.
- b. Electrostatic precipitator/electron photomicrograph samples.
- c. Computed by the Hatch formula (1, 4) from the number median radius of (b) above.
- d. Computed from
  \[ A = a^{\left(\frac{2}{3}\right)} \]
  where \( \rho_p \) = particle density (= 1.18 g cm\(^{-3}\)).
- e. No data available.
TABLE 2. Experimental Details: Ultrasonic Nozzle Experiments

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Particle Radius ( a ) (( \mu ))</th>
<th>Mass of Tracer Released ( Q ) (g)</th>
<th>Rainfall Intensity ( J ) (mm hr(^{-1}))</th>
<th>( a/J ) (mm(^{-1}))</th>
<th>( a/J ) (hr(^{-1}))</th>
<th>( A ) (hr(^{-1}))</th>
<th>( B ) (hr(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>UR-3</td>
<td>0.10</td>
<td>0.06</td>
<td>2.1</td>
<td>1.25 (a)</td>
<td>0.22</td>
<td>0.25</td>
<td></td>
</tr>
<tr>
<td>UR-4</td>
<td>0.10</td>
<td>0.05</td>
<td>6.3</td>
<td>1.25 (a)</td>
<td>0.20</td>
<td>(b)</td>
<td></td>
</tr>
<tr>
<td>UR-5</td>
<td>0.21</td>
<td>0.53</td>
<td>1.8</td>
<td>1.51</td>
<td>0.10</td>
<td>(b)</td>
<td></td>
</tr>
<tr>
<td>UR-6</td>
<td>0.21</td>
<td>0.95</td>
<td>2.2</td>
<td>1.38</td>
<td>0.50</td>
<td>(b)</td>
<td></td>
</tr>
<tr>
<td>UR-7</td>
<td>0.21</td>
<td>0.43</td>
<td>3.6</td>
<td>2.2 (a)</td>
<td>0.10</td>
<td>0.25</td>
<td></td>
</tr>
<tr>
<td>UR-8</td>
<td>0.10</td>
<td>0.28</td>
<td>0.81</td>
<td>1.65</td>
<td>0.12</td>
<td>1.3</td>
<td></td>
</tr>
<tr>
<td>UR-9</td>
<td>0.21</td>
<td>1.48</td>
<td>0.44</td>
<td>1.60</td>
<td>0.28</td>
<td>1.3</td>
<td></td>
</tr>
<tr>
<td>UR-10</td>
<td>0.08</td>
<td>0.14</td>
<td>1.8</td>
<td>1.20</td>
<td>0.24</td>
<td>0.64</td>
<td></td>
</tr>
<tr>
<td>UR-11</td>
<td>0.10</td>
<td>0.23</td>
<td>2.1</td>
<td>1.50</td>
<td>0.38</td>
<td>2.0</td>
<td></td>
</tr>
</tbody>
</table>

\( a \). Estimated from other Quillayute experience.

\( b \). No data available.

The experimental "apparent" washout coefficients for Arc A (radius 22.9 m) and Arc B (45.7 m) are indicated as \( A \) and \( B \), respectively. The quantity \( a \) is defined by

\[
\lambda = aE,
\]

where \( E \) is a mean collection efficiency for the particle spectrum and a given raindrop spectrum. \( a \) -- the washout coefficient if \( E \) is unity -- is a description of the rainfall:

\[
a = \frac{2}{3} \pi F D_2^2,
\]

where \( F \) is the total rain flux (mm\(^2\) hr\(^{-1}\)) and \( D_2 \) is the mean areal raindrop diameter (mm); \( a \) is generally proportional to rainfall rate in continuous prefrontal rain. Thus the usually observed rate dependency of \( \lambda \) is attributable to \( a \), and for raindrop sizes in the range of interest here, the collection efficiency -- for inertial washout -- is practically independent of raindrop size. The dependency is chiefly on particle size and density.\(^{(5)}\)

\[\text{ANALYSIS.}\]

Experimental collection efficiencies for the current runs and previous experiments (pneumatic atomization),\(^{(2)}\) are listed in Table 3, and are plotted in Figure 1, as a function of expected particle radius \( a \). The solid lines represent the inertial washout theory as compiled by Slinn,\(^{(5)}\) for particle densities appropriate to solid rhodamine (1.38 g cm\(^{-3}\)) and methanol (0.8 g cm\(^{-3}\)). Three potentially influential processes were considered in an attempt to explain the divergence between experiment and theory.
1. Dry Deposition

The "raw" collection efficiencies $E_A$ and $E_B$ describe the total deposition, and may be expected to contain a component due to dry deposition. The deposition velocity, $V_D$, is defined by:

$$V_D = \frac{F_D}{x},$$

where $x$ is the air concentration of tracer (g cm$^{-3}$) and $F_D$ is the dry component of the tracer flux (g cm$^{-2}$ sec$^{-1}$). Washout measurements provide a flux $F_T$ (total wet and dry) to the precipitation collector.

If $x$ is suitably measured or calculated, a maximum dry deposition velocity can be calculated:

$$V_M = \frac{F_T}{x}.$$

$V_M$ was evaluated for each arc of each run; these are listed in Table 3. The air concentration at the plume centerline, ground level, was calculated* from the bivariate normal plume with reflection:

$$x = \frac{Q}{T} \cdot \frac{1}{\pi\alpha y z} \exp \left[ -\frac{1}{2} \left( \frac{h}{\alpha z} \right)^2 \right].$$

where:

- $Q$ = mass of tracer released in time $T$;

* Air concentrations were measured with filters on several of the runs. The results were scattered and the filter air flow rates were not precisely known. Agreement with calculations was within a factor of two or three, however. Measurements at similar distances from the source by Dana, et al. (6) showed similar agreement with $SO_x$ as the tracer.
\( \sigma_y, \sigma_z \) = plume distribution parameters (determined from the deposited mass distributions, and through \( \sigma_y = \frac{1}{2} \sigma_z \); \( \sigma_z = \sigma_y \), where \( \sigma \) is the range from the source).

Values of \( V_M \) comparable to a reasonable choice of deposition velocity (e.g., 1 cm sec\(^{-1}\)) would indicate the likelihood of a considerable dry component in the observed deposition.

The values of \( V_M \) in Table 3 are generally greater (especially for Arc A) than 1 cm sec\(^{-1}\); i.e., they are such that a considerable dry deposition velocity is required for the dry component to constitute a significant portion of the total deposition. Consequently, dry deposition appears to be a minor contributor to Arc A results and does not completely explain the divergence from the theory.
2. Undercutting of the Plume by Precipitation

Figure 2 schematizes the process by which the rain which falls onto Arc A may be significantly that which has not encountered the tracer plume. The cutoff diameter, the diameter of a raindrop which just passes the tower top enroute to Arc A, was calculated for each run. From Figure 2, it is evident that

\[ V_{tco} = \frac{h\bar{u}}{x_A} \]  \hspace{1cm} (6)

where \( V_{tco} \) is the terminal velocity of the cutoff raindrop, \( h \) the tower height, \( \bar{u} \) the mean wind speed, and \( x_A \) the distance from the source to Arc A. For raindrop sizes of interest here, \( V_t \approx 4000D \), so

\[ D_{co} = \frac{h\bar{u}}{4000 x_A} \]  \hspace{1cm} (7)

All raindrops smaller than \( D_{co} \) were eliminated from the rain spectra, and new values of the "washout-effective" rain flux and mean areal diameter \( (D'_2) \) were compiled. By (2), this leads to new washout factors \( a' \), and thus new collection efficiencies \( E'_A \). The latter are shown in Table 3. The increase in washout is relatively small—even when 90% of the raindrops are eliminated by the cutoff—because while \( D_2 \) increases, \( F \) decreases.

The trajectory of the new mean raindrop diameter defines an interception time

\[ t_A = \frac{x_A}{\bar{u}} - \frac{h}{4000 D'_2} \]  \hspace{1cm} (8)

The cutoff effect is slight for Arc B, and was neglected; thus the interception time for Arc B is:

\[ t_B = \frac{x_B}{\bar{u}} - \frac{h}{4000 D'_2} \]  \hspace{1cm} (9)

![Figure 2: Mean Raindrop Trajectories and Plume Interception Times](image-url)
This cutoff effect, a combination of rain spectra and wind characteristics, serves to provide an increased value of the collection efficiency on Arc A, and an interception time for Arc A which is still quite short in most cases. Perhaps most significantly, the cutoff effect tends to bring the interception times for Arc A and B closer together. Thus, any aerosol characteristics which may be relatively slowly changing in time may be influential upon the washout results for both arcs.

3. Washout of Unevaporated Methanol Droplets

Concern arose from the above since in the event that the methanol droplets (initially ~20 μ radius for pneumatic atomization, 2.5 μ for ultrasonic atomization) do not evaporate quickly, they could be washed out, and undesirably increase the arc deposition results. Assuming that the washout for Arc A doses conform to the inertial theory for methanol (ρ_d = 0.8), the appropriate effective radii for washout were found from Figure 1, and plotted vs. t_A in Figure 3. Since dry deposition should not be dismissed, the values E_A were adjusted for an arbitrary deposition velocity of V_D = 1 cm sec⁻¹. This adjustment can be made by using a variation of (3):

$$E'_A (V_D) = E_A (1 - V_D/V_M) \quad (10)$$

Figure 3 demonstrates the plausibility of the assumption of methanol droplet washout, and indicates that the evaporation rate is sufficiently slow in some cases to affect the washout on Arc B as well. The points marked "H" are for runs in which the methanol release rate was significantly higher than the others; these points suggest a tendency for slower evaporation as the release rate is increased.

It is clear that the atomized methanol droplets immediately begin to evaporate upon introduction into the methanol-vapor-free atmosphere, decreasing in size sharply. But at some point the rate of evaporation slows due to the increase of the methanol vapor pressure to which the droplets are exposed.
Using the apparent evaporation rates indicated by Figure 3, values of $a_B$ were found that correspond to $t_B$. Applying (10) to $B$:

$$E_B(V_D) = E_B(1 - V_D/V_M), \quad (11)$$

with $V_D = 1 \text{ cm sec}^{-1}$ -- led to the values of $E_B(V_D = 1)$ plotted in Figure 4 versus $a_B$. For some of the runs (those for which $V_M \leq 1$) washout was eliminated completely, the suggestion being that dry deposition could have accounted for all of the observed deposition.

There remain several points on Figure 4 which diverge from the theory. It is interesting to note that four of these (33, 34, 39, 8) were runs where $t_B$ was relatively large: the particles

---

**FIGURE 4.** Arc B Experimental (with Dry Deposition Correction) and Theoretical Collection Efficiencies for Rhodamine Dye Particles
were exposed to the atmosphere for a longer time than in most other experiments. The indication of high washout rate may be due to another size-increase effect, namely that of condensation growth of the soluble rhodamine particles. The ability of rhodamine particles to grow in this manner in a near-saturated water vapor environment is expected to be evaluated soon.

THEORETICAL CONSIDERATIONS

While it seems plausible that the observed washout is compatible with the above analysis, additional theoretical developments should not be dismissed. For small particles (\(\leq 0.1 \mu\)) Brownian diffusion comes into play as a washout mechanism, leading to higher values of the collection efficiency than those due to inertial effects alone. Thermophoresis may be an important enhancer of washout in the range of particle size up to about 1 \(\mu\), if the temperature of the raindrop is significantly cooler than that of its surroundings.\(^{(7)}\) Electrical effects may also be significant for this size range. Enhanced collection efficiencies have been observed\(^{(8)}\) for electrical conditions which may have been extant at the time of the experiments reported here.\(^{(9)}\) Finally, in the particle size range \(\geq 10 \mu\), the inertial theory can allow collection efficiencies in excess of unity.\(^{(10, 11)}\) The approximation to the inertial theory used in Figures 1 and 4 predicts collection efficiencies approaching unity asymptotically.

REFERENCES


AEROSOL PARTICLE SIZE DISTRIBUTIONS IN THE NORTHWEST*
A. J. Alkezweeny and W. G. N. Slinn

Surface level, aerosol particle size distributions were measured at several sites in Washington State: near Richland, at Seattle, and on the Olympic Peninsula. The Richland data are representative of an unpolluted continental atmosphere and for particle diameter, $D$, in the range 0.2 to 2 microns, $dN/dD \sim D^{-a}$, with $a \approx 5$. The data obtained on the Olympic Peninsula have characteristics expected of a maritime aerosol. Preliminary results are also given of the effect of wind on resuspension of aerosol particles from a desert terrain.

INTRODUCTION

Knowing the characteristics of background atmospheric aerosols and gases is important for the evaluation of the effect of contaminant releases. To see this, consider some of the possible atmospheric interactions for a particular release. Its physical and chemical properties may be modified within the atmosphere via interactions with trace gases; it may attach to background aerosol particles and its subsequent dry deposition may depend on the size of the aerosol particle to which it attached; it may enter into cloud forming processes and either its behavior in the cloud will be dictated by the

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cloud droplet and ice crystal nucleation capabilities of the background aerosol particle to which it is attached and/or it will be in competition with the background concentration of these nuclei; if it is not removed by precipitation scavenging, then its properties upon leaving the cloud environment will probably be significantly different from its properties when it entered, as a result of enhanced in-cloud coagulation with other background aerosol particles and gases, within the cloud droplets. This scenario could obviously be continued but perhaps the above sufficiently illustrates the essential roles played by background aerosols and gases.

Toward determining more information on background aerosols, and to circumvent past difficulties encountered with developing an airborne research platform, we have obtained the size distribution of background aerosol particles measured near ground elevation. Little is known about continental and maritime aerosols in an unpolluted atmosphere. Here, we report on several size distributions, over the particle size range 0.015 to 5 μm radius, measured at three sites in Washington State: near Richland, at Seattle, and on the Olympic Peninsula. Some preliminary studies of the influence of wind on resuspension from a desert terrain are also reported.

The three instruments used to characterize the aerosol particles were a General Electric Aitken nuclei counter, a Royco Optical Counter (model PC 200 A), and a Whitby Aerosol Analyzer. Temperature, relative humidity and surface level (2 m) winds were either measured on site or obtained from nearby weather stations.

**SIZE DISTRIBUTION MEASUREMENTS**

The Richland surface measurements (probe height of about 3 meters) were made on the AEC reservation 50 km north of the city, and about 300 km east of the Pacific Ocean, on October 7, and 8, 1971 (See Figure 1).
During this period the wind speed was less than 7 mph and the relative humidity was less than 50%. Visibility was excellent; the Aitken nuclei counter recorded concentrations in the range 2 to $3 \times 10^3$ particles/cm$^3$. A total of four size distributions was obtained, each being an average over several distributions.

The resulting distributions are plotted in Figure 2. It can be seen from the figure that the Royco Optical Counter and the Whitby Aerosol Analyzer agree quite well in the size range where they overlap. The concentrations of particles of a specific size measured at different times, differ at most by a factor of two.

The data for the size distribution for particles between 0.2 to 2 microns in diameter, can be approximated by

$$\frac{dN}{dD} = C D^{-\alpha},$$

where $\frac{dN}{dD}$ is the number of particles per cm$^3$ per micron interval; $C$ is constant; $D$ is the particle diameter; and $\alpha$ is about 5. This data can be considered as representative for a continental atmosphere with little pollution, as indicated by the good visibility and the low Aitken nuclei count. Therefore, such a value for $\alpha$ is reasonable.\(^{(1)}\)

In Seattle, the aerosol size distributions were obtained at two sites on October 19: one, at the Battelle Seattle Research Complex near the University of Washington, and the other from the observation deck of the Space Needle (Figure 3) 520 feet above ground level. Prior to the measurements it was raining but during the measurements the clouds were dissipating. The visibility was good; the Aitken Nuclei Counter indicated about $4 \times 10^4$ particles/cm$^3$. Figure 4 shows the size distribution measured at the two locations.

The third measurements were made at two locations on the Olympic Peninsula. The first site was about 50 feet from the ocean at LaPush, and the second site was located at the Quillayute

emergency landing field, about 10 km from the ocean. Because of instrument difficulties only the Royco Counter was used in the study.

At LaPush the measurements were made on October 20, 1971. During the morning considerable rain had fallen in the area. The data were collected just after the rain had stopped. The wind was calm (<5 mph, temperature 52°F), and the relative humidity decreased to 70% during the period of measurement. A slight increase in the particle concentration was observed during the measurement period. Averages of all the data points collected over a period of two hours are shown in Figure 5. The distribution shows a peak at about 0.7 micron diameter which is typical of sea-spray distribution.

In the morning of the next day, another series of measurements was made at Quillayute. The wind fluctuated between 5 and 15 mph, temperature was 50°F and the relative humidity was 64%. Average of all the distribution is shown in Figure 5. It can be
in cloud physics. Those which are active as a cloud condensation nuclei may produce large cloud droplets, enhance droplet growth by coalescence, and hence increase area precipitation. On the other hand, because of their large surface area, they are most likely to act as ice nuclei. Ice nucleation activity of large particles has been demonstrated experimentally by Rosinski et al.\(^3\) They have shown that the percentage of soil particles active as ice nuclei increases with increase in particle size and that the concentration of particles larger than 1.5 microns in diameter on the ground increased by several orders of magnitude during severe storms.

In Table 1, the concentrations of particles larger than 0.4 micron in diameter are shown for different surface (2 m) wind speeds. For wind speeds in the range of 2 to 7 mph, no significant change in the concentrations was detected.

<table>
<thead>
<tr>
<th>TABLE 1. The Effect of Wind Speed on Aerosol Particle Concentration (#/cm(^3))</th>
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</thead>
<tbody>
<tr>
<td>Particle Diameter Range, (\mu)</td>
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<tr>
<td>---------------------------------</td>
</tr>
<tr>
<td>0.4 - 0.5</td>
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<tr>
<td>0.5 - 0.64</td>
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<tr>
<td>0.64 - 0.8</td>
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<tr>
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<td>3.2 - 4.0</td>
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<tr>
<td>5.0 - 6.4</td>
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<tr>
<td>6.4 - 8</td>
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</tbody>
</table>

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FIGURE 5. Aerosol Particle Size Distributions at LaPush (on the coast) and at Quillayute (about 10 km inland) on the Olympic Peninsula.

seen from the figure that the concentrations of particles of all sizes decreased considerably, and no peak was found in comparison with the LaPush distribution. It is possible that dry deposition on foliage between the two locations is responsible for this effect; this will be studied further.

EFFECT OF THE WIND ON THE PARTICLE CONCENTRATION

Particles larger than one micron in diameter are of particular interest
However, for a wind speed of 25 mph there is a considerable increase in the concentration of particles larger than one micron. A decrease in the concentration of particles smaller than 0.5 micron was also observed. No explanation for this effect is available at this time; instrument inadequacies are not yet dismissed. Further experiments to determine the entrainment/resuspension of aerosol particles are in progress.

CONCLUSIONS

Even though the data are preliminary a few conclusions may be drawn:

1. The size distribution of background aerosol particles near Richland for particles between 0.2 to 2 microns in diameter can be approximated by $D^{-\alpha}$ where $D$ is the particle diameter and $\alpha$ is about 5.

2. Significant variations in particle size distributions were observed on different days near the coast, at sites separated by only a few tens of kilometers and under similar meteorological conditions.

3. Resuspension of aerosol particles by winds can be relatively easily studied by determining the changes in the size distribution.

ACKNOWLEDGEMENT

We wish to thank the managers of the Seattle Space Needle for their hospitality.

REFERENCES


ON THE DRY DEPOSITION OF SEA SALT NUCLEI*

W. G. N. Slinn**

An analysis is made of the dry deposition of sea salt nuclei in an attempt to estimate the time constant for the removal of sea salt by dry deposition, under the assumption that the rate limiting process is the turbulent diffusion of the nuclei to the earth's surface. A general analytical solution is found to the appropriate convective diffusion equation for power-law profiles of the diffusivity and mean wind. The resulting time constant for the exponential decrease of sea salt nuclei concentrations over continents is of the order of 10 hours.

INTRODUCTION

It is a gross understatement to say that an adequate evaluation of the influence of dry deposition on the budgets of atmospheric contaminants is yet to be determined. Not only is there not an adequate evaluation, there are uncertainties of orders of magnitudes. For example, there is the often quoted estimate that precipitation scavenging accounts for approximately 90% of the removal of bomb debris, while dry deposition, only the remaining 10% (Small),\(^1\) and yet there is conjecture (e.g., see Junge,\(^2\) pp. 307-311) that precipitation scavenging of sea salt nuclei is only 1/2 to 1/4 as important, over a long time period, as dry deposition.

The uncertainties appear to arise because of different vertical profiles of the contaminants, their different chemical and physical properties, and different dry deposition receptors and precipitation types.

It is not even clear what is the rate limiting stage of the dry deposition process. Presumably for a chemically inert substance such as \(^{85}\)Kr, the rate limiting stage would be the final biological/chemical/physical interaction at the collector-atmosphere interface. Apparently, for the case of dry deposition of aerosol particles on surfaces in wind tunnels, it is various microphysical processes (Brownian diffusion, photoretic effects, etc.) acting within the viscous sublayer, (Davies,\(^3\) Sehmel,\(^4\) as apparently it is also, for the dry deposition of gaseous radiiodine onto carbon plates in the atmosphere (Markee).\(^5\) On the other hand, if one accepts Chamberlain's\(^6\) theoretical prediction, then perhaps

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\(^**\) Work supported in part by Battelle Institute's Physical Sciences Program and in part by U.S. Atomic Energy Commission Contract AT(45-1)-1830.
turbulent transfer through the atmospheric boundary layer is the rate limiting mechanism for the deposition of $^{131}$I onto grass (Markee, 5; Pasquill). (7)

But for global contaminant budget considerations perhaps the dry deposition problem should be viewed from a different perspective. Perhaps the vegetation cover of a continent should be viewed as a huge fibre filter, of thickness which varies from a few centimeters (cut grass) to as much as a hundred meters (rain forest). What this filter loses because of its relative inefficiency, it makes up for in length. From this view it seems reasonable to inquire: what would be the dry deposition velocity and thereby, the appropriate time constant for removal of contaminants, if a continent's surface were treated as a perfect sink and the rate limiting process were the turbulent diffusion of the contaminant to the "filter" at the surface? Seeking an answer to this question for the case of sea salt nuclei is the subject of this brief note.

PROBLEM

The specific problem we consider is that of the profile and dry deposition of sea salt nuclei. We choose this example because appropriate experiments are in progress. Mathematically we approximate the problem by attempting to solve the two dimensional convective diffusion equation subject to specific boundary conditions, where $u = u(z)$ is the mean wind speed in the $x$ direction, $K = K(z)$ is the eddy diffusivity and $\chi$ is the concentration of the contaminant. Diffusion in the $x$ direction is ignored as is any gravitational settling of the contaminant. The boundary conditions imposed are: (1) at $x = 0$, for example at the sea coast, $x(x = 0, z) = \chi_0$; (2) $x(x > 0, z = 0) = 0$, that is, we assume that the vegetation is a perfect sink for the sea salt nuclei, and (3) $\chi(x \to \infty, z = h) = 0.$ Actually, it would be more realistic to introduce a mixing layer height, $h$, above which the concentration of nuclei is zero. To circumvent this added complication, the solution will be considered for inland distances $x$ for which $x(x, z = h)$ is still essentially $\chi_0$, where $h$ is 5 to 10 km. Power law profiles are used for $u$ and $K$:

$$u = u_0 \left( \frac{z}{h} \right)^{1/n}, \quad K = k_0 \left( \frac{z}{h} \right)^{1/m} \quad (2)$$

Equation (1) has been the subject of a number of investigations. However, the problem as stated above apparently has not been solved previously. Our method is to take a Laplace transform in $x$ (parameter $s$), which leads to

$$u_0 \left( \frac{z}{h} \right)^{1/n} \left[ \frac{\partial}{\partial z} \left( k_0 \left( \frac{z}{h} \right)^{1/m} \frac{\partial \chi}{\partial z} \right) \right]$$

$$= \frac{\partial}{\partial z} \left[ k_0 \left( \frac{z}{h} \right)^{1/m} \frac{\partial \chi}{\partial z} \right] \quad (3)$$
The solution to (3) which satisfies the boundary conditions at \( z = 0 \) and \( z = \infty \) is

\[
\hat{\chi}(s,z) = \frac{\chi_0}{s} \left[ 1 - \frac{2}{\Gamma(v)} \left( \frac{s}{z} \right)^v \mathcal{K}_v (\lambda z^u) \right]
\]

(4)

where \( \mathcal{K}_v \) is a modified Bessel function of order \( v \) and where

\[
v = \frac{m-1}{2}\mu m
\]

(5)

\[
z_u = 2 - \frac{1}{m} + \frac{1}{n}
\]

(6)

\[
\lambda = \frac{(\cos)^{1/2}}{v}
\]

(7)

\[
\rho = \frac{u_0}{k_0} \frac{1}{l/m}
\]

(8)

and \( \Gamma(v) \) is the usual gamma function.

Inverting (4) gives the desired solution

\[
\chi(x,z) = \chi_0 P(v,\xi).
\]

(9)

where

\[
\xi = \frac{\lambda z^2}{4u^2 x}
\]

(10)

and

\[
P(v,\xi) = \frac{1}{\Gamma(v)} \int_0^\xi e^{-t} t^{v-1} dt.
\]

(11)

in an incomplete gamma function.

The flux of the nuclei at any elevation is given by

\[
F = -X(z) \frac{\partial \chi}{\partial z},
\]

(12)

Evaluating this one obtains

\[
F(z) = -\frac{k_0}{1/m} \frac{\chi_0}{\Gamma(v)} 2 \mu \left[ \frac{0}{4u^2 x} \right]^v \exp\left\{ -\xi \right\}
\]

(13)

It is interesting both that the flux as given by (13) is essentially independent of \( z \) for small \( z \) (provided \( \chi \gg \rho z^2 u/(4u^2) \) ) and that for the assumed profile of \( \chi(z) \), even though \( \chi(z) \to 0 \) as \( z \to 0 \), yet the flux remains finite at the surface.

**DEPOSITION VELOCITIES FOR SPECIAL CASES**

I. \( m = n = \infty \)

If the mean wind and eddy diffusivity are assumed to be independent of height, i.e.,

\[
U = u_0 \left( \frac{z}{1_k} \right)^{1/n}, \quad K = k_0 \left( \frac{z}{1_k} \right)^{1/m}.
\]

(14)

with \( m, n \to \infty \), then the general solution, (9), reduces to

\[
\chi(x,z) = \chi_0 \eta \left( \frac{1}{2}, \xi \right) = \chi_0 \text{erf}\left( \frac{u_0 z^2}{4k_0 x} \right)^{1/2}.
\]

(15)

If we define the deposition velocity to be the magnitude of the surface flux, divided by the concentration at some reference height \( z_0 \), and if this height and \( x \) are chosen such that the argument of the error function is small compared with unity (e.g., \( u_0 = 10 \text{ m sec}^{-1}, \ k_0 = 10^6 \text{ cm}^2 \text{ sec}^{-1}, \ x > 1 \text{ km}, \text{then} \ z_0 < 100 \text{ m} \), then we obtain

\[
\nu_d = \frac{|F|}{\chi(x,z=z_0)} = \frac{k_0}{z_0}.
\]

(16)
II. \( m = 1, n \) arbitrary

If the diffusivity increases linearly with height \((m = 1)\), and the wind profile is an arbitrary power law, then writing the general solution \((9)\) in the form

\[
\chi(x,z) = \chi_0 \xi^\nu \gamma^*(\nu, \xi).
\]

(17)

where

\[
\gamma^*(\nu, \xi) = e^{-\xi} \sum_{n=0}^{\infty} \frac{\xi^n}{\Gamma(n+1)}.
\]

(18)

and taking \( \nu = \frac{m-1}{2m} = 0 \),

leads to the surprising result

\[
\chi(x,z) = \chi_0 h(z).
\]

(19)

where \( h(z) \) is the Heaviside function. Thus the concentration is uniform with height and drops sharply to zero at \( z = 0 \). It remains \( \chi_0 \) even though there is deposition because we have assumed \( \chi = \chi_0 \) at \( x = 0 \), for all \( z \).

Thus there is an infinite reservoir. From (13), the deposition velocity based on \( \chi_0 \) becomes

\[
v_d = \frac{m+1}{m} \frac{k_0}{1_k}
\]

(20)

III. General Case (except \( m \neq 1 \))

To estimate the deposition velocity in the case of arbitrary power law profiles for \( U \) and \( K \) (except \( m \neq 1 \)), we expand (9) for small \( \xi \) as in Case I. Then

\[
v_d = \frac{|F(x,z=0)|}{\chi(x,z=x_0)} = \frac{m+1}{m} \frac{k_0}{\xi_0} \left( \frac{z_0}{1_k} \right)^{1/m}
\]

(21)

DISCUSSION

In the above analysis an attempt has been made to determine what deposition velocities would result for sea salt nuclei, under the condition that dry deposition from the atmosphere is controlled (i.e., rate limited) by the turbulent diffusive transfer of the sea salt to the collecting surface. The result is that if this were true then \( v_d = 0(k_0/1_k) \).

Numerically, if we use values suggested by Pasquill(7) then \( v_d = 5 \times 10^3 \text{ cm}^2 \text{ sec}^{-1}/200 \text{ cm} = 25 \text{ cm sec}^{-1} \); or using values as given by Markee(8) for a neutral atmosphere, then \( v_d = 3 \text{ m}^2 \text{ sec}^{-1}/10 \text{ m} = 30 \text{ cm sec}^{-1} \); or if we assume that momentum and contaminants have diffusivities of similar magnitudes, then \( v_d = k u_*, \)

where \( k \) is von Kármán's constant and \( u_* \) is the friction velocity, which leads to similar numerical values.

Although these deposition velocities are typically an order of magnitude larger than the few values which have been obtained experimentally for material other than sea salt nuclei, nevertheless this author believes that it is not justifiable to conclude that turbulent diffusion is not the rate limiting stage for the dry deposition of sea salt nuclei. If the calculated results correctly describe nature then, for example, the concentration of sea salt nuclei in a column of height \( h \) and base area \( A \), moving with the mean wind speed, would decrease according to \( d(yhA)/dt = -yA \ v_d \ \text{ or } \chi = \chi_0 \exp(-t/t) \)

where \( t = h/v_d \). If \( h = 5 \text{ km} \) and \( v_d = 20 \text{ cm sec}^{-1} \), then \( t = 10 \text{ hours} \).
It is not yet clear whether or not this is an unrealistic time constant. But even if it is, the objective of this inquiry has been met: we have determined within the approximations utilized, what the deposition velocity would be if turbulent diffusion were the rate limiting stage of the dry deposition of sea salt nuclei.

REFERENCES


CLOUD CONDENSATION NUCLEI AND VISIBLE POLLUTION IN LOS ANGELES*

A. J. Alkezweeny and T. J. Lockhart**

Simultaneous measurements of cloud condensation nuclei and light scattering in Los Angeles down to a visibility of 7.5 miles are presented. It is shown that, in Los Angeles, air pollution is a good source of nuclei. Their concentration increases with decrease in visibility.

INTRODUCTION

A change in the concentration of cloud condensation nuclei can play an important role in the cloud formation and precipitation development. For example, Warner(1) reported a decrease in precipitation downwind of a sugar plantation during the past 60 years. He associated this decrease with an increase in sugar cane production. He also pointed out that this effect is caused by the increase in the concentration of cloud condensation nuclei. On the other hand, Hobbs, et al., (2) have reported an increase in precipitation downwind of pulp mills in Washington State. They have also shown that these mills are a good source of cloud condensation nuclei.

Although cloud formation and precipitation development is a complex process, it is generally accepted that cloud condensation nuclei could decrease or increase precipitation depending upon the sizes and concentrations of the nuclei. An increase in the concentration may produce more small cloud droplets with a narrow size distribution which would impede drop growth by coalescence, coalescence being an essential mechanism for warm rain production. However, the production of giant nuclei leads to the formation of large cloud droplets and thus a wider size distribution which could enhance coalescence (Fletcher). (3)

Several observations on the effect of air pollution, in the Los Angeles area, on the concentration of cloud condensation nuclei have been published (Alkezweeny and Green, (4) Hidy, et al., (5) and Alkezweeny and Smith). (6) However, all these measurements were either preliminary or lacking quantitative measurements of pollution levels. In this paper simultaneous measurements of cloud condensation nuclei concentrations and

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** Meteorology Research, Inc., Altadena, California.
light scattering coefficients are presented and discussed.

METHOD OF OBSERVATION

The data were collected in Altadena, California, during the months of January, February and April, 1971. Two instruments were used in this study. A cloud condensation nuclei (CCN) counter and an integrating nephelometer. The CCN counter is similar in design to that developed at the University of Washington (Radke and Hobbs). (7) In this instrument the air is drawn into a diffusion chamber where the supersaturation is controlled. The active nuclei at the fixed supersaturation, grow into uniform size droplets. Their number is measured by an optical scattering technique and recorded on a chart recorder. In the integrating nephelometer (Charlson, et al.,) (8) the aerosols are drawn into the instrument, the light scattering from all the particles is measured and recorded.

The two instruments were located inside the building of Meteorology Research, Inc., and sampled air from the same port connected to a 2 inch diameter tube bringing air from outside the building. They were operated continuously. No corrections were made on the data as to the losses of particles in the tube and the effect of relative humidity. However, in view of the size of the tube, the losses are expected to be very small; probably less than the losses in the sampling tube of the instruments. Furthermore, the relative humidity of the sampled air will probably be decreased considerably before reaching the instruments because of the temperature increase of the sampled air, within the tube inside the building.

RESULTS AND DISCUSSION

In Figure 1 the concentration of cloud condensation nuclei active at 0.5 percent supersaturation are plotted as a function of time. Nuclei concentrations fluctuate from about 100/cc to about 7000/cc. The fluctuation is effected by the degree of pollution in the atmosphere, change in air masses, and the presence of precipitation. For example, on the 17th of April, a concentration in excess of 1000/cc was recorded. However, after a rain shower, the count dropped by an order-of-magnitude, indicating a removal of the nuclei by the rain drops or change in air mass and pollution history. The large change in the nuclei concentration was accompanied by a large change in the aerosol light scattering at the same time as detected by the nephelometer.

The positive correlation between the periods of low CCN concentration shown in Figure 1 and pressure troughs at nearby west Los Angeles (see top of Figure 1) suggests that an air mass change associated with a frontal passage could account for the large variation in numbers of CCN. Other phenomena associated with frontal or trough passages such as turbulent mixing and precipitation may also be the cause of large changes in the distribution of total numbers of cloud condensation nuclei.
 FIGURE 1. Variation in CCN Concentration with Time

The two instruments were tracking each other, and roughly experienced the same variation. The values of the light scattering coefficient and the corresponding values of the CCN concentration active at 0.6 percent supersaturation obtained over a period of about 80 hours are shown in Figure 2. It is clear from the figure that an increase in the light scattering coefficient is associated with an increase in the CCN concentration. A similar effect was found by Alkezweeny and Smith (6) in the same location for nuclei activation of 0.65 percent supersaturation. The best fit between all pairs was calculated by the least squares method, and gives the following relation

\[ b_{\text{scat}} = 6 \times 10^{-7} N^{0.84}. \]  

(1)

where \( b_{\text{scat}} \) is the light scattering coefficient in \( \text{m}^{-1} \), and \( N \) is the CCN concentration in \( \text{#/cc} \) active at 0.5 percent supersaturation. The best fit line shows a better correlation in the CCN concentration range about 700 to 2000 nuclei/cc than outside this range. The reason for this is that most of the data were obtained in this range, and each point may represent several points from different times.

The \( b_{\text{scat}} \) is related to the visibility \( (L_v) \) in meters, and mass concentration of suspended aerosol particles \( (M) \) in \( \mu g/m^3 \) through the following relations (Charlson, et al.): (8)
PROGNOSTIC ISENTROPIC TRAJECTORIES

W. E. Davis, J. L. Heffter* and M. E. Smith*

A kinematic, prognostic, isentropic trajectory program has been successfully run. Plans are discussed for future changes in the program as well as methods for evaluating the worth of the prognostic trajectories.

INTRODUCTION

In August, 1970, a meeting concerning prognostic trajectories was held in Silver Springs, Maryland. At the meeting it was decided that a comparison of different techniques for trajectory computations should be made. At a second meeting a decision was made for a cooperative effort between Battelle, Pacific Northwest Laboratories (PNL) and NOAA (Silver Springs, Md.) to produce a prognostic isentropic trajectory routine which would then be compared with previous techniques for computing trajectories. The following is a report of the progress made in programing prognostic isentropic trajectories.

Since flow in the first approximation is adiabatic, isentropic surfaces should be used to describe the flow of these air parcels. The same is true when one approaches the problem of prognostic trajectories. An adiabatic surface, along which one can produce an energy balanced trajectory, needs to be constructed from the prognostic information available. Thus, the first step for a prognostic trajectory computation is to be able to construct an isentropic surface and the second is to obtain wind speed information in order to be able to move the trajectory. (Later this information can serve as the basis for diabatic trajectory analysis if a decision is made to program one.)

Programing of an Isentropic Trajectory and Summary

The data used are from the Primitive Equation (P.E.) model. Temperature, height and wind fields for pressure surfaces were utilized to calculate the height and wind fields for a specific potential temperature. A series of check-out runs was made in December, 1971, and in January-February, 1972. After alterations

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* NOAA - Silver Springs, Md.

** Note: An isentropic surface is one where a moving parcel will conserve potential temperature.
in February, 1972, a successful computer run predicted wind and height fields which checked out successfully against the input data from the P.E. model. Several trajectories have been run using these fields. The same technique was used for computing u and v components for running the trajectories as was used by Peterson and Smith. The results of one of these trajectories tested against hand calculations indicate that the program is now running successfully.

Future Work

A check-out of these trajectories on a post-facto basis is planned as well as changes in the trajectory computer program to make the trajectories energy-consistent.

REFERENCES


HOURLY LOW-LEVEL ISENTROPIC TRAJECTORIES

W. E. Davis

Surface altimeter settings and temperatures as reported hourly are used to compute low-level stream functions. From these the mean flow can be determined for a scale of approximately 50 miles.

INTRODUCTION

Previous work on low-level isentropic and diabatic trajectories (Davis and Scott, (1); Engelmann and Davis)(2) have been based on maps of twelve hour increments. This work using such measurements could only describe macro-scale features of transport of the parcel. It was hoped that work could
be initiated at some time on a smaller scale. This would help determine the mesoscale flow patterns affecting transport as well as deposition of a material released in the lower atmosphere. The following is a report of such an attempt to reduce the scale from macroscale to mesoscale.

**Low-Level Stream Functions**

Previously Davis and Scott computed surface potential temperatures on a hourly basis to designate mesoscale areas of increased mixing. Further study indicated that the surface data could be applied to the calculation of stream functions on isentropic surfaces near the ground.

In studying Danielsen's work in which he calculated upper-level stream functions, the logical step would be to apply the same method to the surface stations. First, the altimeter setting report was used in a back calculation of surface pressure. Second, the surface pressure and the surface temperature were used to calculate the surface potential temperature (θ),

\[ θ = T \left( \frac{1000}{P} \right)^{R/C_p} \]

where θ - Temperature ° absolute
P - Pressure (mb)
R - Gas constant for air
C_p - Specific heat of air

Third, the surface Montgomery stream function (ψ) was calculated where

\[ ψ_{sfc} = C_p T + g z \quad (1) \]

where g - gravitational acceleration
z - the height of the station.

Fourth, Danielsen's equation in which

\[ Δψ = C_p \left( \frac{T}{θ} \right) Δθ \]

was changed to the computational form

\[ Δψ = C_p \left( \frac{P}{1000} \frac{R/C_p}{C_p} \right) Δθ; \quad (2) \]

here the bar indicates averaging with respect to θ.

To calculate \( \left( \frac{P}{1000} \right)^{R/C_p} \) several methods can be used. A linear approach was used:

\[ \left( \frac{P}{1000} \right)^{R/C_p} = \frac{1}{2} \left[ \left( \frac{P_{sfc}}{1000} \right)^{R/C_p} + \left( \frac{P_{est}}{1000} \right)^{R/C_p} \right] \]

\( P_{est} \) is the estimated pressure for the θ surface. The fifth and final step is to use Equations (1) and (2) to find \( ψ_θ = ψ_{sfc} + Δψ \). The term \( ψ_θ \) is the estimate of the stream function for some θ surface above the ground.

**Errors in ψ_θ**

Errors are found in the surface data and in estimating Δψ, Equation (2). Errors in surface temperature of 0.5°K and 0.005 in. of Hg in pressure readings combine to produce errors of \( ν \pm 0.02 \times 10^7 \text{ cm}^2\text{s}^{-2} \) in the stream function. These are minimal errors except in cases of low wind speeds. Larger errors can be generated in Equation (2) from an erroneous estimate of \( \left( \frac{P}{1000} \right)^{R/C_p} \). An error in this
term of 0.01 can generate problems if \( \Delta \phi \) is 5°K. This will cause an error in \( \psi \) of 0.01 \( \times \Delta \phi \times 10^7 \text{ cm}^2\text{ sec}^{-2} \). An error greater than 0.05 \( \times 10^7 \text{ cm}^2\text{ sec}^{-2} \) can create problems in estimating wind speed and directions. Figure 1 indicates the error in geostrophic winds where \( \psi = \frac{1}{E} \) \( \times \) \( \frac{R}{C_P} \); \( f \) is the Coriolis parameter and \( \delta r \) is taken normal to the flow.

Large errors in the term \( \left( \frac{p}{1000} \right)^{\frac{R}{C_P}} \) are not usually expected near the ground because of the slow variation of this function with height (see Figure 2). As long as the analysis is kept near the ground (i.e., \( \Delta \phi < 5° \)) accurate hourly patterns of \( \psi \) are available for an isentropic surface.

A definitive error analysis is difficult because of the nonlinearity in \( \left( \frac{p}{1000} \right)^{\frac{R}{C_P}} \). The foregoing provides guidelines for the use of an analysis so that errors can be minimized.

**CONCLUSIONS**

A method has been developed to estimate Montgomery stream functions on an hourly basis from surface hourly reports. Errors encountered in the method have been pointed out and procedures indicated for computing the stream functions.

**FIGURE 1.** Error in Geostrophic Wind due to Errors in Stream Function
FIGURE 2. Graph of \( \left( \frac{p}{1000} \right)^{R/Cp} \) with Pressure

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CLIMATOLOGICAL MEASUREMENTS ON THE ARID LANDS ECOLOGY RESERVE IN 1971

J. M. Thorp and W. T. Hinds

Measurements to establish a climatology of the ALE Reserve continue. All recorded climatological data are now being collected at a central location and summarized by computer. Resulting monthly data summaries are distributed to ALE Reserve investigators.

During the 1971 bio-year (October 1970 through May 1971) the ALE Reserve rain gages were visited on a monthly basis. The total rainfall and snowfall (in centimeters of water) is shown in Figure 1. Precipitation totals were down from the 1970 bio-year

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FIGURE 1. Bio-year Precipitation (cm of Water)
October 1970-May 1971
amounts, ranging from 11 cm to 22 cm compared to 16-25 cm in 1970.

The 1971 calendar year (actually December 30, 1970 to January 4, 1972) precipitation is shown in Figure 2. As one would expect, the patterns exhibit similar distribution. Spotty summer shower precipitation probably accounts for the somewhat less regular pattern for the calendar year. Precipitation quantities on both charts represent minimum approximations of precipitation that actually fell. No corrections have been applied to account for losses occurring from evaporation, rain driven by high wind, or drifting snow. An oil film is used as an evaporation suppressant, nevertheless there is some loss of gage water by evaporation processes, especially in hot weather. Gages along the crest of Rattlesnake Mountain are particularly subject to wind error during rain or snow and it is possible for drifted snow to completely cover a gage in a location that experiences frequent drifting (#14 is affected this way).

A computer program for ALE climatology has been written and all climatological data from the ALE Reserve are now being summarized by this method. Data are processed to provide applicable weekly, monthly and
annual summations. (1) Table 1 indicates the instrumentation currently in use as source material for the above mentioned data summarization.

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W Weekly
M Monthly

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KINEMATICALLY POSSIBLE FLOWS EXTERIOR TO JETS AND PLUMES

W. G. N. Slinn

A progress report is given of analytical analyses of flows exterior to jets and plumes penetrating into the half space \( z > 0 \). It is shown that the exterior flow is essentially horizontal near a plume and the case of nonzero mean wind and multiple plumes can then be treated using superposition. Qualitative results are presented for bifurcation and merging of plumes.

INTRODUCTION

There exist many studies of atmospheric jets, plumes, and thermals. Three review articles which also reference most of the pertinent literature are those by Turner, Briggs, and Scorer. In contrast, there are relatively few reports which concentrate on the flow exterior to jets and plumes. Such studies are intriguing when one inquires, for example, whether or not there exists an optimum arrangement of cooling towers in the neighborhood of a nuclear reactor, such that if there were an accidental release of radioactivity, it would be entrained by the cooling tower plume, brought to a higher elevation, and thereby dispersed less hazardously. Or, as another example, one wonders of potential hazards caused by entrainment into moist cooling tower plumes of pollutants from fossil-fuel-plant plumes; specifically, there is concern about possible enhanced production of sulfuric acid.

The purpose of this study was to explore the degree to which analytical analysis could provide insight into solutions of these practical problems. The only other analytical studies of exterior flows which apparently are available are that of the flow exterior to a momentum jet emerging into unbounded space (e.g., see Landau and Lifshitz) and those by Taylor. Unfortunately the latter is just a listing of results and is not of much value to the uninitiated.

THE TURBULENT (MOMENTUM) JET

Before considering exterior flows, it seems appropriate to review the analyses of the interior flows. Consider first the flow within a turbulent (pure momentum) jet. Notation to be used is shown in Figure 1. At distances, \( z \), large compared with the initial radius of the jet, \( a \), the only characteristic length for the problem is \( z \). Therefore, by dimensional arguments it is reasoned that the radius of the jet, \( R \), will be proportional to \( z \). Landau and Lifshitz take

* Work supported in part by the Battelle Memorial Institute's Physical Sciences Program.
It is interesting that the above assumptions contain a prediction for the rate of entrainment of external air. To see this, note that the mass flux at any height, \( Q = \rho \omega R^2 \). If \( \rho \) is independent of height, then (1) and (3) give

\[
Q = A z,
\]

(4)

where \( A \) is a constant (which Landau and Lifshitz suggest to be about 1.5 \( Q_0/a \) where \( Q_0 \) is the mass flux emerging from the jet.) From (4), the amount of air entrained through the sides of an infinitesimal slice of the jet of height \( dz \) is

\[
dQ = A dz = A \cos \delta \, dr.
\]

(5)

But the amount entrained is also given by

\[
dQ = -2\pi R \rho_0 \nu_r(\xi = R, z) \, dr,
\]

(6)

where \( \nu_r \) is the \( r \)-component of the velocity, \( \nu \), of the external air and \( \rho_0 \) is its density. If variations of \( \rho_0 \) with height are ignored, then using (1) and (3), (5) and (6) give

\[
\nu_r(R, z) = -\alpha \omega.
\]

(7)

That is, the two assumptions used in the construction of the theory contain the results that the entrainment is proportional to the updraft velocity.

Turning now to the exterior flow, there are a number of ways to solve for the kinematically (but not necessarily dynamically) feasible flow exterior to a jet emerging into the
half space, \( z > 0 \). Proceeding as Landau and Lifshitz, but using different boundary conditions, we solve continuity, \( \nabla \cdot \vec{V} = 0 \) and the postulated condition of irrotationality, \( \nabla \times \vec{V} = 0 \). The solution that vanishes at infinity, satisfies \( v_z = 0 \) at \( z = 0 \) and \( v_\theta = -\alpha w \) on the jet boundary, is

\[
\vec{V} = -\frac{A}{2 \pi \rho_a} \frac{1}{\rho} \left[ \hat{e}_r + \cot \theta \hat{e}_\theta \right].
\]  

(8)

Another way is to solve for the velocity potential; defined via \( \vec{V} = -\nabla \phi \), which satisfies Laplace's equation, \( \nabla^2 \phi = 0 \). In the solution it is necessary to include the infrequently used Legendre polynomials of the second kind, \( Q_n \), which have a logarithmic singularity at \( \theta = 0 \), which is outside the flow region of interest. A third way to obtain a solution which evidently is the method used by Taylor, is to solve for a Stokes' stream function. Distributing a line sink along the axis of symmetry (and from the above, \( dQ \propto R w \) is independent of height and therefore the sink strength per unit length is independent of height) leads to \( d\psi = A dz/2 \pi \rho_a \), or

\[
\psi = -\frac{A}{2 \pi \rho_a} r \cos \theta.
\]  

(9)

from which one can obtain the velocity components.

The most interesting feature of these results is that exterior to a jet emerging into a half space there exists a kinematically possible flow which is horizontal. Thus, for example from (9), we have

\[
v_\xi = \frac{1}{\xi} \left( \frac{\partial \psi}{\partial z} \right) = -\frac{A}{2 \pi \rho_a} \frac{1}{\xi},
\]

(10)

\[
v_z = -\frac{1}{\xi} \left( \frac{\partial \psi}{\partial \xi} \right) = 0.
\]

(11)

Further, the flow is not only horizontal but also, since \( v_\xi \) is independent of \( z \), then the flow appears to be dynamically feasible: no instabilities generated by velocity shear should develop (except in a surface boundary layer).

**BUDYANT PLUMES**

The case of buoyant plumes is more complicated. For the interior flow it is customary to continue to assume that

\[
R = z \tan \delta_b.
\]

(12)

To determine \( w \) and \( v_\theta \), conservation equations are needed. The conservation of mass,

\[
\frac{d}{dz}(\rho w R^2) = -\frac{2R}{\cos \delta_b} \frac{\partial v_\theta}{\partial R},
\]

simply equates the increase of mass within the plume to the mass entrained at the boundary; it yields \( v_\theta \) if \( \rho \) and \( w \) are known. The \( z \)-component of the momentum equation is

\[
\frac{d}{dz}(\rho w^2 R^2) + \frac{d}{dz} R^2 \frac{dR}{dz} = (\rho_a - \rho) g R^2 + 2Rp \tan \delta_b
\]

\[ - 2R v_\theta(R) \rho_a v_z(R)/\cos \delta_b \]

(14)
Upon application of the continuity equation and using \( R = z \tan \delta_b \), we reduce (14) to

\[
\frac{d}{dz}(w^2 R^2) = BR^2 + v_z(R) \frac{d}{dz}(WR^2),
\]

(15)

where we have used the Boussinesq approximation and defined the buoyant force per unit mass to be

\[
B = B(z) = \frac{\rho_c - \rho_a}{\rho_a} g.
\]

(16)

Using (12) we could solve (13) and (16) for the two unknowns \( \rho \) and \( w \) provided the external flow were known.

A solution that is frequently quoted is derived using three additional assumptions:

1. The influence of \( v_z \) on the momentum equation, (15), is negligible.
2. The difference between \( \rho_c \) and \( \rho \) in the continuity equation is negligible.
3. The buoyancy flux is a constant, i.e.,

\[
B \cdot R^2 = B_\infty \cdot \rho_0 R_\infty^2.
\]

(17)

Notice that with these assumptions, we have essentially specified \( \rho \), \( \rho_0 \) and \( v_z \). Using them in (15) leads to

\[
w = w_\infty \left( \frac{z_\infty}{z} \right)^{1/3},
\]

(18)

and using them and (18) in (13) gives

\[
v_\epsilon = -\alpha w,
\]

(19)

with \( \alpha = (5/6) \sin \delta_b \) which from observations is about 0.1. Thus, as for the turbulent jet, the entrainment is proportional to the updraft velocity. [Alternatively, one could have assumed (19) and derived, e.g., (12).]

We now look at the exterior flow. Again we seek only a kinematically possible solution, i.e., one that satisfies continuity. The simplest way to do this is to find a Stokes' stream function which satisfies the boundary conditions imposed by the ground and the plume. By definition, the stream function satisfies continuity.

It is relatively easy to find a stream function that satisfies any distribution of sinks and sources along the plume axis. A distribution that might be of particular interest is shown in Figure 2, where we have attempted to include the effect of the "sink" at the base of a cooling tower (i.e., the inlet) and a thermal inversion at height \( H \).

The infinitely repeated mirror images of sources and sinks are used to force the boundary conditions that \( v_z = 0 \) at \( z = 0 \) and \( z = H \) and to aid in the calculations.

The stream function is given by

\[
\psi(\xi, \zeta) = \frac{1}{4\pi} \int_{\zeta = -\infty}^{\infty} f(\zeta) \frac{z - \zeta}{[\xi - \zeta]^2 + \zeta^2]^{1/2}} \, d\zeta,
\]

(20)

where \( f(\zeta) \) is the source strength per unit length. To solve (20), \( f(\zeta) \) is expanded in a series of cosines

\[
f(\zeta) = \sum_{n=1}^{\infty} a_n \cos nk\zeta,
\]

(21)

where \( a_n \) are the Fourier coefficients and \( k = \pi/H \). Substituting (21) into (20) and assuming uniform convergence of the resulting series, we obtain
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FIGURE 2. Distribution of Sources and Sinks Used to Model the Flow Exterior to a Cooling Tower, Beneath a Temperature Inversion at Height H.

\[ \psi(\zeta, z) = \sum_{n=1}^{\infty} a_n \sin nk \zeta K_1(nk \xi), \]  

(22)

where \( K_1 \) is a modified Bessel function. From (22) we can obtain the velocity components

\[ v_\zeta = \sum_{n=1}^{\infty} a_n nk \cos nk \zeta K_1(nk \xi), \]  

(23)

and

\[ v_z = \sum_{n=1}^{\infty} a_n nk \sin nk \zeta K_0(nk \xi). \]  

(24)

There are a number of interesting features of these results. First, \( v_z = 0 \) at \( z = 0 \) and \( z = \frac{1}{2} \pi k^{-1} = H \), as was desired. Also, for large distances from the axis,

\[ \psi \sim \sum_{n=1}^{\infty} a_n \sin nk \xi \left( \frac{\xi}{2nk} \right)^{1/2} e^{-nk \xi}, \]  

(25)

which vanishes. Thus at large distance the cooling tower-inversion situation appears as a dipole. At the other extreme, i.e., for distances from the axis small compared with \( H \), then

\[ v_\zeta = \sum_{n=1}^{\infty} a_n nk \cos nk \zeta \frac{1}{nk \xi}, \]  

(26)

or
\[ v_\zeta = \frac{1}{2\pi} \zeta, \quad \zeta > 0. \]  
(27)

To obtain information about \( v_\zeta \), we notice from (24) that as \( \zeta \to 0 \), \( v_\zeta \) has a logarithmic singularity. However, this is inside the plume boundary. The streamlines are given by \( \psi = \text{constant} \) and from (22), for small \( \zeta \),

\[ \psi(\zeta - 0, z) = \frac{1}{2\pi} \sum a_n \sin nkz \zeta \frac{1}{nk\zeta}. \]
(28)

Thus, for small \( \zeta \), the streamlines are in surfaces of constant \( z \) and therefore the flow is horizontal.

To determine if the flow might be dynamically possible, we look at the vorticity

\[ \omega = \nabla \times \mathbf{v} = \phi \left[ \frac{\partial v_\zeta}{\partial z} - \frac{\partial v_z}{\partial \zeta} \right]. \]
(29)

Substituting (23) and (24) into (29) gives \( \omega \equiv 0 \). Thus the solution appears to be dynamically acceptable except for the inadequacy at \( z = 0 \), where \( v_\zeta \neq 0 \), whereas in reality there should be a no slip condition and a boundary layer from which vorticity diffuses into \( z > 0 \). However, even this inadequacy is probably not too significant since the boundary layer may be thin in the favorable, stabilizing pressure gradient caused by the sink at \( z = 0 \). Since \( \omega = 0 \), a velocity potential exists and it is easily found to be

\[ \phi = \frac{1}{2\pi} \sum a_n \cos nkz K_0(nk\zeta). \]
(30)

The reality of the above solution could be challenged, especially the significance of the infinitely repeated distribution of sources and sinks. In response to this, we note that in view of the above we have essentially solved \( \nabla \times \mathbf{v} = 0 \), \( \nabla \cdot \mathbf{v} = 0 \) and satisfied all appropriate boundary conditions. Therefore the solution we have obtained is the desired solution, and the method used to obtain it (method of images) is irrelevant. Further confidence in the result is obtained via comparison of these results with those recently obtained numerically by Trent. (6) His numerical solutions show that in the neighborhood of both jets and plumes emerging into a half space, the exterior flow is essentially horizontal.

**The Influence of a Mean Wind and/or Adjacent Plumes**

If there exists a mean wind then the analysis could become very complicated since the symmetry of the problem is destroyed. This lack of symmetry also precludes numerical calculations in reasonable computing times. However, we shall explore this problem for the case that the mean wind is light enough so that the dynamics of the plume are still described by the above analysis. To determine the flow exterior to the jet or plume, we utilize the above results that in both cases, the flow in the neighborhood \( (\zeta < H) \) of the jet or plume is essentially horizontal. Then, since the equations for \( \phi \) or \( \psi \) are linear, superposition can be used.

Then consider the flow in some plane \( z = \text{const} \). The total stream function will be the sum of the stream function for a source of strength per
unit length, \( f(z) \), and that for a uniform stream of speed \( U(z) \). Taking the derivatives of the resulting stream function gives the velocity components
\[
v_\xi = \frac{f(z)}{2\pi} + U \cos \phi
\]
and
\[
v_\phi = -U \sin \phi,
\]
where \( \phi \) is the angle from the downwind direction as shown by Figure 3. There we have sketched the flow field for a particular \( z \) at which the plume is still entraining fluid.

It is interesting that the higher the wind speed, the closer the stagnation point (s.p.) will move toward the plume boundary. In this naive picture the stagnation point could move inside the jet boundary and would set up two counter-rotating vortices within the plume. It is suggestive that the ingredients of this picture are essentially correct for cases of light turbulence in the exterior flow, and that this is the cause of the frequently observed bifurcation of plumes. In this picture, bifurcation would occur at height \( z \) given by \( v_\xi(\xi = R, \phi = 0; z) = 0 \). That is, from (31), \( f(z) = -2\pi RU \). For a jet, the source

FIGURE 3. Sketch of a Plan View of the Flow Exterior to a Jet or Plume at some Height \( z \) for the Case of Nonzero Mean Wind. In the top half of the figure, the velocity vectors and components are shown at a number of locations. In the lower half, streamlines are sketched.
strength per unit length is the constant value \(-A/\rho_a\). Therefore, using \(R = z \tan \delta\), it is suggested that in a light wind and for low-level turbulence in the exterior fluid, bifurcation might set in at a height

\[
\zeta = \frac{C}{2\pi \tan \delta \rho_a U} \sim \frac{Q_0}{\rho_a U} \sim \frac{\nu_E a}{U},
\]

(33)

where \(\nu_E\) is the jet's exit velocity. For the buoyant plume discussed earlier, for which \(f(z) \sim z^{2/3}\), then \(z_0 \sim \left(\frac{\rho}{U^3}\right) z_0\), where \(z_0\) is the height at which \(w = w_0\). Extension of these considerations to line vortices cast off by convective storms is intriguing.

In reality, when the stagnation point approaches the jet or plume boundary, the inner flow would not be able to entrain fluid from the exterior flow and the assumptions of a vertical plume would have to be abandoned. To obtain a qualitative idea of the initial tilt of a plume, we can estimate the \(x\) (downwind) component of the momentum equation to be

\[
\frac{d}{dz} (R^2 u) \sim 2awRU.
\]

(34)

For arbitrary \(w\), using \(R = z \tan \delta\), (34) leads to

\[
\tan \theta (z) \equiv \frac{U}{w} = \gamma \frac{U}{w}
\]

(35)

with \(\gamma = \alpha \cot \delta \approx 1\). The result (35) is what might have been decided by dimensional arguments.

If there exist adjacent jets or plumes, then the flow field can be found in a manner similar to the above. We look at the flow in each plane and use superposition. In Figure 4 a simple example is sketched. This example is particularly interesting in that two stagnation points appear on the centerline. If the axes of two plumes were initially a distance \(D\) apart, then they would bend toward each other and merge. From (35) an estimate of the height at which this would occur is \(z = D/c\) where \(c = \gamma \tan \delta = \alpha \approx 0.1\).

**SUMMARY**

This above represents a progress report of a brief look at the flow exterior to jets and plumes. It contains some interesting features but the analysis is primitive and it is premature to draw any substantial conclusions. It should be reemphasized that we have considered only the case for jets or plumes emerging into the half space \(z > 0\). Thus, in particular for the solution to the flow exterior to a momentum jet, the results should not be applied to describe the flow exterior to a tall stack. This problem has been solved\(^{(4)}\) but since it does not predict horizontal flow, the effect of mean wind and adjacent plumes cannot be handled so simply.

The return on the small investment of effort that has been put into this analysis is very encouraging. It has been seen that exterior to jets and plumes emerging into a half space, the flow is essentially horizontal and the speed varies as the reciprocal of the distance from the axis. Thus, an estimate of the fate of a radiation
FIGURE 4. Sketch of a Plan View of the Flow Field Exterior to two Cooling Tower Plumes at some Height $z$, for the Case of Nonzero Mean Wind. A simple case for which there exists considerable symmetry has been chosen. Notice that in this case, of fairly low wind speed, two stagnation points appear on the centerline. At higher wind speeds these stagnation points can vanish and stagnation points appear downwind of each plume.

leak near a cooling tower can be evaluated. Further, because the exterior flow is essentially horizontal, the interaction between jets and plumes and the influence of a mean wind can be studied relatively easily. Thereby, interesting qualitative results have been obtained for the tilt and the merging of plumes and their bifurcation. Another effect which may be within reach of this type of analysis but which was not investigated is the frequently observed kink instability of plumes.

I wish to thank S. A. Slinn for her graphic evaluations of Equations (31) and (32) and sketches of the results (Figures 3 and 4).

REFERENCES

AIRBORNE RELEASE OF PLUTONIUM AND ITS COMPOUNDS
DURING OVERHEATING INCIDENTS

J. Mishina and L. C. Schwendiman

A series of studies to evaluate the fractional airborne release of plutonium under various postulated accident conditions has been in progress for some time. Data generated in earlier laboratory scale studies are reviewed. A larger scale facility -- The Radioactive Aerosol Release Facility -- was placed in operation providing the capability for measurement of fractional airborne release of radioactive particles from burning material on a more realistic scale. The amount and aerodynamic equivalent size distribution of particles that may become airborne under conditions found in some shipping accidents were measured in experiments conducted in a special wind tunnel in the Radioactive Aerosol Release Test Facility.

As the era of nuclear power comes of age, plutonium is handled and utilized in ever-increasing amounts. An abiding interest in assuring the maximum protection to the population prompts a continuing effort to evaluate the potential consequences of situations

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*Work supported by the Atlantic Richfield Hanford Company's Environmental and Regulatory Technology Program.
Incidents in which control criteria involving plutonium and its compounds were exceeded were reviewed,\(^1\) and existing data on the airborne release of plutonium and its compounds presented.\(^2\) Small scale laboratory experiments--yielding order of magnitude estimates of the airborne release of plutonium and its compounds\(^3\)-\(^7\) were conducted during earlier periods. The retrospective review showed that about 70 percent of the incidents occurred during what is nominally considered chemical processing, 28 percent during handling and storage, and only 2 percent in transportation. Fire played a role in 13 of the 20 incidents resulting in airborne contamination.

Based upon the needs revealed by the review of data on the airborne release of plutonium, a laboratory program was conducted to provide information on the fractional airborne release of particles when plutonium and its compounds are subjected to thermal stress and differing airflows.

- **Metal**--Some \(10^{-6}\) to \(10^{-5}\) percent of plutonium present as the ignited metal was entrained by air passing horizontally over oxidizing cylindrical specimens. As much as 0.03 percent of the oxide formed could be elutriated by passing air up through the residue after cooling. The fractional airborne release from large pieces of plutonium metal oxidizing at an accelerated rate was in the same range--the maximum measured value was 0.05 percent. The Median Mass Diameter (MMD) of the particles airborne was 4.2 microns Aerodynamic Equivalent Diameter (AED).

- **Powders**--Entrainment of particles during the heating of powders of plutonium compounds--oxide, partially oxidized oxalate, oxalate, fluoride and the solid residues from the evaporation of nitrate solutions--varied over a wide range. A fractional airborne release of the order of \(10^{-6}\) percent was measured in air passed over the residues from nitrate solution evaporation, heated to 130\(^\circ\)C. When heated to as high as 1000\(^\circ\)C, as much as 0.12 percent could be carried out the chimney in an upswEEP of air. The MMD of these particles was microscopically measured to be about 20 microns. The other powders heated in this manner indicated fractional airborne releases of: (a) up to 0.82 percent from the partially oxidized oxalate, (b) up to 0.9 percent from the oxalate, (c) up to 0.05 percent from fluoride, and (d) up to 0.025 percent from oxide powder less than a 325 mesh. The particles airborne were measured microscopically in some experiments and were quite coarse--a MMD of 25 microns for partially oxidized oxalate, a range of 5 to 60 microns from oxalate and MMD of 26 microns from fluoride powder.

- **Nitrate Solutions**--The amount airborne under the heating of aqueous plutonium nitrate solutions varied widely with the heating rate used. Aqueous plutonium nitrate was chosen as representative of solution behavior since it is frequently encountered at various processing stages. During the
slow evaporation of shallow pools (about 3 mm deep) of concentrated solution at temperatures up to 150°C, as much as $3 \times 10^{-3}$ percent was carried away by dry air (dew-point of -50°F) flowing horizontally over the liquid at 100 cm/sec. With deeper pools (initially about 15 cm deep) of dilute solution as much as $6 \times 10^{-3}$ percent was airborne during the slow evaporation of 90 percent of the volume. As the heat input was increased, the amount airborne increased. Releases as high as 0.084 percent were obtained when the heat input was sufficient to cause surface disturbance and 0.18 percent at a vigorous boil. Characterization of particles airborne was not undertaken due to the small numbers collected.

- **Plutonium Nitrate Dried on Sand:** Some experiments were conducted to determine the amount of plutonium release during the heating of sand bearing dried plutonium nitrate. Small quantities of materials were indirectly heated in stainless steel cups at temperatures up to 1000°C for 1 hour periods. Using completely dried material--sand surfaces were completely undisturbed after the run--up to 0.002 percent was found in air drawn up and around the sample at 100 cm/sec. If moisture were present, sand particles were dislodged and some of these particles entrained in the air stream. Releases under these conditions ranged from 0.0053 to 0.028 percent.

- **During Combustion of Flammable Materials Containing Active Particles:** As a prelude to large engineering-scale experiments, various flammable materials; wood, paper and paper products, rubber and plastic were impregnated with plutonium or uranium nitrate solutions or finely-divided uranium dioxide powder (MMD of approximately 6 microns AED). Uranium compounds were used as a stand-in for plutonium. As much as 1 percent of the plutonium incorporated was entrained in an upsweep of air having a nominal velocity of 5.6 cm/sec. Using an upsweep of air having a nominal velocity of up to 100 cm/sec, 8 percent of the uranium incorporated as a dried nitrate could be found on the filter sealing the end of a 4 inch diameter by 30 inch long chimney. In most cases lesser amounts were entrained, generally around 1 percent. Using uranium dioxide powder, in most cases at least 10 percent was made airborne during the fire and in some cases up to 40 percent.

- **A larger scale facility--The Radioactive Aerosol Release Test Facility:** was placed in operation providing the capability for measurement of fractional airborne release of radioactive particles from burning materials at a more realistic scale. Of the several possible materials studied uranium was shown to be the most suitable stand-in for plutonium in these experiments.
The amount and aerodynamic equivalent size distribution of particles that may become airborne under conditions found in some shipping accidents were measured in experiments conducted in a special wind tunnel in the Radioactive Aerosol Release Test Facility. Airborne release from uranium dioxide powder or nitrate liquid dispersed on four types of surfaces were evaluated in twenty-six combinations of conditions believed to be representative of circumstances which would prevail in a transportation accident. Two wind speeds less than 4 and 23 mph were used, both with and without a fire.

The fractional airborne releases measured under these conditions are tabulated in Table 1 and 2. Table 1, "Aerodynamic Entrainment of Uranium from Various Surfaces," are fractional airborne releases in the absence of a gasoline fire, and Table 2, "Percent of Source Airborne in Respirable Range During Gasoline Fires," are for the experiments involving a gasoline fire. These values were obtained by averaging the three samples obtained in each experiment -- two filters and an 8 stage cascade impactor. Since inhalation is the principal hazard, the gross quantity airborne was weighted by the fraction less than 10 microns AED (nominal respirable range) to give the percent of source airborne in the respirable range. Two areas of burning (approximately 2.9 and 1.2 sq ft) were used.

No single variable of those encompassed appeared to exert an over-riding controlling influence on the fractional releases in the absence of a gasoline fire.

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### TABLE 1. Aerodynamic Entrainment of Uranium from Various Surfaces (Percent of source per hour in respirable size range.)

<table>
<thead>
<tr>
<th>Surface</th>
<th>Uranium Dioxide Powder</th>
<th>Uranium Nitrate Solution</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>&lt;4 mph</td>
<td>23 mph</td>
</tr>
<tr>
<td>Bare Ground</td>
<td></td>
<td></td>
</tr>
<tr>
<td>~2.9 sq ft</td>
<td>0.016</td>
<td>0.12</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Vegetation Cover</td>
<td></td>
<td></td>
</tr>
<tr>
<td>~2.9 sq ft</td>
<td>0.00007</td>
<td>0.0059</td>
</tr>
<tr>
<td>Stainless Steel</td>
<td></td>
<td></td>
</tr>
<tr>
<td>~1.2 sq ft</td>
<td>0.015*</td>
<td>—</td>
</tr>
<tr>
<td>Asphalt-Gravel</td>
<td></td>
<td></td>
</tr>
<tr>
<td>~1.2 sq ft</td>
<td>0.0011*</td>
<td>—</td>
</tr>
</tbody>
</table>

* Solid residue remaining after gasoline fire.
** Amount released assumed to be linear with time during period when flame was visible.
† Solid residue of air-dried UNH solution.
TABLE 2. Percent of Source Airborne in Respirable Range During Petroleum Fires

<table>
<thead>
<tr>
<th>Experiment ID</th>
<th>Source Description</th>
<th>Air Velocity (mph)</th>
<th>Surface Area (sq ft)</th>
<th>Grams Uranium In Source</th>
<th>Grams Uranium (sq ft)</th>
<th>Percent Source Airborne in Respirable Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>18a</td>
<td>UNH solution on stainless steel</td>
<td>23</td>
<td>1.23</td>
<td>25.5</td>
<td>20.7</td>
<td>4.3</td>
</tr>
<tr>
<td>19a</td>
<td>UNH solution on stainless steel</td>
<td>&lt;4</td>
<td>1.23</td>
<td>25.5</td>
<td>20.7</td>
<td>0.44</td>
</tr>
<tr>
<td>14</td>
<td>UO₂ powder on vegetation cover</td>
<td>23</td>
<td>2.89</td>
<td>69.5</td>
<td>24</td>
<td>0.59</td>
</tr>
<tr>
<td>15</td>
<td>UNH solution on vegetation cover</td>
<td>23</td>
<td>2.89</td>
<td>51.1</td>
<td>17.7</td>
<td>0.17</td>
</tr>
<tr>
<td>20a</td>
<td>UNH solution on asphalt/gravel mixture</td>
<td>&lt;4</td>
<td>1.23</td>
<td>25.5</td>
<td>20.7</td>
<td>0.16</td>
</tr>
<tr>
<td>17a</td>
<td>UO₂ powder on stainless steel</td>
<td>&lt;4</td>
<td>1.23</td>
<td>19.4</td>
<td>15.8</td>
<td>0.079</td>
</tr>
<tr>
<td>16</td>
<td>UNH solution on vegetation cover</td>
<td>&lt;4</td>
<td>2.89</td>
<td>51.1</td>
<td>17.7</td>
<td>0.075</td>
</tr>
<tr>
<td>21a</td>
<td>UO₂ powder on asphalt/gravel mixture</td>
<td>&lt;4</td>
<td>1.23</td>
<td>58.1</td>
<td>47.2</td>
<td>0.029</td>
</tr>
<tr>
<td>7</td>
<td>Air dried UNH residue on ground</td>
<td>23</td>
<td>2.89</td>
<td>51</td>
<td>17.6</td>
<td>0.038</td>
</tr>
<tr>
<td>3</td>
<td>UO₂ powder on ground</td>
<td>&lt;4</td>
<td>2.89</td>
<td>74.9</td>
<td>25.9</td>
<td>0.030</td>
</tr>
<tr>
<td>12</td>
<td>UO₂ powder on vegetation cover</td>
<td>&lt;4</td>
<td>2.89</td>
<td>78.3</td>
<td>27.1</td>
<td>0.013</td>
</tr>
<tr>
<td>9</td>
<td>Air dried UNH residue on ground</td>
<td>&lt;4</td>
<td>2.89</td>
<td>51.5</td>
<td>17.8</td>
<td>0.0072</td>
</tr>
<tr>
<td>5</td>
<td>Air dried UNH residue on ground</td>
<td>&lt;4</td>
<td>2.89</td>
<td>51</td>
<td>17.6</td>
<td>0.0045</td>
</tr>
<tr>
<td>10</td>
<td>UNH solution on ground</td>
<td>&lt;4</td>
<td>2.89</td>
<td>47.8</td>
<td>16.3</td>
<td>0.0013</td>
</tr>
</tbody>
</table>

release. The combination of all factors produces a situation favorable or unfavorable to release. The highest fractional release was measured for a uranyl nitrate solution spilled on a stainless steel surface in a 23 mph wind during a gasoline fire, 4.3 percent in the respirable range. Reducing the wind speed reduced the fractional airborne release an order of magnitude. Lesser quantities were released under the other conditions evaluated with fractional releases from bare soil on which uranyl nitrate has been spilled giving the smallest values.

A large scale field test was performed to validate that conditions generated in the lesser scale wind tunnel experiments were an adequate representation of those found in large scale gasoline fires outdoors. Five hundred gallons of gasoline were dumped onto an instrumented 30 ft × 30 ft plot of typical Hanford terrain and ignited. Observed and measured parameters were in reasonable agreement with those generated in the wind tunnel simulations. The burning characteristics considered important were the duration of intense burning, the subsequent
period of flame breakup and extinguishment, temperatures achieved, and areal extent of the fire per gallon of fuel.

REFERENCES


CHARACTERIZATION OF RADIOACTIVE PARTICLES IN A PLUTONIUM PROCESSING PLANT EXHAUST SYSTEM

J. Mishima and L. C. Schwendiman

Filter and cascade impactor samples were taken of the stack gases and various exhaust streams of a plutonium processing plant to characterize by aerodynamic characteristics the amounts and distribution of particles with their associated radioactivity. Only general conclusions can be drawn from the limited data obtained thus far: (a) the overall efficiency of the exhaust system is high; (b) little, if any, of the alpha activity leaving the stack is being recycled back into the ventilation system; (c) the plutonium present appears to be attached to large, nonactive particles.

INTRODUCTION

The Plutonium finishing plant at the Hanford site has a long history of safe operation, and radioactive material release is controlled well within the established limits. But an abiding public concern of possible effects of plutonium processing operations on the atmospheric environs justifies a continuing effort to reduce potentials for release to the lowest practicable level. Routine stack monitoring is performed as part of the radiological responsibility but does not provide all the data necessary to determine the fate of particles after emission. Knowledge of the distribution of the activity's aerodynamic particle size fractions is necessary to determine the airborne behavior of material released into the atmosphere. The contribution of the various components of the exhaust system to the total emission may be helpful in delineating areas of potential gain or identifying potential trouble points within the system. A body of information has been published on the amounts and size distribution of plutonium particles in various types of installations and in accident situations, (1-11) but little information is available on the nature of particles within the ventilation system of plutonium processing plants.

OBJECTIVES

- The current study was undertaken to determine the distribution of aerodynamic sizes of plutonium-containing particles present in the plant gaseous effluents. The total concentration of plutonium as particulates was also to be determined.

* Work supported by the Atlantic Richfield Hanford Company’s Environmental and Regulatory Technology Program.
By direct use of the aerodynamic characteristics for classification, such factors as particle density and shape, agglomeration, etc., need not be of concern since the property of interest is measured. Obtaining size distribution data in terms of aerodynamic characteristics will generate information which can be directly used in existing dispersion and deposition models that can forecast spatial distribution and predict any build-up on the terrain.

- A second objective was to determine the amount and distribution of radioactivity associated with particles contributed by various components of the building exhaust system.

SUMMARY

Filter and cascade impactor samples were taken of the processing plant stack gases and of various exhaust streams within the building ventilation system to characterize the amount and distribution by aerodynamic characteristics of particles with their associated radioactivity. Stack samples were extracted at the base of the stack. Exhaust stream samples were taken at various locations downstream of final filtration. Sampling periods ranged from 7 to 63 days and high volume samples (flows up to 72 cfm) taken of streams where extremely low activity concentrations were anticipated. Volumes of gases sampled ranged from $10^4$ to $4.4 \times 10^6$ cubic feet. Two components of the exhaust system—the exhaust from the Incinerator Building and the 26-inch (Process) vacuum system exhaust—may be contributing a significant amount of the total activity emitted. Another system with significant potential to contribute to the loading, the HF system exhaust, was not on the line during the sampling period and could not be evaluated. Air samples taken in the inlet plenum of the supply to the building showed low activity concentrations—$10^{-8}$ dpm/cu ft to $10^{-7}$ dpm/cu ft—indicating that little, if any plutonium is recycled from the stack. The activity distribution into aerodynamic fractions was approximately log-normally distributed and surprisingly coarse—with one exception, Median Activity Diameter (MAD) ranged from 1.6 to ~20 micron Aerodynamically Equivalent Diameter (AED).

Because these measurements were the first of their kind ever taken within the facility and because the plant operations during the sampling period were not regarded as fully representative, the results to date are regarded as indicative rather than definitive. A few general conclusions can be drawn from the limited data obtained.

- Although the level of activity in the various exhaust streams could be determined with high accuracy with the data obtained, the overall efficiency of the exhaust system is high. The low quantity of alpha particle emitters leaving the stack and low activity concentrations in the exhaust streams indicate satisfactory performance of the system.
- Little, if any, of the alpha-emitters emitted via the stack is being recycled back into the ventilation system.
- The plutonium present appears to be attached to large, nonactive particles.
REFERENCES


Aircraft Instrumentation for Atmospheric Research

INTRODUCTION

A Cessna 411 aircraft is being instrumented for the purpose of conducting airborne measurements of parameters which are important to our ongoing AEC atmospheric sciences research programs. The first phase of this modular instrumentation is directed especially toward measuring concentrations, size distributions, and other characteristics of atmospheric aerosols as a function of meteorological conditions. In the second phase, which is about to get underway, a module will be installed for measurement of trace gas concentrations; specifically, an airborne quadrupole mass spectrometer and associated equipment, real-time SF₆ and SO₂ samplers, and possibly a nucleogenic gas detector will be installed. Other phases of instrumentation will be used for cloud physics, fundamental turbulence, and atmospheric transport and diffusion studies. The module to measure standard meteorological parameters is installed and will be common to all phases of instrumentation.

The all-weather, radar equipped, twin engine aircraft is capable of cruising at speeds between 110 to 230 mph for a period of 5 hours and up to altitudes of 25,000 ft. In the following sections electrical power distribution, sampling probe, measuring instruments, data recording, and data processing are described. Also, preliminary results of aerosol particle data collected during recent flights are presented.
ELECTRICAL POWER DISTRIBUTION

The instrumentation presently installed in the aircraft requires both 28 VDC and 115 VAC-60 Hz. The aircraft 28 V bus is supplied by two, 100 amperes alternators. The bus supplies power directly to certain instruments and also to a 115 VAC - 60 Hz inverter which provides up to 1 kW output to the other instruments.

All power distribution to the various instruments is controlled via a central power control panel. The control panel is divided into three functional areas: inverter control, AC power, and DC power. Selection of the inverter, application of the inverter load, and selection of an external AC source is provided in a manner which prevents the application of inverter and external AC power simultaneously. A switch selects an external AC source which is supplied via the external AC jack at the rear of the control panel chassis, and is used for ground operations (maintenance, calibration, etc.). A pair of switches and relays apply the 28 VDC to the inverter and connect the inverter output load. The AC power is distributed to the instrumentation via a group of standard AC outlets which are protected by individual circuit breakers.

Distribution of 28 VDC power is also controlled by a group of circuit breakers. A main 15 A circuit breaker/switch feeds five branch circuit breakers which service the DC outlets for instrument power.

Metering is provided for visual monitoring of the DC input current to the inverter, AC and DC volts and total AC and DC current load. All the power to the inverter and the instruments can be disconnected by a main 60 amperes breaker/switch accessible to the pilot. All breakers can be manually operated, and are MIL and FAA approved.

THE SAMPLING PROBE

The sampling probe consists of two parts. The first part is made of stainless steel and is located on the upper lefthand side of the aircraft (Figure 1). The probe extends about 10 inches from the fuselage and has an area expansion ratio of 16 to slow the flow speed before the stream is deflected inside the aircraft. The second part is made of aluminum with five stainless steel tubes. It is connected to the first part, inside the aircraft, with a rubber hose and clamps. Detail design of the probe is shown in Figure 2.

To maintain isokinetic sampling, the 1/2 inch tube may be connected to a pump whose pumping rate can be adjusted so that the air velocity at the probe's orifice is the same as the aircraft speed. Unfortunately there is not enough data in the literature to determine the losses of particles to the inside wall of the probe as a result of the 90 degree bend of the probe. However, in view of recent wind tunnel data by Sehmel (1) we can assume that for particles less than 5 microns in diameter, the loss is less than 10 percent.
FIGURE 1. The Locations of the Various Sensors on the Aircraft.
1) Sampling Probe
3) Temperature and Relative Humidity Sensor
2) Dewpoint Sensor
4) Turbulence Sensor

FIGURE 2. The Sampling Probe
INSTRUMENTATION

The instruments mounted and to be mounted onboard the aircraft are listed in Table 1 and photographs of their locations inside the aircraft are shown in Figures 3 and 4. In the following paragraphs the instruments are described briefly.

The Condensation (Aitken) Nuclei Counter (General Electric) samples at the rate of 100 cc/second. It is capable of detecting particles larger than 0.001 micron in size by measuring the light scattered from the water droplets formed on particles which are exposed to a high supersaturation achieved by a sudden expansion. The intensity of light scattering is automatically related to the particle concentration; response time is about 2 seconds.

The Royco Particle Counter, Model PC 200A (Royco Instruments, Inc.) operates on the principle of 90° light scattering from single particle of diameter 0.32 micron and greater, with a sampling rate of 100 cc/minute. The instrument can be operated in either of two modes. In the first, it measures particle concentrations in 15 individual channels in the range of 0.32 to 8 microns and greater. In this case the data are printed in

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Parameter</th>
<th>Range</th>
<th>Accuracy</th>
<th>Flow Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>G. E. Condensation Nuclei Counter</td>
<td>Condensation (Aitken) Nuclei</td>
<td>10^10 - 10^19 particles/cm^3</td>
<td>100%</td>
<td>100 cm^3/sec</td>
</tr>
<tr>
<td>Royco Particle Counter, Model PC 200A</td>
<td>Size and concentration of particles</td>
<td>0.32 to 8 μm and 0.32 to 10^4 μm</td>
<td>10%</td>
<td>100 cm^3/min</td>
</tr>
<tr>
<td>Metrodata System, Model N8</td>
<td>Airspeed</td>
<td>50 - 350 knots</td>
<td>2 ft</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Altitude</td>
<td>5000 - 50,000 ft</td>
<td>100 ft</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Temperature</td>
<td>50 - 550 °C</td>
<td>0.5 °C</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Humidity</td>
<td>30 - 95%</td>
<td>4%</td>
<td></td>
</tr>
<tr>
<td></td>
<td>VOR 1</td>
<td>0 - 360°</td>
<td>1°</td>
<td></td>
</tr>
<tr>
<td></td>
<td>VOR 11</td>
<td>0 - 360°</td>
<td>1°</td>
<td></td>
</tr>
<tr>
<td></td>
<td>DME</td>
<td>1 - 100 n mile</td>
<td>1.5 n mile or 3%</td>
<td></td>
</tr>
<tr>
<td>NRI Universal Indicated System</td>
<td>Turbulence intensity</td>
<td>0 - 10 cm^2/s sec^-1</td>
<td>10%</td>
<td></td>
</tr>
<tr>
<td>EGCG, Cambridge Aircraft Hygrometer</td>
<td>Dew point temperature</td>
<td>-50 - -50 °C</td>
<td>20.5 °C</td>
<td></td>
</tr>
</tbody>
</table>

**Instruments to be added in the near future**

- NRI Cloud Condensation Nuclei Counter
- NRI Ice Nuclei Counter
- Quadrupole Mass Spectrometer, Scientific Instruments

**Cloud condensation nuclei:**

- 0.2 - 25 supersaturation

**Ice nuclei:**

- 0 °C - 10 °C activation temperature and 0.1 to 10^2 particles/1
Neg 720804-2

FIGURE 3. The Locations of the Instruments Inside the Aircraft.

1) Power Control Panel
2) Recorder Input Panel
3) Metredata Magnetic Tape Recorder
4) Condensation Nuclei Counter
5) Turbulence Indicator System Panel
6) Hewlett-Packard Model 500A High Speed Printer
7) Metredata System, MS, Direct Reader
8) Royco Particle Counter

a digital form on a Hewlett-Packard Model 500A High Speed Printer. In the second mode, the instrument provides a count of particles greater than a preset diameter. The data from this mode of operation are recorded on the aircraft recording system. It should be noted here that the count represents a cumulative count, but can easily be reduced to concentration in time and space.
Meteorological and aircraft parameters are measured via three different systems. The first is a Metrodata Systems, Inc., Model M8. This instrument measures temperature, humidity, airspeed, altitude, bearings from two points (VOR), distance from one point (DME), and compass heading. The temperature and humidity are obtained from sensors in a probe mounted on the nose of the aircraft (Figure 1). The temperature sensor uses a shielded, linear response, thermistor head, and the humidity uses a carbon-strip hygrister, similar to that used in radiosondes. The aircraft pitot-static system is connected
to a potentiometric transducer to measure airspeed and altitude. The VOR and DME data is derived from the navigational receivers of the aircraft. These and the altitude are used to determine the exact position of the aircraft at any time.

The second system is a Cambridge Systems Model 137-C3 Aircraft Hygrometer manufactured by EG&G for measuring dew point temperature. The sensor is mounted on the lefthand side of the aircraft nose (Figure 1). It contains a platinum thermometer.

The third system is a Universal Indicated Turbulence System, Model 1120, Meteorology Research, Inc. It measures the turbulence intensity of a specific frequency band which lies within the inertial subrange. It also measures the airspeed. The location of the sensor on the outside of the aircraft is shown in Figure 1.

All the instruments which are described above are presently onboard the aircraft. Other instruments which will be installed in the near future are:

1) A Cloud Condensation Nuclei Counter (Meteorology Research Inc.) which is capable of measuring cloud condensation nuclei concentrations at adjustable supersaturations in the range 0.2 to 2 percent. The data are measured and recorded automatically.

2) An Ice Nuclei Counter (MEE Industries) which is a fast-response counter for detecting ice nuclei in the concentration range $10^4$ nuclei per liter over any activation temperature from 0°C to -30°C with automatic recording.

3) A Quadrupole Mass Spectrometer (Scientific Instruments, Inc.) which measures the concentration of gases in the range 1 to 300 atomic mass units, with sensitivity of 0.1 ppm real-time. With appropriate molecular sieves, concentrations in the ppb range can be attained.

**DATA RECORDING SYSTEM**

A recorder input panel functions as a convenient connecting point for all signal inputs to a Metrodata Model DL620 magnetic tape data logger. Precision 10-turn potentiometers at each input terminal permit the operator to attenuate and/or calibrate each data input to a full-scale range which is appropriate to the Metrodata recorder. All data lines utilize coaxial or shielded, twisted pair cable, and, insofar as is possible, all lines and interconnections external to the various instruments are shielded to preserve data integrity.

The Metrodata recorder may be used in several input configurations ranging from ±10 millivolts to ±5 volts full scale with a 2000 point resolution. The instrumentation presently installed utilizes ±10 millivolt and ±1 volt ranges. The recorder samples 18 analog inputs (plus 2 real-time channels) at rates up to 48 channels/sec. At the minimum sampling interval, 60 minutes of recording may be accomplished on a single tape cartridge. Analog inputs are converted to digital form and recorded as four character BCD digits (sign + 3 digits). Hours, minutes and seconds
data from the real time clock are recorded at the beginning of each 20 channel scan.

A visual numeric display on the recorder permits the operator to check any one of the several input signals during acquisition and recording to verify the validity of the data.

DATA ANALYSIS

The airborne recorded magnetic tapes are brought to the Battelle-PNL computer facility for further processing and analysis. The central processor in the facility is a Systems Engineering Laboratories SEL 510A. The magnetic tape records acquired in-flight are read into the computing system via a Metrodata Model DL 622 tape reader. Presently, the tape record can be output either as a numerical record or as a graphical representation as a function of time by means of a high-speed electrostatic printer/plotter. In the event that more detailed analysis of the recorded data is required, the Metrodata tapes can be read into the 840A computer, preprocessed and/or formatted, and rewritten onto industry-compatible magnetic tape which can then be processed on a more extensive computer system such as the UNIVAC 1108.

ILLUSTRATIVE DATA

Figure 5 shows some preliminary data of the concentration of condensation (Aitken) nuclei as a function of altitude. Curve (1) was obtained

![Diagram of Concentration of Condensation Nuclei as a Function of Altitude]

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FIGURE 5. Concentration of Condensation Nuclei as a Function of Altitude
on February 20, 1972 over the Hanford project about 30 miles north of Richland, Washington. During the measurement, broken scattered clouds were present with bases at about 2000 feet. It can be seen from this curve that the concentration decreases with height to some altitude between 1000 to 2000 feet and then seems to fluctuate at higher altitudes. Other measurements were made on the same day, 25 miles southeast of Wenatchee, Washington in a clear sky (Curve 2). The count decreased with height up to about 4500 feet. At that time a patch of cloud drifted into the area, and the count increased. The next day it was overcast with some haze; cloud base was estimated at about 20,000 feet. Approximately 3 miles north of the Pasco Airport, Washington, another flight was made and Curve (3) of Figure 5 shows a plot of the collected data. Here again the count decreased with height, then remained almost constant, and finally at an elevation of about 11,500 feet, increased to values approaching the ground level concentration.

On February 21, 1972, particle size distributions were obtained at three different altitudes in the same area. These are shown in Figure 6 with the corresponding altitude marked on each distribution. Even from the meager statistics of these data, one can deduce that with increase in altitude, there is a significant decrease in the particle concentrations of all sizes.

Although the data obtained so far with this airborne laboratory do not warrant significant meteorological interpretations, we are certain that future results will be impressive.

REFERENCE

TECHNIQUES USED TO STABILIZE TRACER SOLUTIONS FOR AEROSOL GENERATION
C. W. Thomas

Methods are described to dispense tracer aerosols of several rare elements from airborne, acetone generators.

Finely divided aerosols of several rare elements are generated by acetone-burner techniques on aircraft and serve as tracers in studying precipitation scavenging rates and mechanisms. Techniques were developed to put tracers of gold, iridium, ruthenium, tellurium and selenium either in solution or in a colloidal suspension in a solvent mixture that had excellent ignition and burning characteristics. A solution of acetone containing about 20 percent by volume of n-heptane was found to be compatible with almost any tracer that was moderately soluble in acetone and/or alcohol and had ignition and burning qualities that were congruous with a forced air aerosol generator. Laboratory and field studies were conducted and the following recipes were formulated and successfully demonstrated for both ground level and aircraft releases from acetone-burner generators of the types used in AgI generators.

GOLD

One hundred and seventy-five grams HAuCl₄·3H₂O was dissolved in 13 liters of acetone in a 50 liter polyethylene carboy. Three and two-tenths liters of pyridine was added to prohibit reduction of gold by the metal surfaces of the generator. Six and five-tenths liters of n-heptane was added. This solution was stable for two weeks after which the gold in solution was slowly reduced to the metal.

IRIDIUM

Thirty-two grams of IRCl₃·XH₂O was dissolved with difficulty in 10.5 liters of acetone by the following technique: The iridium chloride salt was stirred into a liter of acetone and filtered. The filtrate was placed in a 50 liter polyethylene carboy. The residue was dried and fumed with concentrated hydrochloric acid and taken to just dryness. The residual salts were added to a liter of acetone and stirred and filtered. The filtrate was added to the polyethylene carboy. This process was repeated until no appreciable amounts of residual salts remained after filtration. Additional acetone was added to make up to 16.5 liter volume and 4.5 liters of n-heptane added. The solution was stable for weeks.
RUTHENIUM

One hundred and fifty grams of \( \text{RuCl}_3 \cdot \text{xH}_2\text{O} \) was dissolved in acetone by the method used for iridium and made up to 16.5 liters with acetone. Four and five-tenths liters of \( \text{n-heptane} \) was added. The resulting solution was colloid but stable for one week after which slow coagulation occurred over a period of several weeks.

TELLURIUM

Three hundred grams of \( \text{TeCl}_4 \) was dissolved in 16.5 liters of acetone and 4.5 liters of \( \text{n-heptane} \) added.

This solution was stable for weeks.

SELENIUM

Selenium tetrachloride was not compatible with either acetone or \( \text{n-heptane} \), both of which resulted in a rapid reduction to the metal. Alcohol was a suitable solvent for selenium, thus, 450 grams of \( \text{SeCl}_4 \) was dissolved in 4 liters of absolute ethanol. This solution was metered simultaneously with \( \text{n-heptane} \) to the burner compartment of the generator. Since this required separate reservoirs for each solution it has not been as suitable for aircraft release.

AN INEXPENSIVE INFUSION SAMPLER FOR ATMOSPHERIC TRACER STUDIES

J. M. Hales and D. W. Glover

An inexpensive infusion sampler for atmospheric tracers has been constructed. These samplers will be used for \( \text{SF}_6 \) and halogenated hydrocarbons in large scale field studies such as those being conducted on the Olympic Peninsula.

Description of plume and air mass behavior in dispersion and washout studies has necessitated the design of a simple and inexpensive infusion sampler for remote field use. Previously-used samplers are generally unsatisfactory for a variety of reasons, including expense, operational difficulties, and the frequent need for external power sources.

The sampler designed and fabricated to meet present requirements is described schematically in Figure 1. It consists of a Plexiglas cylinder (2 inch bore tubing) containing a piston (2 inch diameter Plexiglas rod) which is raised at a constant rate by a 12 volt d.c. timing motor. Power for the motor is provided by replaceable "D" cell batteries, and the unit is housed in a small plywood shelter.
Depending on the size of the drive wheels, this sampler can obtain its total sample volume of approximately 0.5 liter over periods ranging from 1 hour to 2 hours. These samplers can be deployed rapidly, simply by placing at their prescribed locations and throwing the starting switch. Sampling is terminated automatically upon filling the sampler volume by action of a top-mounted limit switch.

These samplers have been constructed primarily for use with SF₆ and halogenated organics. The samples will be injected into a gas chromatograph and analyzed using a procedure similar to that described by Clemmons et al. (1)

Cost of these samplers is about $30.00 per unit, including parts and
labor. Twenty of them have been constructed at present, with plans for several more to be built during the coming year. This large quantity of samplers will allow accurate definition of time-averaged tracer concentration over large areas, and should be particularly useful in large-scale dispersion and washout studies such as those currently being conducted on the Olympic Peninsula.

REFERENCE


ANALYTICAL PROCEDURES FOR MEASUREMENT OF $^{214}$Pb AND $^{214}$Bi IN RAINWATER

C. W. Thomas, J. A. Young, and N. A. Wogman

A technique is described for rapid continuous separation of $^{214}$Pb and $^{214}$Bi from rainwater at flows up to 30 liters per minute.

The short-lived daughters of radon, $^{214}$Pb (26.8 min) and $^{214}$Bi (19.7 min), are potentially useful tracers of precipitation scavenging processes because they are attached to the natural aerosols, they are present in easily measurable concentration in rainwater, and their half-lives are of the same order of magnitude as the time scale on which these processes occur. The major problem in using the radon daughters as tracers is the difficulty of rapidly separating them from large volumes of rainwater after short consecutive collection periods. A technique was recently developed to continuously separate $^{214}$Pb and $^{214}$Bi from rainwater at flow rates of up to 30 liters per minute and consecutive sampling periods of 5-10 minutes. Rainwater from a 3000 ft$^2$ polyethylene surface is collected in a polyethylene 55-gallon drum and pumped through a fiber-glass filter followed by an anion exchange bed (0.64 cm thick by 28 cm diameter of Dowex 500W × 8 H$^+$ form, 200-400 mesh). Bismuth-214 is removed at greater than
90 percent efficiency by the anion exchange bed. Bismuth-214 is measured immediately by counting the coincident photons emitted by the sample with a pair of 13-1/2 inch diameter by 6 inch thick NaI(Tl) detectors. No measurable amount of 214Pb is removed by the anion exchange bed, however, greater than 90 percent of the 214Pb is retained on the cation exchange bed which in turn does not retain 214Bi. After the collection period the cation bed is set aside for about 1 hour to allow ingrowth of 214Bi which is then measured using the above detector system and the 214Pb concentration is calculated from this growth.

FALLOUT RATES AND MECHANISMS

R. W. Perkins

The concentrations of trace elements, cosmogenic radionuclides, and nuclear-weapons-produced radionuclides were measured in the air from ground level to 19 km and in rain and seawater in order to determine the origin and chemistry of the atmospheric aerosol and to study the rates of atmospheric and oceanic mixing, the rates of air-sea interchange, and the rates and mechanisms of dry and wet fallout. Brief reports on these topics are presented by the principal investigators.

AIR CONCENTRATIONS OF 90Sr AND 55Fe AT RICHLAND, WASHINGTON FROM 1963 TO 1970 - C. W. THOMAS AND J. C. LANGFORD

The atmospheric concentrations of 90Sr and 55Fe were measured in ground level air samples collected at Richland from 1963 to 1970 as part of a program to define the rates of long-term stratospheric processes in the northern hemisphere. Seasonal variations in the concentrations of 90Sr were similar to those radionuclides of stratospheric origin, decreasing from 1963 to 1966 as a result of decay and deposition on the earth's surface. Atmospheric nuclear bomb testing since 1967 have maintained the surface air 90Sr concentrations at about the same level from 1967 to 1970 (Figure 1). In contrast, the seasonal variation of the concentrations of 55Fe were not so pronounced as radionuclides of stratospheric origin, (Figure 2), and suggest
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FIGURE 1. The Concentration of $^{90}$Sr in Ground Level Air at Richland, Washington from 1963-1970

Neg 715669-2

FIGURE 2. The Concentration of $^{55}$Fe in Ground Level Air at Richland, Washington from 1964-1968
that a portion of the $^{55}$Fe may come from the Hanford Project.

**COMPARISONS OF ATMOSPHERIC RADIONUCLIDE CONCENTRATIONS AT NEAH BAY AND RICHLAND, WASHINGTON - C. W. THOMAS**

The atmospheric concentrations of several radionuclides were measured continuously in ground level air from 1968 through 1970 at Richland (46 °N, 118 °W) and west of Neah Bay, Washington on the Makah Indian Reservation (49 °N, 125 °W) near the Pacific coast. At both sites, large 350 cfm air pumps are mounted several feet above the ground and draw air through membrane filters. The radionuclide concentrations on the filters are measured using large volume NaI(Tl) gamma ray spectrometers.

As seen in Figure 3, the concentrations of $^{7}$Be, $^{95}$Zr, $^{106}$Ru, $^{137}$Cs, and $^{144}$Ce at the two sites show the seasonal variation that is characteristic of radionuclides of stratospheric origin. However, the concentrations are generally higher in the Richland samples, especially during the summer when the concentrations are at a maximum. A possible explanation for this effect is increased vertical mixing following passage of air over the coastal and Cascade Mountain Ranges.

**COSMOGENIC RADIONUCLIDE PRODUCTION RATES IN ARGON FROM 3 TO 19 km - J. A. YOUNG, C. W. THOMAS, AND N. A. WOGMAN**

For the past four years the concentrations of the cosmogenic radionuclides $^{24}$Na (19 hr), $^{36}$Cl (57 min), and $^{39}$Cl (55 min) have been measured in air filter samples collected by aircraft. These radionuclides are produced at a relatively constant rate in the atmosphere by spallation reaction of cosmic-rays with atmospheric argon. In order to determine whether the measured cosmogenic radionuclide disintegration rates corresponded to the actual production rates, metal spheres were lined with rubber meteorological balloons, filled with argon under pressure, and flown at altitudes ranging from 3 to 19 km. At the end of each flight the argon was vented through a charcoal impregnated filter. The filters and rubber liners were then composited and the cosmogenic radionuclide concentrations measured by direct analysis on a NaI(Tl) multi-dimensional gamma-ray spectrometer. In addition to $^{24}$Na, $^{36}$Cl, and $^{39}$Cl, the cosmogenic radionuclides $^{18}$F, $^{20}$Mg, $^{34}$mCl, and $^{38}$S were measured on some of the 15 to 19 km flights.

The measured $^{24}$Na production rates were almost identical to the measured atmospheric disintegration rates, indicating that the air filters are removing essentially all of the $^{24}$Na from the air. The $^{36}$Cl and $^{39}$Cl production rates, however, were much higher than their measured atmospheric disintegration rates. Possible explanations for this include that the lifetimes of the $^{36}$Cl and $^{39}$Cl nuclei are too short for them to become quantitatively attached to filterable atmospheric aerosol particles, or that their production rate is enhanced in the argon spheres by low energy radiation produced by the reaction of cosmic rays with the aircraft and the metal spheres. Higher energies are
required for $^{24}$Na production, so the $^{24}$Na production rate would not necessarily be enhanced by the latter effect.

On March 11, 1971, several spheres were filled with a total of 60 M$^3$ (STP) of argon and flown for 350 minutes at 15.3 km. The largest amount of argon that had been flown on any previous flight was 42 M$^3$. These measurements permitted a significant improvement in the accuracy of cosmogenic radionuclide production rates; however, an attempt to measure $^{32}$P (14.5 d) and $^{33}$P (25 d) was unsuccessful even with very nearly quantitative chemical separations. Production rate measurement of $^{32}$P and $^{33}$P will require a long exposure of the argon-filled spheres at some high altitude location.

**VERTICAL PROFILES OF RADIONUCLIDE CONCENTRATIONS IN THE ATMOSPHERE**

J. A. YOUNG AND N. A. WOGMAN

Vertical profiles of atmospheric radionuclide concentrations have been determined on a monthly basis since 1967 from air filter samples collected by RB-57 aircraft at 1.5 to 3.0 km altitude intervals from 0.3 km to an
altitude of over 18 km. In the past, most of the vertical profiles were measured either south of Spokane, Washington at 47°N, 117°W, or east of Barbados, British West Indies from 12 to 18°N and 45 to 59°W. The profiles at Barbados were taken during the summer of 1969 as part of the BOMEX project. The concentrations of cosmogenic radionuclides, nuclear weapons produced radionuclides, and radon and thoron daughters were determined using NaI(Tl), multidimensional gamma ray spectrometers and, beginning in late 1971, a dual, anticoincidence shielded, Ge(Li) gamma-ray spectrometer.

A surprising feature of the profiles at all latitudes has been the fairly common occurrence of very pronounced concentration minimums at altitudes from 6 to 9 km, for all commonly measured radionuclides, except 24Na (15 hr). Sometimes the concentrations at these altitudes are orders of magnitude lower than the concentrations at lower elevations; and the presence of 24Na at normal concentrations in the samples virtually eliminates the possibility of sampling error. Whatever the cause of this phenomenon (in-cloud precipitation scavenging being a likely candidate) it is apparent because 24Na is relatively insensitive to it, that it occurs on a time scale large compared with the 15 hr half-life of 24Na.

A specific example of a few of the radionuclide profiles is shown in Figure 4. The profiles were obtained

FIGURE 4. Radionuclide Disintegration Rates on November 16, 1971 at 39°N, 103°W
THE DETERMINATION OF AIR-SEA EXCHANGE AND OCEANIC MIXING RATES USING $^7$Be DURING THE BOMEX EXPERIMENT - J. A. YOUNG AND W. B. SILKER

The concentrations of $^7$Be and other radionuclides were measured in air, rain, and seawater in a region east of Barbados, British West Indies in the summers of 1968 and 1969 during the BOMEX experiment. The deposition of $^7$Be on the sea surface by rainfall in August 1968 and May through July 1969 was only 13 and 26 percent, respectively, of that necessary to maintain the measured seawater inventories. The calculated deposition velocity of $^7$Be on the sea surface by wet and dry deposition was 1.0 cm sec$^{-1}$. During May through June of 1969 the calculated flux of $^7$Be across the sea surface averaged about 3.8 $\times$ 10$^{-2}$ atom cm$^{-2}$ sec$^{-1}$ and the inventories were increasing at a rate of 1.0 $\times$ 10$^{-2}$ atom cm$^{-2}$ sec$^{-1}$. Due to seasonal variations in the atmospheric $^7$Be concentrations the $^7$Be seawater inventories should vary seasonally with maximum inventories occurring around July and minimum inventories around February. The yearly average of the flux of $^7$Be across the sea surface was calculated to be 1.6 $\times$ 10$^{-2}$ atom cm$^{-2}$ sec$^{-1}$. The vertical eddy diffusion coefficient, $K_z$, in the sea decreased from greater than 7 cm$^2$ sec$^{-1}$ at the surface to 0.25 to 0.85 cm$^2$ sec$^{-1}$ at 30 to 40 meters and then possibly increased somewhat from 40 to 100 meters. The $^7$Be inventory in the sea varied rapidly due to the horizontal motion of seawater through the sampling region. The $^{103}$Ru to $^{95}$Zr ratio in the top 15 meters of the sea indicated that large amounts of $^{103}$Ru and $^{95}$Zr from the French nuclear test series at 23 $^\circ$S beginning July 7, 1968 entered the sampling region on August 15, 1968 and also was present in June, but not in May 1969. The debris possibly was carried into the sampling region by the Guiana Current which flows northwestward along the coast of South America.

ATMOSPHERIC FALLOUT DURING 1971 - C. W. THOMAS AND J. A YOUNG

The atmospheric concentrations of 25 radionuclides in ground level air were measured continuously during 1971 at Point Barrow, Alaska (71 $^\circ$N) and Richland, Washington (46 $^\circ$N) by filtering large volumes of air through membrane filters followed by gamma-ray spectrometric analysis. The concentration of nuclear-weapons-produced radionuclides, including fission products and neutron activation products, increased steadily from 1967 through 1971 because of the high yield thermonuclear tests conducted by the Chinese at Lop Nor (45 $^\circ$N) during this period with a total yield of over ten megatons. During 1971 essentially all of the nuclear-weapons-produced radionuclides present
in northern hemispheric air originated from these Chinese tests. The concentration of $^{88}$Y (107 day half-life) increased 100-fold over 1968 peak levels and during 1971 reached peak concentration levels comparable with peak concentrations of 1962-1963.

The Chinese nuclear test of November 19, 1971 was detected in low concentrations in ground level air at Richland, Washington five days after detonation. During 1970 the nuclear reactors, which are a part of the Hanford atomic energy complex and use Columbia River water as a straight through coolant, were shut down. The effect of this shutdown was noted by a decrease in air concentration of several radionuclides that were known to be associated with the Richland operations. Scandium-46 concentrations in air were down by two orders of magnitude, $^{60}$Co was down one order of magnitude, and $^{124}$Sb is now below detection levels. Also affected was $^{65}$Zn air concentration which now shows an excellent seasonal variation and is down by an order of magnitude.

A COMPARISON OF MEASURED AND PREDICTED EXPOSURES FROM ELEVATED SOURCES

R. K. Woodruff and C. E. Elderkin

A comparison was made of data from elevated continuous source diffusion experiments. The experiments were conducted in neutral and stable conditions and the data have been classified into stability classes by both the standard deviation of the azimuth angle, $C_b$, and the Richardson number, $R_i$. As is common in safety and environmental impact analyses, for comparison with Pasquill estimates. The comparison indicates that estimates based upon Pasquill's curves are conservative downwind of the maximum in the ground level centerline exposure, and unconservative at short distances. This result indicates that the elevated plumes are growing faster than Pasquill's curves indicate, the latter being based primarily upon surface release experiments. The classification of the data by Richardson number gives a better stratification of the data than the $C_b$ classification, although the scatter in the data indicates that the stability stratification may not be significant. It is anticipated that further analysis using the Richardson number to determine $C_b$, and $C_b$ to determine $C_{eq}$ will produce a better organization of the data.

The increasing importance of the environmental effects of industrial emissions is resulting in greater demands upon diffusion model accuracy.
and applicability. In nuclear power plant safety and environmental impact analyses, it is common to use the plume growth curves attributed to Pasquill. These curves are primarily based upon data obtained from surface releases to distances of about a kilometer. However, Pasquill's curves are commonly extrapolated to distances as great as 100 kilometers and applied to elevated sources. These extensions of Pasquill's curves have been generally supported by subsequent data as being conservative under the neutral and stable conditions, which are generally of most concern, at distances of less than about 10 kilometers as shown in Meteorology and Atomic Energy, 1968(1) (Figures 4.4 and 4.6). The slope of the stable data in Figure 4.6 suggests, however, that concentrations beyond about 10 kilometers might exceed those predicted by the extrapolation of Pasquill's results.

Additional elevated source experiments with particulate zinc sulfide and fluorescein dye have been conducted in the last few years at several elevations at Hanford. Figures 1 and 2 summarize the normalized ground level centerline exposure data.

![Normalized Centerline Exposure Classified into Pasquill Categories by Richardson Number.](Neg 721420-7)
EUT/Q, for 57 tests released at an elevation of 26 meters under predominantly neutral and stable conditions. The tests are grouped in Pasquill categories by the Richardson number and the standard deviation of the wind direction, $\sigma_B$, in Figures 1 and 2, respectively, according to the following classifications.

<table>
<thead>
<tr>
<th>Stability Class</th>
<th>$z_1$</th>
<th>$\sigma_B$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Extremely Unstable</td>
<td>-1.0</td>
<td>0.7 25.0 Degrees</td>
</tr>
<tr>
<td>Moderately Unstable</td>
<td>-0.5</td>
<td>-0.4 20.0</td>
</tr>
<tr>
<td>Slightly Unstable</td>
<td>-0.17</td>
<td>-0.15 15.0</td>
</tr>
<tr>
<td>Neutral</td>
<td>0</td>
<td>10.0</td>
</tr>
<tr>
<td>Slightly Stable</td>
<td>+0.83</td>
<td>0.65 5.0</td>
</tr>
<tr>
<td>Moderately Stable</td>
<td>+0.87</td>
<td>0.31 2.5</td>
</tr>
<tr>
<td>Very Stable</td>
<td>+0.11</td>
<td>1.1</td>
</tr>
</tbody>
</table>

As these classifications are discontinuous, tests falling between classes were divided at the midpoints between classes. The Richardson numbers were determined for the layer from 7 to 50 ft for the period of the release. The $\sigma_B$'s were determined from 20-second averages of the wind direction and were calculated over the release period, which was 30 minutes with only a few exceptions.

Figures 1 and 2 also contain tables of the number of observations averaged for each plotted point. The smooth curves are the ground level centerline exposures predicted using Pasquill's curves for stability classes A, D, and F as indicated by the circled letters.
COMPARISON OF RESULTS

The most obvious difference between the data and the predictions is the conservatism of the Pasquill prediction near the peak and beyond, especially for the neutral and stable cases. This feature is consistent with the results of the experiments summarized in Figures 4.4 and 4.6 of Reference (1). However, the slope of a smoothed curve through the data would not suggest that concentrations at greater distances would exceed those which would be predicted by Pasquill's curves. The conservatism of estimates based on Pasquill's curves is not fully shown by these data since sampling errors due to subisokinetic flow rates tend to make the observed exposures high. Qualitative observations indicate that these errors can be as high as a factor of two. No conclusions can be drawn from the few unstable cases, although those that are shown do not disagree significantly with the predicted.

Another significant feature of the data is the consistently higher concentrations in the region between the source and a point downwind occurring just before the maximum centerline exposure for the neutral and stable cases. The magnitudes of the differences are quite significant although the downwind distance to which the differences are great is relatively short; e.g., in both Figures 1 and 2 the observed exposures fall below the predicted at a distance of only 700 meters. The Brookhaven oil fog results in Figure 4.4, of Reference (1), show a similar tendency. It is apparent that a translation along the x axis would significantly improve the fit of the data to the Gaussian form. This could be accomplished by a translation of Pasquill's $\sigma_y$ and $\sigma_z$ curves to higher values. In other words, the Gaussian model for elevated releases is reasonably verified by the data although the rates of plume growth for the elevated sources are greater than those obtained from Pasquill's curves for neutral and stable cases.

Another feature of importance is that neither Ri number nor $\sigma_0$ classification of the data provide very good stratification of the data for neutral and stable conditions. Although there is a stronger tendency for correlation with $\sigma_0$, the scatter of data about each data point (not plotted) would likely make it difficult to demonstrate that the neutral and stable classifications are significantly different. This is important because frequently in safety and impact studies considerable significance is placed upon stability class frequency distributions. Also of interest is the fact that the trend towards decreasing dilution with increasing Ri reverses for large Ri as indicated by the relative positions of the curve designated as $G$ to those of less stable classes. This tendency may be the result of increasing low frequency plume meander in addition to plume spreading due to vertical shear in the wind direction with increasing stability.

The above results suggest that a better stratification of the data might be obtained if the $\sigma_0$ number
were used to classify the vertical diffusion and $\sigma_z$ to the lateral diffusion. As $Ri$ and $\sigma_z$ are virtually independent, there would be $(7)^2$ possible classifications. As such a large number of classes would be meaningless, it would be desirable to establish a meaningful number of diffusion curves, each representing a group of different stability class pairs. This type of tabulation has not been completed at this time.

Crosswind integrated exposures classified with $Ri$ would also be expected to show better organization than if classified by $\sigma_z$ since the lateral variance does not appear in the Gaussian model for crosswind integrated exposure. This is also true of the form of the Gaussian model used for long term exposure calculations. As these forms of the Gaussian model are used in safety and environmental impact analyses, it will be important to demonstrate in additional analyses the relative merits of stability classification based upon $\sigma_z$ and $Ri$.

REFERENCES


Comparison of Maximum Exposures at Ground Level Resulting from Simultaneous Release of Tracers from Two Elevations

P. W. Nickola

Results from field experiments in which two atmospheric tracers were simultaneously released from different elevations are used to compute the ratio of exposures resulting from the two levels of release. The magnitude of the ratios is generally related quite strongly to atmospheric stability. These experimentally generated ratios are compared to similar ratios calculated with the aid of Pasquill's curves for various diffusion categories. The experimental data show considerably smaller benefits from additional stack height than do the data resulting from use of Pasquill's curves.

In the previous annual report (1) in this series, Nickola reviewed the technique employed at Pacific Northwest Laboratory in which two tracers...
are simultaneously released from two elevations on a tower and subsequently collected on a common set of filters for assay. Nickola employed results of these dual tracer releases in investigating the effect of release height on plume centerline exposures as measured at ground or breathing level (≈2 m). Specifically, the effect of increasing release height from 2 meters to 26 meters was investigated. It is the purpose here to report results of comparisons of other release height differences, and to compare these experimental results with predictions which can be made with the aid of the commonly used Pasquill diffusion categories.

In comparing the exposures resulting from the one-half hour tracer releases, the data have been normalized to the mass of tracer emitted, but not to wind speed.

The maximum exposures measured at distances as far as 12.8 km from the source were put in the form of a ratio with the exposure resulting from the lower release elevation as the denominator. Thus, the benefit to persons located at plume centerlines is directly indicated. A ratio of 1.0 indicates no benefit from an increased elevation of release. A ratio of 0.5 indicates that release from the upper level (H_U) reduces the exposure to one half that which is observed from release at the lower elevation (H_L).

As was done in the previous study, the data were grouped into rather arbitrary stability classes on the basis of Richardson numbers (Ri) computed for the layer between 2 m and 15 m. The classes were moderately stable (MS), slightly stable (SS), slightly unstable (SU), and moderately unstable (MU) with corresponding Ri ranges of >0.10, 0.10 to 0, 0 to -0.10, and <-0.10.

The data reported in the previous annual report for H_U = 26 m and H_L = 2 m are shown by the solid curves on Figure 1. (There is an anomaly in these data above the point where the MS curve exceeds the ratio for the SS curve. Since only 3 field tests contributed to the MS curve, it is felt the SS curve is more reliable for distances beyond 2 km.) The solid curves on Figures 2, 3, and 4 indicate exposure ratios observed during other combinations of release heights and stabilities. Only SS and MS stability categories were observed during the 18 dual field releases not previously reported. Note in Figure 3 that the observed differences in ratio for the SS and MS data for H_L = 26 m and H_U = 56 m were so small that a single curve was plotted as representative.

It should be mentioned that the pairings of release elevations actually used in the field experiments were 2 and 26 m, 26 and 56 m, and 56 and 111 m. Exposure ratios for the other pairings of H_L and H_U can easily be computed from ratios of these de facto pairings.

Example interpretations from these curves can be made from Figure 4. In a slightly stable atmosphere, release at an effective elevation of 111 m would reduce plume centerline exposure at a distance of 2 km to about 32% of the exposure which would result from release at an elevation of 56 m. At
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**FIGURE 1. Centerline Exposure Ratios for \( H_L = 2 \) m and \( H_U = 26 \) m for Various Atmospheric Stabilities**

Neg 721169-2

**FIGURE 3. Centerline Exposure Ratios for \( H_L = 26 \) m for Various Values of Stability and \( H_U \)**

Neg 721169-4

**FIGURE 2. Centerline Exposure Ratios for \( H_L = 2 \) m for Various Values of Stability and \( H_U \)**

10 km, there would still be a 50% reduction. In moderately stable atmospheres, the reductions at 2 and 10 km resulting from the increased effective stack height would be to 8% and 32% of the exposures from the lower stack. It is of interest to compare the curves developed above with predictions that can be generated with the
The differences between Pasquill's graphed data for heights of 30 m and 100 m and the PNL release heights of 26 m and 111 m were ignored in the comparisons that follow. In fact, there is considerable room for debate as to the wind which should be used as representative in the normalization of exposure. The avoidance of the necessity of making these \( \overline{u} \) selections is part of the appeal of the exposure ratio approach presented in this paper.

In Figure 1, the dotted curves indicate ratios of exposure for \( H_y = 30 \) m and \( H_y = 2 \) m under the SS winds mentioned above. The dashed curves were generated assuming the MS wind speeds mentioned. The nomenclature D, E, and F are Pasquill's neutral, slightly stable, and moderately stable conditions, respectively. One might conclude that the discrepancy between Pasquill's curves and the experimental SS and MS data is not out of bounds at distances out to about 2 km, but beyond that distance, Pasquill's curves significantly overestimate the benefits expected from the increase in elevation of release.

For \( H_y \) other than 26 m, the assumption of either an SS or MS wind profile made minimal difference in the computed centerline ratios developed using Pasquill's curves. Thus only one set of Pasquill-generated curves (represented by dot-dash lines) is presented in Figures 2 and 3.

In Figures 2 and 3, the tendency for significant overestimation of the benefits of additional stack height is repeated. Although it should be

aid of Pasquill's frequently used diffusion categories. Curves representative of these categories are given in Appendix A-3 of Meteorology and Atomic Energy, 1968. Pasquill's curves give centerline exposure \( (E_{dp}) \) normalized to mean wind speed \( \overline{u} \) and source strength \( Q \) in the form \( E_{dp} \overline{u}/Q \) versus distance from source. In order to employ these curves in the current study, it is necessary to assume a "typical" wind speed for the elevations under consideration. An examination of the winds measured on the 122-m meteorology tower during tracer releases suggested \( \overline{u} \) values of 2.6, 4.4, and 7.0 mps at elevations of 2 m, 30 m, and 100 m, respectively, during SS conditions. Corresponding values of \( \overline{u} \) during MS conditions were 1.2, 3.2, and 5.1 mps.
noted that the definitions of "slightly stable" and "moderately stable"
offered by Pasquill and by the current study likely embrace quite different
ranges in stability, it is felt that the discrepancies between the observed
ratios and those which can be pre-
dicted by use of Pasquill’s diffusion
categories are cause for caution in
the application of Pasquill’s curves.

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A $^{85}$Kr FIELD MONITORING SYSTEM AND ITS APPLICATION TO MAXIMUM
AIR DOSE MEASUREMENTS OF A RADIOACTIVE GAS

J. D. Ludwick and P. W. Nickola

Improvements to the $^{85}$Kr diffusion grid are described which
include an increase in the number of detectors, an expansion of
the programmer memory, and an increase in data transfer speed.

Our $^{85}$Kr field monitoring system
has been expanded by doubling the num-
ber of detector stations from 64 to
128 positions and by installing auto-
matic cover-adsorber plates on each
detector for energy discrimination.

An improved plume definition was
sought by selective positioning of
the added sensors although little, if
any, improvement in sampling density
was accomplished by the additional 64
stations. Monitoring stations were
extended to the 1600 meter arc on the
meteorological grid. Definition of
the upper extent of typical plumes
will be accomplished through measure-
ments now available from sensors lo-
cated up to 70 meters above ground
elevation.
Modifications of the data collection center include the expansion of the programmer memory to identify information from 256 separate locations rather than the previous 64. In this way the instrumentation is ready for any further expansion of the field locations beyond the existing 128. The real-time monitoring information collected on two 4096 address memory banks is also handled in a new manner. An electronic coupler was built to facilitate transfer of the 4096 information pieces to DEC-tape transports such as used in PDP-15 data processing computers.

There are several important advantages of this new system. The rapid information transfer ability of the coupler-tape system conserves real-time monitoring time, allowing continuous measurements of plume characteristics in time increments as small as 0.1 second. In addition, the computer compatible nature of the new tape system allows data readout in a form designed to significantly reduce the many man-hours previously needed for necessary information presentation.

PRELIMINARY CONCENTRATION MEASUREMENTS
OF PUFFS RELEASED DURING NEAR CALM WINDS
P. W. Nickola and J. D. Ludwick

The Hanford inert gas field grid has been revised in order to better measure puffs of $^{85}$Kr released under stagnant atmospheric conditions. To date, two puffs have been released on this grid. Results from these tests suggest that if tracer concentration is considered as a function of distance, the stagnation conditions produce lower concentrations than do releases into stable atmospheres with steady wind directions. In considering short period (5 sec) mean concentrations as a function of time, stagnation tests produced higher concentrations than the steady wind case at times greater than 500 seconds after release.

Field diffusion experiments employing the inert radioactive gas $^{85}$Kr were first carried out at Hanford during the fall of 1967. Numerous references which describe the system, present the data generated and describe research generated from use of this system were listed in last year’s annual report in this series. Further research involving data generated by this system is presented in this and other contributions elsewhere in
this annual report, or is referenced in the section on Publications and Presentations at the end of this volume.

The early krypton field releases, involving both puffs and plumes, were carried out during periods when predicted steady winds were expected to carry the krypton tracer through two reasonably dense arrays of samplers located at distances of 200 and 800 m from the release point. In July 1971, the first tracer release was made on a revised grid designed to investigate diffusion under extremely low wind speed or stagnation conditions. This grid, incorporating a number of field samplers between the previously established 200 m and 800 m arrays, is diagrammed on Figure 1. Each dot represents a Geiger-Müller field detector located at an elevation of 1.5 m. Tracer release in these erratic wind direction situations was near the center of the array.

To date, two puffs of krypton tracer (puffs P8 and P9) have been released at ground level on the stagnation grid. Figure 1 diagrams for test P9 the extent of detectable concentrations of the tracer at several moments in time. It is interesting to note that although the tracer is still detectable at the source location after 14 minutes, tracer also was detected at a distance greater than 1.2 km from

![Figure 1](image-url)

**FIGURE 1.** Areal Extent of Detectable $^{85}$Kr Concentration at 1.5 m Elevation During Field Test P9, 30 July 1971. Puff release at 0451 PST.
the source. This indicates that despite the tracer being at ambient temperature at release (i.e., nonbuoyant) a portion of it must have moved at a mean speed of greater than 3 mph. As indicated on Figure 1, the mean wind speeds were 0.7 mph and 2 mph at the 6-ft and 50-ft elevations, respectively, and the atmosphere was quite stable thermally. It is surprising that a portion of the tracer moved at a speed characterized by winds at an elevation above 50 ft.

Puff P10, released from the same location but on a different day, drifted in a direction opposite the movement of puff P9. Thus, no data are available for distances beyond 0.5 km. Winds averaged 1.8 mph at the 7-ft elevation during this test. A 6°F temperature inversion existed between the 3 and 50-ft elevations.

Figure 2 compares maximum short period (5-sec) mean concentrations \(\chi_p\) for all 10 puff releases to date. The \(\chi_p\) values were normalized to unit source strength by dividing by the amount of tracer emitted \(Q\). Puffs P1 through P8 were those released at ground level during more steady wind directions. Puffs P1, P2 and P8 were released into stable atmospheres; P3, P4, P5 and P6 into neutral atmospheres; and P7 was released during unstable atmospheric conditions. As mentioned previously, stagnation puffs P9 and P10 were released into stable atmospheres.

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**FIGURE 2.** Puff Maximum Short Period (5-sec) Concentrations Versus Distance and Versus Time
One might intuitively expect concentrations under stagnation conditions to be higher than for those conditions in which higher wind speeds are observed. The right-hand portion of Figure 2, in which $x_p/Q$ is plotted as a function of time after release, confirms the intuitive feeling. However, the left-hand portion of this figure indicates that, in terms of distance, the stagnation $x_p$ values for tests P9 and P10 rank relatively low.

One might also speculate that the maximum exposure $(E_p/Q)$—the maximum quantity of tracer a stationary receptor could experience—would be higher in near-calm conditions. Figure 3 suggests that this is not the case. Exposures experienced during the relatively steady winds during stable tests P2 and P8 were at least as great as those observed during the two stagnation tests. (Wind speed during P2 was 2.7 mph and during P8 was 3.4 mph at the 1.5 m elevation.) No normalization to wind speed is incorporated in the exposures presented in Figure 2.

A qualitative explanation as to why P2 exposures exceeded those from the stagnant puffs P9 and P10 can be advanced. Higher wind speeds lead to lower exposures. Variable wind directions lead to lower exposures. In the case of test P2, the effect of steadiness of direction may have outweighed the effect of the higher wind speed. Alternatively, it is possible that the relatively wide spacing between sensors during P9 and P10 precluded the obtaining of measurements near locations of peak exposure. For like reasons, the $x_p/Q$ values reported in Figure 2 for tests P9 and P10 may be low. During tests P1 to P8 when steady winds carried tracer through the relatively dense network of samplers, it is unlikely that the measured maxima differed greatly from the actual maxima.

Further releases of puffs in stagnant atmospheres are planned in order to better define diffusion in this relatively uninvestigated area.

REFERENCES

A COMPARISON OF EXPOSURES RESULTING FROM INERT GAS
RELEASES WITH EXPOSURES PREDICTED WITH
THE AID OF PASQUILL'S DIFFUSION CATEGORIES
P. W. Nickola

Normalized exposures resulting from near ground level releases and sampling of plumes of an inert gas tracer were lower than exposures predicted with the aid of graphs based on Pasquill's diffusion categories. Since concentration reducing processes other than turbulence and diffusion should be minimized by use of an inert tracer, these results suggest that Pasquill's curves are too conservative. Exposures resulting from instantaneous puff releases were found to average three times the magnitude of those resulting from 10 to 20-minute plume releases.

Pasquill's diffusion categories and associated curves of exposure versus travel distance are relatively widely used in estimating plume centerline exposures. A number of these graphical aids are given in Appendix A-3 of Meteorology and Atomic Energy, 1968. (1) Measurements of exposure made during a series of releases of the inert gas 85Kr(2) present an opportunity to compare these observed exposures with those resulting from use of Pasquill's curves.

For a given meteorological condition, the use of an inert gas tracer should result in higher downwind concentrations than would be expected with particulate or chemically reactive pollutants. The effect of depleting mechanisms such as fallout, chemical reaction with other atmospheric constituents and impaction on vegetation or other terrain features are absent.

Table 1 lists the duration of release and diffusion measurements made during the period of transport of the tracer across the measurement arcs. These arcs were located at distances of 200 and 800 m from the sources. It is assumed that since tracer release height was 1 m and sampling height was 1.5 m, the measured concentrations were near plume axial values.

The left-hand portion of Figure 1 gives plume centerline exposures (E_p) normalized to amount of tracer released (Q) and to wind speed (U) measured at an elevation of 1.5 m. The dotted lines correspond to Pasquill's diffusion categories qualitatively described as extremely unstable (A) through neutral (D) to moderately stable (F). Note that only the very
stable test C1 falls above Pasquill's D or neutral classification. Pasquill's test number indicates that an instantaneous puff of krypton gas was released shortly before the plume release of tracer. An asterisk following

TABLE 1. Krypton-85 Tracer Releases

<table>
<thead>
<tr>
<th>Test</th>
<th>Duration (min: sec)</th>
<th>Wind Speed at 1.5 m (mps)</th>
<th>Richardson No. for Layer 2 to 15 m</th>
<th>Qualitative Thermal Stability</th>
</tr>
</thead>
<tbody>
<tr>
<td>*C1</td>
<td>15:28</td>
<td>1.3</td>
<td>0.13</td>
<td>Very Stable</td>
</tr>
<tr>
<td><em>C2</em></td>
<td>15:05</td>
<td>3.9</td>
<td>-0.04</td>
<td>Neutral</td>
</tr>
<tr>
<td><em>C3</em></td>
<td>14:15</td>
<td>7.1</td>
<td>-0.03</td>
<td>Neutral</td>
</tr>
<tr>
<td>*C4</td>
<td>9:58</td>
<td>3.9</td>
<td>-0.16</td>
<td>Unstable</td>
</tr>
<tr>
<td>C5*</td>
<td>19:51</td>
<td>2.6</td>
<td>0.07</td>
<td>Stable</td>
</tr>
</tbody>
</table>

FIGURE 1. Plume and Puff Peak Exposures Versus Distance from Source

In Table 1, an asterisk preceding a test number indicates that an instantaneous puff of krypton gas was released shortly before the plume release of tracer.
a test number indicates a puff release shortly after plume release. (Of course, tracer from one release was permitted to clear the sampling grid prior to the following release.) Meteorological conditions were reasonably constant throughout each pairing of releases. Peak normalized exposures resulting from the puff releases are shown at the right of Figure 1. It is interesting to note that although Pasquill's curves were designed for use with continuous releases in this instance, they more reasonably predict exposures for the puff releases.

Quite aside from the comparison of the krypton releases with Pasquill's curves, the sequential releases of puffs and plumes provide the opportunity to compare the magnitudes of puff and plume exposures. Ratios of $E_p/U/Q$ for each puff to $E_p/U/Q$ for its accompanying plume were computed. Although individual ratios ranged from a high of 6.4 to a low of 1.5, the geometric mean ratio was 3.0 for samples at both 200 m and 800 m arcs. Ratios of peak exposure in which no normalization to $U$ was effected were also computed. These $E_p/U/Q$ ratios for 200 m and 800 m again, fortuitously, were identical at 3.1. No dependence on atmospheric stability was obvious in either case.

REFERENCES


EXAMINATION OF THE RELATIONSHIP BETWEEN THE SIZE
AND SHAPE OF THE REAL PLUME AND ITS POSITION

J. V. Ramsdell

The assumption that the size and shape of a real (instantaneous) plume are independent of the position of its centroid at any fixed distance from the source is crucial to fluctuating plume models. This assumption is examined using 85Kr data and time series analysis.

The discussions on the relationship between the real (instantaneous) plume and the apparent (mean) plume which have been presented in past volumes of this series(1,2) have tacitly assumed that the diffusion and meandering are statistically independent processes. This assumption was explicitly identified in a recent paper by Högström.(5) Specifically, the assumption required is that the shape of the real plume and its physical size must be independent of the instantaneous horizontal or vertical position of its centroid. Elementary time series analysis provides an easy means of checking the validity of this assumption.

One of the short-duration continuous releases of 85Kr made in 1967(4) provides sufficient data for an initial analysis. The time-history of this plume at both 200 and 800 m is shown in Figure 1. The release, CS, was made during slightly stable conditions with a mean wind speed of 2.6 m/sec. As can be inferred from the figure, there was a dominant low frequency meander of the wind direction which had a period of oscillation of about 30 min. It might be expected that this relatively organized meander would enhance or exaggerate any relationships between the size and shape of the real plume and its position. If this is the case, the plume will provide a good test of the assumption in question.

The size and shape of the real plume were parameterized by taking the first 4 moments of the crosswind concentration distribution for each time increment. These parameters are shown in Table 1 for a middle portion of plume passage at 200 and 800 m. The centerline columns represent the position of the real plume axis for each cross section, in degrees azimuth. The size of the real plume cross sections is represented in \( \bar{\sigma} \) (standard deviation) and \( \bar{\sigma}^2 \) (variance) columns, and the shape of the real plume cross sections is represented by the skew. (skewness) and kurt. (kurtosis) columns. The \( \bar{\sigma} \) and \( \bar{\sigma}^2 \) entries are in degrees, while the skew. and kurt.
FIGURE 1. Time-History of $^{85}$Kr Concentration in a Diffusing Plume, Test C5

<table>
<thead>
<tr>
<th>TIME</th>
<th>CENTERLINE</th>
<th>$\Delta^2$</th>
<th>SKW</th>
<th>KURT</th>
<th>CENTERLINE</th>
<th>$\Delta^2$</th>
<th>SKW</th>
<th>KURT</th>
</tr>
</thead>
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<td>11</td>
<td>10 3.47</td>
<td>3.41</td>
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<td>17</td>
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<tr>
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<td>0.62</td>
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<td>29</td>
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<td>0.62</td>
<td>0.42</td>
</tr>
</tbody>
</table>

TABLE 1. Real Plume Position, Size and Shape Parameters for a Segment of C5
entries are pure numbers (coefficients of skewness and kurtosis). The mean and standard deviation rows at the bottom of the figure indicate the mean and standard deviation for each of the columns for the entire plume passage. The last row of numbers in the figure indicates the size and shape of the apparent plume resulting from this release. The various columns in this figure are only portions of each of the time-series analyzed in the remainder of the discussion.

An initial point of interest is whether autocorrelations of the size and shape parameters are significant or not. It is obvious that the autocorrelation of the position of the real plume segments should be high, even at long time-lags. As a result, it will not be discussed further. On the other hand, in Table 1 there does not appear to be a significant autocorrelation for any of the other parameters. This inference is examined in Figure 2, which shows the autocorrelograms for real plume width, skewness and kurtosis. Indeed, this figure shows that the autocorrelations are insignificant for each of the parameters at 200 m and for the coefficient of kurtosis at 800 m. However, at 800 m and a time-lag of 38.4 sec (1 time increment), the autocorrelations of real plume width (c) and the coefficient of skewness are both statistically significant. The question is then raised whether these correlations are physically real and related to plume position.

To examine this question and the ultimate validity of the original assumption, correlograms were constructed for the relationships between the real plume size and shape parameters and the position parameter. These correlograms are shown in Figure 3. None of the correlograms is significantly different from zero in the statistical sense, but the nature of the correlograms for the 800 m width and skewness is of some interest. In particular, the fact that the curves are relatively smooth and are almost mirror images of each other suggests that there may be a physical relationship between the real plume size and shape and its position.

In the light of the implications of a relationship between the real plume size and shape and its position and because of the lack of evidence of such relationship in the correlograms
\[
L_v = \frac{4.7}{b_{\text{scat}}} \tag{2}
\]

\[
M = 3.8 \times 10^5 b_{\text{scat}} \tag{3}
\]

On the top of Figure 2 the visibilities corresponding to 10, 25, and 100 miles are shown by arrows. Several CCN spectra and light scattering coefficients were also measured. A summary of all the data points is shown in Table 1, and the CCN spectra are plotted on Figure 3. Visibility values were calculated from Equation (2). The five curves for January 26th, Table 1. Summary of CCN Spectra and Light Scattering Coefficient (\(b_{\text{scat}}\))

<table>
<thead>
<tr>
<th>Date</th>
<th>Time</th>
<th>Average Number of CCN/cc</th>
<th>Number of Points</th>
<th>% S.S.</th>
<th>(b_{\text{scat}}) 10(^{-4})/m</th>
<th>Visibility Mile</th>
</tr>
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<tr>
<td>1-26-71</td>
<td>1503-1507</td>
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centrations were in the order of $10^4$ particles/cc. However, on a few occasions the concentration was about $10^3$ particles/cc. The lowest observed CCN concentrations in this area were 75/cc, 250/cc and 490/cc at supersaturations of 0.5, 0.75 and 1.0 percent, respectively.

The top curves in Figure 3 were taken on February 2nd and 3rd with a moderately smoggy day. Visibility was less than 10 miles.

CONCLUSIONS

From the data presented in this paper we can conclude that Los Angeles air pollution is a good source of CCN. More data are needed to establish the generality of Equation (1) in Los Angeles, and at a different supersaturation. It is also desirable to conduct similar investigations at different locations characterized by different types and different concentrations of pollution.

REFERENCES


FLUCTUATIONS IN TRACE GAS CONCENTRATIONS IN THE TROPOSPHERE

A. G. Gibbs and W. G. N. Slinn

A simple model is devised and approximations are made in an attempt to explain the observed dependence on removal rates of fluctuations in trace gas concentrations in the troposphere. The model suggests that the coefficient of dispersion \((\sigma^2/\langle x^2 \rangle)\) of the fluctuations of the gas concentration, \(x\), arising from variations in the production rate are proportional to \(r/\bar{x}\) where \(r\) is the rate of removal of the gas.

Junge, et al.\(^{(1)}\) recently remarked that "There is a general tendency for time and space fluctuations of atmospheric trace gases within the free troposphere to decrease as the residence time of the component increases."

Earlier, Junge\(^{(2)}\) suggested "If the space and time distribution of sources and sinks were the same for all gases, the time and space variations of all constituents would be proportional to (their removal rate) r."

This is certainly a reasonable result. If the removal rate is slow, then the influence of sources and sinks would have time to "spread" throughout the entire troposphere, and therefore the concentration fluctuations would be smaller than if the removal rate..."
were fast. It is also a potentially useful result for global contaminant budget evaluations, since it provides a simple estimate of the removal rate, based on measured fluctuations in the trace gas concentrations. Junge, et al.\(^1\) used this method to estimate the removal rate for CO.

**THE MODEL**

The purpose of this note is to explore a simple model in the hope that it might shed a little light on this atmospheric phenomenon. Toward this end, suppose that the concentration of a particular trace gas in a specific tropospheric air mass is the random variable \(\chi(t;\xi)\). The symbol \(\sim\) under a variable will be used to indicate that is a stochastic variable; the parameter \(\xi\) labels a specific air mass or a specific realization of the stochastic process.

As a specific air mass moves, the trace gas concentration varies because of time-dependent production and removal processes. Thus, if we use a coordinate system moving with the air mass, and ignore diffusion, then

\[
\frac{d}{dt} \chi(t;\xi) = p(t;\xi) - r(t;\xi), \tag{1}
\]

where \(p\) and \(r\) describe the production and removal processes experienced by the air mass. We propose to consider only the case of first order removal processes; i.e., those for which \(r = r_{\chi}\). Then (1) becomes

\[
\frac{d}{dt} \chi(t;\xi) + r(t;\xi) \chi = p(t;\xi). \tag{2}
\]

**LINEARIZATION**

Unfortunately we have not been able to make much headway trying to gain information from the complete Equation (2). What we seek is at least the first two moments of \(\chi\): its mean, \(\langle \chi \rangle\) and mean square \(\langle \chi^2 \rangle\) (or variance, \(\sigma^2 = \langle \chi^2 \rangle - \langle \chi \rangle^2 \)) where the averages in the above operations are taken over different realizations, \(\xi\); i.e., over different air masses. From the long-time \(t \gg r^{-1}\) solution to (2)

\[
\chi(t;\xi) = \int_0^t \exp\left\{ - \int_t^\tau r(t';\xi) \, dt' \right\} \chi(t';\xi) \, dt', \tag{3}
\]

it is relatively easy to find \(\langle \chi \rangle\) if \(r\) and \(p\) are uncorrelated, but we have been unable to evaluate \(\sigma^2\).

Instead, we linearize (2). If the fluctuations are small quantities about the mean value, then we write (2) as

\[
\frac{d}{dt} \langle \chi \rangle + r_{\chi} \langle \chi \rangle = \langle p \rangle + \delta p, \tag{4}
\]

and ignore the term \(\delta r \delta \chi\) which is of second order in small quantities. The lowest order terms in (4), i.e., the terms that exist if there are no fluctuations, satisfy

\[
\frac{d}{dt} \langle \chi \rangle + \langle r(t) \rangle \langle \chi \rangle = \langle p(t) \rangle. \tag{5}
\]
It seems reasonable to treat \(<r^{*}\) and \(<p^{*}\) in (5) as the time-independent, global-averaged removal rate, \(r\), and production rate, \(p\). In this case, the solution to (5) for \(t>>r^{-1}\) is

\[ <\chi> = p/r. \]  

which is a familiar result.

If there is a fluctuation in \(p^{*}\) or \(r^{*}\), then from (4) it "propagates" to \(\chi^{*}\) via

\[ <\delta\chi(t_1)\delta\chi(t_2)> = e^{-r(t_1+t_2)}\int_0^{t_1} e^{rt} \int_0^{t_2} e^{rt'} <g(t')g(t'')> dt dt'. \]  

\[ \delta\chi(t) = \int_0^t e^{-rt} \int_0^{t'} g(t';\xi) dt'. \]

Notice that \(<\delta\chi> = 0\) since \(<\xi> = 0\).

From (8) the autocorrelation of \(\delta\chi\) at large time is

\[ <\delta\chi(t_1)\delta\chi(t_2)> = e^{-r(t_1+t_2)}\int_0^{t_1} e^{rt} \int_0^{t_2} e^{rt'} <g(t')g(t'')> dt dt'. \]  

\[ \int_\tau e^{2\pi t\tau} dt' = \frac{1}{2\pi} \left(e^{2\pi \tau} - e^{-2\pi \tau}\right). \]

Using these results in (9), recalling that

\[ <g(\sigma)g(-\tau)> = <\xi> <g(\sigma)g(-\tau)>, \]

for a stationary process, and then upon collecting terms and letting \(t->\infty\), leads to

\[ <(\delta\chi)^2> = \frac{1}{r} \int_0^{\infty} e^{-rt} <g(\sigma)g(\tau)> dt. \]
ASSUMPTIONS

To proceed, a specification of the autocovariance of the fluctuations in the production and removal rates is needed. For want of something better, we assume that they decay exponentially:

\[ \langle \delta p(0) \delta p(\tau) \rangle = \sigma^2 p e^{-\lambda_p \tau}, \]
\[ \langle \delta p(0) \delta r(\tau) \rangle = 2 \alpha \sigma_p \sigma_r e^{-\lambda pr \tau}, \]
\[ \langle \delta r(0) \delta r(\tau) \rangle = \sigma^2 r e^{-\lambda_r \tau}. \]

Note that it may be reasonable to take the correlation coefficient, \( \alpha \), to be zero since production and removal rates could be expected to be uncorrelated. Substituting the above into (7) and (11) and evaluating the integrals gives

\[ \frac{\langle (\delta x) \rangle^2}{\langle x \rangle^2} = \left( \frac{r}{r^2 + \lambda_p \rho} \right) \frac{\sigma^2 p}{\rho^2} + 2 \alpha \left( \frac{r}{r^2 + \lambda pr} \right) \frac{\sigma_p \sigma_r}{\rho \tau} + \left( \frac{r}{r^2 \tau} \right) \frac{\sigma^2 r}{\tau^2}. \]

(12)

We expect that the average over different air masses of the "memory" of fluctuations in production or removal rates is short compared with the removal rates, themselves; i.e., \( \lambda >> \tau \). If this is true then (12) reduces to

\[ f_x^2 = \frac{r}{\lambda_p \rho} f_p^2 + 2 \alpha \frac{r}{\lambda pr} f_p f_r + \frac{r}{\lambda_r} f_r^2, \]

(13)

where the \( f \)'s are the coefficients of dispersion (standard deviation divided by the mean) of the appropriate quantities.

SUMMARY

In summary, the objective of this brief study was to determine a relationship between the fluctuations in trace gas concentrations and their removal rates. We linearized the general expression (2), and note that the resulting theory is self-consistent insofar as the solution (13) does predict that the neglected term, \( \delta r \delta x \), is much smaller than those which were retained, \( O(\delta x \delta r) \). Thus from (13)

\[ \frac{\langle \delta x \delta r \rangle}{\langle x \rangle \langle r \rangle} = O(f_x) << 1. \]

If the gas sampling is done at a single location for which the concentration fluctuations could be attributed to different long range trajectories of the air masses, then (13) suggests that fluctuations in the production rate alone would cause concentration fluctuations that vary with the removal rate according to \( f_x \propto r^{-1/2} \). In contrast, fluctuations in the removal rate alone would lead to \( f_x \propto r^{1/2} \delta r \), which is proportional either to \( r^{1/2} \) or to \( r^{-1/2} \), depending on whether the fluctuations in \( r \) are characterized by \( \sigma_r \) or by \( f_r \). In the more general case when both \( r \) and \( p \) experience fluctuations, \( f_x \) has the form given by Equation (13).

REFERENCES


EXAMPLE OF THE EVALUATION FUNCTION
FOR A GOOD SET OF DOMAIN VALUES

Of all the physically realistic sets of domain values used, possibly the best analysis was achieved for \( H_c = 15, S \) in \( S = 4, C_1 \) in \( B = 600, \) and \( C_2 \) in \( V_d = 3.0. \) This set may be written \( (15, 4, 600, 3.0). \) This cannot be considered an optimum set yet since further statistical evaluation of various sets of domain values is advisable. Also, further improvements in the basic model are possible. Nevertheless, using the above set of domain values, calculation with 116 \( r_{ij} \) yields

\[
N = 1.35 \quad S = 2.25
\]

The histogram of the \( r_{ij} \) and the \( f_{r_{ij}}, \) i.e., folded \( r_{ij} \) (the larger of \( r_{ij} \) or its reciprocal) is given in Table 1.

Attention is drawn to the following:

1. 63% of the calculated crosswind integrated exposures (CIE's) are within a factor of 2 of the measured CIE values.
2. 90% of the calculated CIE's are within a factor of 4 of the measured CIE's.
3. No \( r_{ij} \) values were found to be zero in this analysis.

The fourth element in the evaluation function, referred to earlier in the report, is simply the sum of the first four numbers in the bottom line of Table 1 and provides a measure of the spread of the \( r_{ij} \) values about the desired value of unity. The sum reaches a maximum value of 4 if all the \( f_{r_{ij}} \) are less than 2 and reaches a minimum of zero if all the \( f_{r_{ij}} \) values are greater than 16. The sum, which will be called the weighted count, W.C., in this case is 3.49. The largest of the \( r_{ij} \) was 17.6 (seemingly anomalous).

At this point it is instructive to tabulate the evaluation function for a selection of sets of domain values, illustrating the manner of variation of the evaluation function when all elements in the domain are held constant but one (Table 2).

The detailed computer output gives a considerable variety of output parameters for each arc of each experiment. For the same domain \((15, 4, 600, 3.0)\) detailed output from three experiments is given in Table 3 -- one from each stability classification.

<p>| TABLE 1. Histogram of the ( r_{ij} ) and ( f_{r_{ij}} ) |
|---|---|---|---|---|---|---|---|---|---|
| ( r_{ij} ) Intervals | 0.0-0.0625 | 0.0625-0.125 | 0.125-0.25 | 0.25-0.5 | 0.5-1.0 | 1.0-2.0 | 2.0-4.0 | 4.0-8.0 | 8.0-16.0 |
| ( f_{r_{ij}} ) count | 0 | 1 | 2 | 8 | 30 | 43 | 23 | 7 | 1 |
| ( r_{ij} ) count | 73 | 31 | 9 | 2 | .1 |
| Cumulative normalized ( f_{r_{ij}} ) count | 0.63 | 0.90 | 0.97 | 0.99 | 1.00 |</p>
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<th>B in B</th>
<th>C₁ in B</th>
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The unfamiliar symbols are: M.E. - measured crosswind integrated exposure; P.S. - portion of tracer calculated to be still suspended at the i-th arc as determined from the integral involving deposition velocity; BF.R. - B filter ratio, i.e., the ratio of $o_2^a$ as computed using the Hay-Pasquill equation to $o_2^{0.2}$, the plume spread if B were infinite.
### TABLE 3. Detailed Data from Three Experiments for One Set of Domain Values

**Experiment U-78**

\( U_c = 5.5 \text{ mps}; \bar{U}(2) = 3.7 \text{ mps}; Ri = 0.005; \delta = 7; \)

\( u* = 0.35 \text{ mps}; V_d = 0.099 \text{ mps}; \phi_m = 1.09 \)

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<th>1200</th>
<th>1600</th>
<th>2200</th>
<th>3200</th>
<th>5000</th>
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<td>0.56</td>
<td>0.41</td>
<td>0.25</td>
<td>0.16</td>
</tr>
<tr>
<td>( BF.R. )</td>
<td>0.152</td>
<td>0.081</td>
<td>0.055</td>
<td>0.042</td>
<td>0.031</td>
<td>0.021</td>
<td>0.014</td>
<td>0.010</td>
</tr>
</tbody>
</table>

**Experiment U-79**

\( U_c = 5.8 \text{ mps}; \bar{U}(2) = 1.6 \text{ mps}; Ri = 0.08; \delta = 52; \)

\( u* = 0.38 \text{ mps}; V_d = 0.063 \text{ mps}; \phi_m = 3.12 \)

<table>
<thead>
<tr>
<th>X (Meters)</th>
<th>400</th>
<th>800</th>
<th>1200</th>
<th>1600</th>
<th>2200</th>
<th>3200</th>
<th>5000</th>
<th>7000</th>
</tr>
</thead>
<tbody>
<tr>
<td>M.E. (gm sec m(^{-2}))</td>
<td>3.38</td>
<td>11.66</td>
<td>7.29</td>
<td>9.82</td>
<td>5.09</td>
<td>2.79</td>
<td>3.66</td>
<td>0.59</td>
</tr>
<tr>
<td>( r_{ij} )</td>
<td>2.45</td>
<td>1.08</td>
<td>1.57</td>
<td>0.99</td>
<td>1.05</td>
<td>0.98</td>
<td>0.53</td>
<td>0.93</td>
</tr>
<tr>
<td>( c_z )</td>
<td>14.5</td>
<td>22.0</td>
<td>27.7</td>
<td>32.5</td>
<td>46.8</td>
<td>58.8</td>
<td>70.3</td>
<td>94.6</td>
</tr>
<tr>
<td>P.S.</td>
<td>0.98</td>
<td>0.88</td>
<td>0.78</td>
<td>0.69</td>
<td>0.45</td>
<td>0.30</td>
<td>0.21</td>
<td>0.09</td>
</tr>
<tr>
<td>( BF.R. )</td>
<td>0.520</td>
<td>0.189</td>
<td>0.133</td>
<td>0.103</td>
<td>0.053</td>
<td>0.035</td>
<td>0.025</td>
<td>0.014</td>
</tr>
</tbody>
</table>

**Experiment U-82**

\( U_c = 3.4 \text{ mps}; \bar{U}(2) = 0.6 \text{ mps}; Ri = 0.15; \delta = 94; \)

\( u* = 0.095 \text{ mps}; V_d = 0.045 \text{ mps}; \phi_m = 9.17 \)

<table>
<thead>
<tr>
<th>X (Meters)</th>
<th>400</th>
<th>800</th>
<th>1200</th>
<th>1600</th>
<th>2200</th>
<th>3200</th>
<th>5000</th>
<th>7000</th>
<th>12,800</th>
</tr>
</thead>
<tbody>
<tr>
<td>M.E. (gm sec m(^{-2}))</td>
<td>0.56</td>
<td>5.83</td>
<td>5.36</td>
<td>5.20</td>
<td>2.09</td>
<td>3.42</td>
<td>1.83</td>
<td>1.83</td>
<td>1.83</td>
</tr>
<tr>
<td>( r_{ij} )</td>
<td>0.16</td>
<td>0.48</td>
<td>1.25</td>
<td>1.95</td>
<td>3.25</td>
<td>1.18</td>
<td>0.53</td>
<td>0.53</td>
<td>0.53</td>
</tr>
<tr>
<td>( c_z )</td>
<td>6.9</td>
<td>10.3</td>
<td>12.9</td>
<td>21.5</td>
<td>26.9</td>
<td>31.9</td>
<td>43.2</td>
<td>43.2</td>
<td>43.2</td>
</tr>
<tr>
<td>P.S.</td>
<td>1.00</td>
<td>0.99</td>
<td>0.96</td>
<td>0.65</td>
<td>0.42</td>
<td>0.26</td>
<td>0.07</td>
<td>0.07</td>
<td>0.07</td>
</tr>
<tr>
<td>( BF.R. )</td>
<td>0.224</td>
<td>0.124</td>
<td>0.085</td>
<td>0.034</td>
<td>0.022</td>
<td>0.015</td>
<td>0.008</td>
<td>0.008</td>
<td>0.008</td>
</tr>
</tbody>
</table>

Analysis for a few of the domain sets has also been made with the data divided into the three stability classifications identified previously. For the domain (15, 4, 600, 3.0), the following are obtained:
Comparatively speaking, the graphs suggest that the measurements of exposure are somewhat low at 1200 meters, high at 1600 meters, low at 2200, and high at 3200 meters. Differences in sampling flow rates and associated nonisokinetic sampling errors appear to be the cause.

The graph for the most stable case shows a tendency of the model to overestimate for the arcs at intermediate distances and underestimate at the extreme distances. Since the critical Richardson's number is reached at a height not too far from the surface for this case, the absence of turbulent diffusion above that height might well be responsible for higher concentrations, and thus lower ratios at
FIGURE 2. Ratios, $r_{ij}$, for Test Numbers 69, 72, 74, and 79.

FIGURE 3. Ratios, $r_{ij}$, for Test Numbers 65, 71, 82, and 83.
the greater distances from the source, than would be estimated using only the turbulence at lower level characteristic height.

DISCUSSION

In examining Table 2, it is clear that selecting the set of domain values (15, 4, 600, 3) as initially optimum resulted from tempering statistical analysis results with insight into reasonableness of values based on prior independent investigations of turbulence structure, \( \delta \), and deposition. However, our basic understanding of these factors is incomplete enough that selecting the best final set of input parameters awaits further experimental evaluation.

There has been no criticism so far of the basic turbulence model accepted from similarity theory along the A.F.C.R.L. and Hanford formulae. Attention must be drawn to the fact that for stable cases the nondimensional shear, \( \sigma_m \), is a rapidly increasing function of \( \text{Ri} \), that for the most stable cases analyzed this parameter shifted the spectrum to the right by a full decade. If this is physically unrealistic for the extreme cases, then physically unrealistic values of \( \delta \) must be used to counteract the effect. Extension of direct turbulence modeling for greater stabilities through the height range of interest should be carried out to settle this question. The question of the role that critical Richardson's Numbers plays affecting the diffusion should also be answered in further turbulence investigations.

Also, from Table 2 it is seen that the analysis changes rapidly as a function of \( C_1 \) and of \( C_2 \), but slowly as a function of \( H_C \) and of \( B \). This suggests that the manner of variation of \( \delta \) with stability is more critical to ascertain than its neutral value. Thus, independent evaluation of \( \delta \) as a function of stability or turbulence intensity is important.

The important effect of \( C_2 \) variation point to the need for further independent evaluation of the deposition process. The best description of deposition velocity as a function of meteorological and surface variables should be sought.

Future studies should be carried out to extend the model to unstable conditions and to greater heights. The crosswind spread should also be included in future adaptations of the model, accounting for the spectrum of the lateral wind component, \( \delta \) for the lateral wind component, and the effects of wind direction shear.

REFERENCES


PROFILE-DERIVED ESTIMATION OF CROSSWIND INTEGRATED EXPOSURE FROM ELEVATED RELEASES: METHOD OF INVESTIGATION

D. C. Powell and C. E. Elderkin

A method is proposed for estimating without turbulence measurements the crosswind integrated exposure at ground level to a depositing tracer released at an elevated source in the surface boundary layer. The meteorological inputs for the calculations are: (1) the quantity of tracer released; (2) the height of release; (3) the mean wind speeds measured at 15 and 2 meters; (4) the Richardson number calculated from data taken at the same two heights. From these data the friction velocity, \( u_* \), is first estimated, values of \( a_z \) for the tracer at downstream distances corresponding to sampling are locations are calculated using a spectral model and the Hay-Pasquill beta equation, and finally, the crosswind integrated exposure is calculated from a Gaussian equation and corrected using a deposition velocity that is a function of the input variables.

INTRODUCTION

Estimation of downwind exposure to continuous release of effluent from an elevated source is of interest at many locations at which turbulence measurements are not available. This paper describes the initial steps in developing a method, in which the estimation is limited to crosswind integration of the exposure, which reduces the study to that of vertical dispersion as a function of wind and temperature profile derived parameters. The method may be divided into four major parts: (1) estimation of the friction velocity, \( u_* \), as a function of Richardson number; (2) estimation of the variance of the vertical dispersion, \( a_z^2 \), from \( u_* \) and from the vertical velocity spectrum and predicted values of the Lagrangian-Eulerian scale ratio for appropriate travel time; (3) estimation of crosswind integrated exposure from a Gaussian equation; and (4) calculation of a deposition velocity which is a function of the meteorological input parameters.

CALCULATION OF \( u_* \) FOR THE CONSTANT FLUX LAYER

From the relationship given by Businger, et al.\(^{(1)}\) for \( z/L \) (L is the Monin-Obukhov length) and the Richardson number, \( Ri \), for stable conditions, the following solution for \( z/L \) may be obtained. This value is assumed to apply at the geometric mean height between the two heights used for computing \( Ri \) and is therefore designated, \( z_g \).

\[
\frac{z_g}{L} = \frac{0.74 + 9.4 \cdot Ri + \sqrt{4.9 \cdot Ri} - 0.35}{9.4 - 44 \cdot Ri}
\]

(1)
When the log-linear wind equation from the same paper is solved for the friction velocity, \( u_* \), the result is
\[
\frac{u_*}{L} = \frac{z}{\ln(z/z_0)} - 4.7 \frac{z}{L} \; ; \; \frac{z}{z_0} = \frac{z}{z_0} \frac{z}{\zeta}
\]
where \( \overline{U}(z) \) is the mean value of the longitudinal wind component at a height of measurement, \( z ; z_0 \) is the surface roughness parameter; and \( \zeta \) is Von Karman's constant.

**CALCULATION OF \( \sigma_z^2 \) FROM \( u_* \) AND FROM THE SPECTRUM OF \( u_* \)**

The spectrum used is the Hanford logarithmic model for the vertical component.\(^{(2)}\) This may be written
\[
nS_w(n) = \frac{1.15}{1 + 1.5(f/0.46 \sigma_m)^{5/3}} u_*^2 (f/0.46 \sigma_m)
\]
where \( f \) is the nondimensional frequency, \( n = U_c \) and \( \sigma_m \) is the dimensionless wind shear, calculated according to Husinger, et al.\(^{(1)}\) by
\[
\sigma_m = 1 + 4.7 \frac{z_c}{L}
\]
where \( U_c \) is the calculated wind at a height \( z_c \) that is assumed to be a characteristic height for the diffusion process between the height of release, \( H \), and the height of sampling. Currently \( z_c \) is assumed to be between 0.5 \( H \) and \( H \).

Assuming that the vertical dispersion is a function of turbulence only, leads to use of the Hay-Pasquill equation exhibiting \( \sigma_z^2 \) as a function of travel time, \( T \). The equation may be written\(^{(3)}\)
\[
\sigma_z^2 = T^2 \int_0^\infty nS_w(n) \left( \frac{\sin^2(n\pi T/h)}{(n\pi T/h)^2} \right) d(n\pi T/h)
\]
where \( nS_w(n) \) is the logarithmic energy spectrum for the vertical component, and \( h \) is theoretically the ratio of the Lagrangian time scale to the Eulerian time scale of the turbulence. If the value of \( h \) is adjusted empirically, it may also be a function of other physical factors not included in the model, such as the shear of the mean wind.

Since the spectrum and travel time are functions of the height, so is \( \sigma_z^2 \). For an arc at distance \( x \) downwind from the source the travel time has been calculated according to
\[
T = \frac{x}{U_c}.
\]

The final computation of the crosswind integrated exposure for a non-depositing tracer with settling velocity \( V_s \) is from a Gaussian equation
\[
E(x/U_c) = \frac{\sqrt{\pi}}{\sigma_z(x/U_c)} \sqrt{\frac{Q}{\sigma_z(x/U_c)^2}} \exp \left\{ -\frac{1}{4} \left( \frac{H - x V_s}{\sigma_z(x/U_c)} \right)^2 \right\}
\]
where \( Q \) is the total tracer mass released. The dependency is given as a time function rather than as a distance function because Fuquay, et al.\(^{(4)}\) and Nickola\(^{(5)}\) have found that diffusion is better exhibited as a function of travel time than of travel.
distance. The inclusion of the settling velocity has the effect of tipping the cloud downward at an angle to the horizontal of \( \tan^{-1} \frac{V_s}{U_c} \).

**EFFECT OF DEPOSITION**

Equation (7) assumes a nondepositing tracer, or equivalently, a virtual source below the surface at height, \( H \). An equation accounting for the depletion of the tracer is given by Van der Hoven, (6) which when modified to include the settling effect may be written:

\[
\ln \frac{E_x}{E_x'} = \frac{V_d}{U_c} \int_{x'}^{x} \frac{1}{z} \left( H - x' \frac{V_s}{U_c} \right)^2 \exp \left( -\frac{1}{2} \frac{V_s}{U_c} \sigma_z^2 \right) dx' \tag{8}
\]

where \( E_x \) is the final exposure, calculated as a function of deposition, and \( V_d \) is the deposition velocity. \( \sigma_z(x) \) is as computed in Equation (4) with \( T = x/U_c \).

Exposures calculated according to Equation (8) can be compared to measure exposures summed along an arc of the Hanford diffusion grid, multiplied by the distance between samplers. The comparisons so obtained are the test for the method.

**REFERENCES**


DETERMINATION OF THE VON KARMAN AND KOLMOGOROV CONSTANTS
FROM MEASUREMENTS OF WIND STRESS AND SPECTRA

T. W. Horst

Two relationships between the von Karman and Kolmogorov constants are presented which independently provide constraints on the allowable values for these two constants. Combined, they permit the determination of both constants from the measurement of the stress, mean wind, and power spectrum in the inertial subrange with a single, fast response, three-dimensional anemometer during neutral atmospheric stability conditions.

Recent independent measurements of the stress and wind shear\(^1\) have indicated that the parameter relating these two quantities under neutral stability conditions, von Karman's constant, \(k\), is about 12 percent smaller than the previously well-established value of 0.40. If this latest value is correct, use of the previous value would lead to a 30 percent overestimate in the profile-measured wind stress. This same error would be carried into estimations of the turbulent kinetic energy within the atmospheric boundary layer, since it is the wind shear-wind stress interaction which produces most of the turbulent energy under all but very unstable conditions. Knowledge of turbulent energy levels is vital for the prediction of many aspects of aerosol transport within the atmospheric boundary layer: diffusion, deposition, resuspension, etc. Although a 30 percent error may in many instances be unimportant compared to the approximations which become necessary when theory must be applied to a real problem, this may also be an unacceptably large error when carefully controlled experiments are performed to develop the theory.

A search for other means of measuring this important constant has brought to light a relationship between \(k\) and the Kolmogorov constant of the power spectrum in the inertial subrange. Under neutral stability conditions within the constant flux layer, the equations defining the von Karman and Kolmogorov constants may be combined to obtain a relation between them and a third parameter, which can easily be measured to high accuracy with a single three-dimensional anemometer. A recent theoretical extension of Heisenberg's formula for the spectral transfer of turbulent energy to the problem of the turbulent boundary layer by Roth\(^2\) provides an independent relationship which closes the system of equations and permits evaluation of both constants.

Using Taylor's hypothesis for the relationship between spatial and temporal descriptions of turbulence, the one-dimensional power spectrum of the
longitudinal component of the wind in
the inertial subrange obeys the power
law,

\[ \omega S_u(\omega) = a(\overline{cU}/\omega)^{2/3} \tag{1} \]

where \( \omega \) is angular frequency, \( S_u(\omega) \) is
the power spectrum, \( a \) is the Kolmogorov
constant, \( c \) is the viscous dissipation,
and \( \overline{U} \) is the mean wind speed. If,
under conditions of neutral stability,
the terms which account for the ver-
tical divergence of turbulent and pres-
sure transport of kinetic energy can
be ignored, the steady state, horizon-
tally homogeneous turbulent kinetic
energy budget reduces to

\[ c = u_*^2 \frac{3\overline{U}}{\zeta} \tag{2} \]

where \( z \) is the vertical coordinate and
\( u_* \) is the friction velocity (equal to
the square root of the surface stress
divided by the air density). Von
Karman's constant, \( k \), is defined by
the equation

\[ \frac{2U}{3} = \frac{u_*}{kz} \tag{3} \]

which is strictly valid in the con-
stant stress layer only for a neutral,
steady state, horizontally homogeneous
situation.

These three equations can be com-
bined to give a relationship between
the von Karman and Kolmogorov
constants,

\[ ak^{2/3} = \frac{\omega S_u(\omega)}{u_*^2} \left( \frac{\zeta}{U} \right)^{2/3} \equiv C \tag{4} \]

Under the conditions stipulated above,
the quantity \( C \) is a constant which
has a very desirable property. If a
single three dimensional anemometer
which has frequency response into the
inertial subrange is used to measure
the power spectrum, stress and mean
wind, the resulting estimate for the
constant \( C \) is insensitive to calibra-
tion errors since they will only be
evident in the determination of the
mean wind. An instrumental error
will be made in the quantity
\( S_u(\omega) u_*^{-2} \) only if the hori-
tontal and vertical axes of the instru-
ment have inconsistent calibrations or if
these axes are inadequately aligned
to the true horizontal and vertical.
Equation (4) has been plotted in
Figure 1 for several values of the
constant \( C \). Reasonable values for
the von Karman and Kolmogorov con-
stants are seen to limit \( C \) to the
approximate range 0.8 to 1.2.

This equation may be combined with
an independent relationship between
\( a \) and \( k \) developed by Roth(2) to give
both of these constants from an ex-
perimental measurement of the single
constant \( C \). Roth has utilized the
hypothesis of Heisenberg for the spec-
tral transfer of turbulent kinetic
energy to derive the relationship

\[ ak^{4/3} = \left( \frac{4}{3} \right)^{2/3} = 0.126 \tag{5} \]

This derivation also applies only
when the conditions (1)-(3) are satis-
fied. Equation (5) has been plotted
in Figure 1, the intersection with
the lines parameterized by \( C \) deter-
mining \( a \) and \( k \) according to

\[ a = 0.50 \ C^{2/3} \tag{6} \]

\[ k = 0.355 \ C^{-1/2} \tag{7} \]
The same data set which led to a value of 0.35 for von Karman's constant gave a value of 0.50 for Kolmogorov's constant (3), confirming Equation 5,

\[ a \kappa^{4/5} = 0.125, \quad (8) \]

and producing a value of 1.01 for the constant \( C \).

A reexamination of Hanford data (4) for one test during neutral stability indicates that \( C \) may be as large as 1.1 to 1.2. Combined with the results of other investigators (5) that Kolmogorov's constant may be 10 percent greater than the value quoted above, this also lends support to the value 0.35 for \( k \). More data need to be collected during neutral stability conditions for the simultaneous, independent measurement of the two constants defined by Equations (1) and (3). One such series of measurements has been performed by Battelle-Northwest in cooperation with the University of Washington Atmospheric Sciences Department. The relationships presented here will provide a framework for examining these data and checking the results for internal consistency.

* See following contribution "Turbulence Measurements on the Bonneville Salt Flats" by the same author.
REFERENCES


TURBULENCE MEASUREMENTS ON THE BONNEVILLE SALT FLATS

T. W. Horst

A recent field trip to the Bonneville Salt Flats to make fast response measurements of the wind with three-component hot film and sonic anemometers had three objectives: the determination of the von Kármán and Kolmogorov constants, turbulence measurements within the lowest centimeters of the atmosphere, and comparison of the two types of anemometers. Fifteen hours of data were obtained and analysis is in progress.

A series of turbulence measurements was made during the period September 13-21 in cooperation with the University of Washington Atmospheric Sciences Department. Although the data will be useful for many purposes, the participation of Battelle-Northwest had three specific objectives: to obtain data during periods of neutral stability for the determination of the von Kármán and Kolmogorov constants, to obtain turbulence data within the lowest centimeters of the atmospheric boundary layer for the study of deposition and resuspension, and to investigate the performance of the three-dimensional hot film anemometer which is to be used for turbulence measurements from the Battelle-Northwest aircraft. The site for these measurements was the Bonneville Salt Flats.
near Wendover, Utah, a choice dictated by the requirements of the first objective for horizontally homogeneous terrain and by the desire to measure very close to the surface. The Salt Flats (Figure 1) provided a vast, extremely uniform plain, bordered by peaks which rise to heights of 300-1000 ft above it, and measurements were made with fetches from 80 to 50 km. Rougher elements were generally less than a centimeter in height.

The determination of the von Kármán and Kolmogorov constants requires measurements of the wind shear, Reynolds stress, power spectra, and dissipation during periods of neutral atmospheric stability.* Since in the constant flux layer the stability approaches neutral as the height decreases, the smoothness and uniformity of the Utah site were a great advantage, allowing measurements to be made quite close to the surface. A Kaijo-Denki three-dimensional sonic anemometer-thermometer was used at a height of 2 m for measurements of the Reynolds stress, power spectra, and heat flux (or stability). Measurements of the wind shear were provided by the U. of W. profile system (1).

* See preceding contribution, "Determination of the von Kármán and Kolmogorov Constants from Measurements of Wind Stress and Spectra."
using cup anemometers, and the dissipation was measured with the time differentiated output from a Thermo Systems three-dimensional hot film anemometer. This anemometer also provided a supplementary measurement of the Reynolds stress and, with a thermistor temperature probe, of the heat flux, as well as higher frequency measurements of the power spectra. The hot film anemometer was placed at either 1 m or 2 m. At the greater height, it was situated within the sonic array for a calibration check and for direct comparison of all aspects of its performance with that of the sonic. The 1 m location provided a closer realization of neutral stability.

* See contribution, "Deposition Velocities as a Function of Particle Concentration Reference Height and Atmospheric Stability," in this report.

Field studies of deposition and resuspension as well as the application of laboratory studies of these phenomena to the atmosphere require knowledge of the wind structure close to the surface.* This goal, too, was facilitated by the extremely smooth, uniform terrain of the Salt Flats. Additional measurements of the wind were made for this purpose with the hot film anemometer at heights of 10, 24, 44, and 77 cm. Analysis of these data is in progress.

REFERENCE


TURBULENCE MEASUREMENT AND ANALYSIS CAPABILITIES

C. E. Elderkin, A. G. Dunbar and D. C. Powell

A system for measuring and storing turbulence data from an array of sonic and Gill anemometers mounted on towers was developed at Hanford. Computer methods for processing and analyzing the turbulence data were also established.

INTRODUCTION

A system for measuring and storing the turbulence from several points in a large field measurement grid was developed at Hanford for the Air Force in connection with the Take-Off and Landing Critical Atmospheric Turbulence (TOLCAT) program. In addition, the techniques and computer
programs for processing and analyzing the data from this system were established. This measurement and analysis capability has also been found to be appropriate to the needs of the AEC and is currently being used in ongoing turbulence and diffusion studies. A brief description of this capability is given here.

INSTRUMENTATION AND FIELD ARRAY

The multi-point turbulence measurement system utilizes sensors mounted on an array of towers for measurement of turbulence fluctuations in the three components of the wind at various heights and horizontal spacings, and under various meteorological conditions. The measurement array is shown in Figure 1. The data are collected in the field on analog recorders, played back into a medium-size computer for pre-processing the data and are recorded on industry standard tapes for later details processing and analysis on a UNIVAC 1108 computer.

FIGURE 1. The Field Measurement Array
Two types of turbulence sensors are utilized: fast response sonic anemometers for the most accurate determinations of momentum and heat fluxes used in characterizing the stability of the atmosphere, for power spectral definition into the inertial subrange, and for spatial correlations over small separation distances; and less expensive Gill three-propeller anemometers for the large separation distances where their slower response is adequate.

The mountings for both types of instruments are constructed in a manner that assures true sensor leveling and optimum orientation into the wind at inaccessible remote locations on tower tops and on the ends of tower booms. This has been accomplished by utilizing remotely monitored electrolytic levelers on each sensor and an antenna rotor base on each sensor for remote reorientation from centrally located control trailers.

The sensors can be mounted on four permanently erected, 62-m towers or eight 50-m expandable, portable towers which can be repositioned for various measurement configurations to meet different experimental requirements. The sensor booms can be moved to various heights on the 62-m tower and the sensors on the tops of the 50-m towers can be raised and lowered to measure the turbulence at selected heights.

The two control trailers, downwind of the tower array, contain the signal conditioning and data recording equipment. Two 14-channel analog magnetic tape recorders are generally used to record the three wind component signals from each of three sonic anemometers and five Gill anemometers, and the temperature signals from each of two sonic. One channel on each recorder is used for a coordination timing signal.

DATA PROCESSING

After each experiment, tape recordings of the field data are taken to a computer laboratory where the data are played back from one of the analog tape units into an analog-to-digital converter, interfaced to an SEL 840A medium-size computer. The computer is programmed to test the data for reasonableness and to perform preliminary editing; average a selectable number (usually five) of the 100 to 200 basic samples per second together to minimize aliasing and provide a range of optional digitizing rates, make instrument corrections, such as conversion of sonic nonorthogonal measured components to cartesian wind components; perform scaling functions to account for gains and offsets used during measurements; and finally, to record digital values of the three orthogonal wind components and the temperature on an industry standard digital tape recorder interfaced with the SEL 840A, for more thorough and repeated analyses to be performed later on a UNIVAC 1108 computer.

The 1108 programs perform more refined editing and corrects Gill anemometer data for deviations of the propellers from a true cosine response,
which is necessary for adequate turbulence statistics, before re-recording the true wind components in cm/sec and temperature in hundredths of a Centigrade degree on "master tapes." These are then processed by the probability subroutines which, for selectable time periods within the total data segment, calculate and remove means for each of the variables; re-orient the coordinate system in the direction of the mean wind, if desired; provide up to three different high-pass filterings of the data and the probability density functions - individual and joint - for each when requested; and calculate variances for each component and covariances between them, as well as the third and fourth moments. An option to plot any of the time series in the data segment being analyzed is also available. The unfiltered turbulence variables are then selected, two at a time, for analysis by the spectral subroutines. Here, the data are optionally detrended with the best fitting first or second order polynomial for each time series and tapered to prevent ringing oscillations in the fast Fourier transform calculated spectra. Then the Fourier coefficients are calculated from which banded power spectral and cross-spectral estimates are formed. The auto- and cross-correlation functions can, in turn, be transformed from these estimates if required. Variances and covariances for the detrended time series are also available from this program.

With this versatile system of measurement and analysis, data from a number of points arranged in various configurations in a field of turbulence can be accurately recorded and rapidly processed. The variety of analyses available allows the turbulence to be studied comprehensively in space and time.
A COMPUTER ALGORITHM FOR CORRECTING NONCOSINE RESPONSE
IN THE GILL ANEMOMETER

T. W. Horst

A fast computational algorithm is presented which ade-
quately corrects for noncosine response in the Gill UVM
propeller anemometer. Computational time is reduced by per-
forming calculations in integer arithmetic and by indexing
the cosine correction as a function of the cosine of the
angle between the wind vector and the propeller axis, rather
than as a function of the angle itself.

After considering such factors as
initial cost and reliability, the Gill
three-propeller anemometer was chosen
as a basic tool to be used in several
research programs at Hanford to mea-
sure the three components of the wind.
Careful comparison experiments with
the Wind Component Meter and the
Kaijo Denki sonic anemometer have
shown that the most serious error in
the Gill anemometer at frequencies
below about 0.3 Hz is the inability
of an individual propeller to totally respond to the component of the wind
parallel to its axis of rotation.\(^{(1\cdot2)}\)

Above this frequency, the inertia of
the rotating propeller begins to sig-
ficantly reduce the response of the
instrument, but this is not a serious
problem for many applications which
only require information about the
larger eddies. The former problem,
termed noncosine response, can be
adequately corrected for during digi-
tal computer processing of the data
by using the manufacturer's response
data and a computational algorithm
presented here. It has evolved
through several versions, each being
faster than the former. This is an
important point since the algorithm
is applied to each and every digital
data sample (up to 60 per minute of
data) and charges for computer ser-
VICES are based on computation time.

The component of the wind parallel
to a given axis is

\[ u = S \cos \theta \]  \hspace{1cm} (1)

where \( u \) is the wind component, \( S \) is
the magnitude of the wind vector,
and \( \theta \) is the angle between the wind
and the propeller axis. The Gill
propeller anemometer measures less
than this ideal quantity, hence the
term noncosine response, and the de-
viation from cosine response becomes
worse as the angle increases from
0° to 90°. The cosine response
will be worst for the vertical axis
propeller, the wind fluctuating
about an angle of 90°, and hence this
arm is calibrated for a 25 percent
greater output than the horizontal
arms to bring the response closer to
the cosine law within the expected
input range of 60° to 120°. Using
the manufacturer's wind-tunnel-derived response data, corrections to the measured wind have been determined as a function of \( \cos \theta \) and are listed in Table 1.

The algorithm, written in FORTRAN, is listed in Figure 1. Wherever possible, computations are performed in integer arithmetic to decrease execution time. Since the correction for each measured wind component is

---

### TABLE 1. Gill Cosine Correction Factors, Multiplied by 100, as a Function of \( \cos \theta \)

<table>
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<tr>
<th>( \cos \theta )</th>
<th>HORCOR</th>
<th>( \cos \theta )</th>
<th>HORCOR</th>
<th>( \cos \theta )</th>
<th>HORCOR</th>
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<td>173</td>
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Cosine Correction for Vertical Axis

<table>
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<th>( \cos \theta )</th>
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<th>( \cos \theta )</th>
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</table>

VERCOR = 100 for \(| \cos \theta | > 0.50 \)
a function of the angle between that propeller axis and the wind vector and since the correct wind vector may have an orientation different from that of the uncorrected wind vector, an iterative scheme must be used until the results converge. The direction cosines are calculated from the three wind components, the appropriate

```
INTEGER GU,GV,GW,Y,W,S
INTEGER HORCOR(101),VERCOR(101)
C
N=0
I=ISAVE
J=JSAVE
K=KSAVE
C CORRECT DATA FOR NONCOSINE RESPONSE
C USING CURRENT DIRECTION COSINES
40 U=GU+HORCOR(I)/100
V=GV+HORCOR(J)/100
W=GW+VERCOR(K)/100
C CALCULATE NEW DIRECTION COSINES
C AND CONVERT TO SUBSCRIPTS
FU2=U**2
FV2=V**2
FW2=W**2
S=SQRT(FU2+FV2+FW2)*0.5
II=(U*1000+S*10)/20+51
JJ=(V*1000+S*10)/20+51
KK=(W*1000+S*10)/20+51
C COMPARE NEW COSINES WITH OLD
C ITERATE IF TOO DIFFERENT
10 IF IABS(II-I-1)<10*10+20
15 IF IABS(JJ-J-1)<15*15+20
C CHECK NUMBER OF ITERATIONS
C STOP IF TOO LARGE
20 N=N+1
IF (I-N)<30*30+80
C REITERATE WITH NEW DIRECTION COSINES
80 I=II
J=JJ
K=KK
GO TO 40
C REPLACE RAW DATA WITH CORRECTED DATA
30 GU=U
GV=V
GW=W
C SAVE CURRENT COSINES TO INITIALIZE
C CORRECTION OF NEXT DATA SAMPLE
ISAVE=I
JSAVE=J
KSAVE=K
```

**FIGURE 1.** FORTRAN Algorithm for Correcting Noncosine Response in the Gill UVW Propeller Anemometer
corrections are selected and applied to the data, and new direction cosines are calculated from the corrected wind components. A time consuming operation, that of calculating

$$\theta = \cos^{-1}\left(\frac{u}{\sqrt{u^2 + v^2 + w^2}}\right)^{1/2}$$

(2)
is eliminated by tabulating and applying the correction as a function of $\cos \theta$ rather than $\theta$. Less than six repetitions of these steps is normally adequate for successive direction cosines to agree within $10^{-2}$.

Additional comments on the algorithm are listed separately for certain of the FORTRAN statements.

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**Note 1:** GU, GV, GW are the raw data points (in cm/sec $\times 2$) for the three components of the Gill anemometer. U, V, W, S are the corrected wind components and their vector sum.

**Note 2:** HORCOR, VERCOR are the tabulated corrections (Table 1) multiplied by 100.

**Note 3:** N keeps track of the number of iterations.

**Note 4:** I, J, K, etc., are the direction cosines converted to the appropriate subscripts for HORCOR(I) and VERCOR(K). The first iteration for each set of three data points is performed with the corrected direction cosines from the previous data point.

**Note 5:** The root sum of $u^2, v^2, w^2$ must be calculated in floating point arithmetic.

**Note 6:** The 10 added to the direction cosine rounds it off before conversion to a subscript.

**Note 7:** Iteration ceases only if the old and new cosines all agree within $\pm 0.02$ or if six iterations have been performed.
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the mean plume axis, but there is more than 2 orders of magnitude difference between them at positions 2.5 \sigma_y from the mean plume axis.

The relationship between absolute peak-to-mean concentration ratios and the time ratio is clearly shown in Figure 4. As with conventional \( \frac{P}{M} \), \( \frac{P}{M_a} \) monotonically increase with increasing \( T/t \). Unlike conventional \( \frac{P}{M} \), however, the \( \frac{P}{M_a} \) do not start from a common point at \( T/t = 1 \). Rather, \( \frac{P}{M_a} \) appear to converge at some large value of \( T/t \).

In the original discussion of peak-to-centerline-mean ratios, an expression was derived relating \( \frac{P}{M_a} \) with crosswind position through Gifford's Fluctuating Plume Model.\(^{(2,3)}\) This expression, however, contained many unknown quantities. It is hoped that in the future an empirical or semi-theoretical model can be developed which will permit the prediction of absolute peak-to-mean concentrations.

REFERENCES


AN EXTENSION OF THE PEAK-TO-MEAN CONCENTRATION RATIO MODEL

J. V. Ramsdell

The peak-to-mean concentration ratio model developed in 1969 for ground-level plumes is extended to predict ground-level peak-to-mean ratios in elevated plumes.

During 1969 an empirical model was developed which related $\frac{P}{M}$ ratios to the crosswind position in the mean plume and the ratio between the averaging times for the mean and peak concentrations.\(^{(1)}\) This model was based on ground-level measurements of plumes released at ground level and therefore, could not properly be used in connection with problems related to elevated sources. During the past year $^{85}$Kr data taken on towers have been used to evaluate the model in prediction of peak-to-mean concentration ratios in elevated plumes.

The original model was developed using the graphical relationships between $\frac{P}{M}$ and $\frac{|y|}{\sigma_y}$, and $\frac{P}{M}$ and $T/t$, as well as the following 4 theoretical considerations as guidelines:
1. At $T/t = 1$, $\frac{P}{M}$ must equal 1 for all crosswind positions in the plume.
2. $\frac{P}{M}$ ratios are a minimum at $\frac{|y|}{\sigma_y} = 0$.
3. $\frac{P}{M}$ ratios approach a maximum at large $\frac{|y|}{\sigma_y}$.
4. $\frac{P}{M}$ ratios cannot exceed $T/t$ in magnitude.

The final form of the original model was

$$\frac{P}{M} = 1.0 \, y^2 \, x^4 + 0.90 \, x^2 + 1.0 \quad (1)$$

where

$$y^2 = \tanh^2 \left( \frac{|y|/\sigma_y}{x - 1} \right) \quad (2)$$

and

$$x^2 = \ln(T/t). \quad (3)$$

An isometric view of the model is shown in Figure 1.

To extend the $\frac{P}{M}$ model to elevated sources, it was assumed that the rate at which a ground-level plume diffuses upward and the rate of downward diffusion of an elevated plume are equal. In addition, it was assumed that a cross-section of the plume perpendicular to its axis would be roughly elliptical in shape. With these assumptions in mind, the $^{85}$Kr diffusion test was analyzed. Estimates of the standard deviation of the vertical distribution of the tracer ($\sigma_y$) were obtained using the bivariate normal equation and measured values of the centerline concentration, windspeed, and $\sigma_y$. Complete reflection of the plume at the ground was assumed for these estimates.
The $\frac{\text{PM}}{\text{M}}$ ratios were grouped by $T/t$ and the normalized position of the sampler within the plume, where normalized position is defined as

$$r/\sigma_r = \left[ (y/\sigma_y)^2 + (z/\sigma_z)^2 \right]^{1/2}. \quad (4)$$

At the same time, it was assumed that the crosswind term in the original model could be replaced by a radial position term. That is, the $y^2$ term is replaced with an $R^2$.

$$R^2 = \tanh^2 \left[ \frac{r/\sigma_r}{X-1} \right] \quad (5)$$

The extended model is then

$$\frac{\text{PM}}{\text{M}} = 1.9 \ R^2 x^4 + 0.90 \ x^2 + 1.0. \quad (6)$$

To check the validity of the model under these assumptions, the extended model was used to predict a $\frac{\text{PM}}{\text{M}}$ corresponding to each of the observed values.

The peak-to-mean ratios predicted by the model were reasonably close to the observed values as shown in Figure 2. A tendency of the initial model to over predict $\frac{\text{PM}}{\text{M}}$ ratios at large $|y|/\sigma_y$ and small $T/t$ is still evident as the data tend to drop below the line in the middle of the $\frac{\text{PM}}{\text{M}}$ ratio range. But, the most important feature of Figure 2 is that in no case did the
model significantly under predict a P/M ratio. Some caution in use of the extended model is still in order, however, as the vertical extent of the data was limited to \( z/\sigma_z \leq 1.1 \).

REFERENCE


TRACER PARTICLE RESUSPENSION CAUSED BY WIND FORCES UPON AN ASPHALT SURFACE

G. A. Sehmel

Particle resuspension rates caused by wind stresses upon dry tracer particles were determined in field tests on an asphalt surface. For average wind speeds from 5 to 8 mph, the fraction of tracer resuspended/sec was from \( 5 \times 10^{-9} \) to \( 6 \times 10^{-8} \). For average wind speeds from 3 to 14 mph, the fraction of tracer resuspended increased when a man walked once through the tracer. The fraction resuspended per walk through was from \( 1 \times 10^{-6} \) to \( 7 \times 10^{-6} \).

The resuspension into the atmosphere of deposited particulate material presents potential inhalation hazard problems. Unfortunately, little is known about the effectiveness of the wind to raise deposited contaminants into the
air from the surface. It is clear, however, that the properties of the atmosphere, the properties of the surface, and the properties of the particulate contaminants are all important to the problem.

Initial experiments were performed in the field to determine wind-caused or-aided particle resuspension rates. The current site for some resuspension experiments is shown schematically in Figure 1. Shown is an asphalt surface onto which tracer ZnS particles were placed and a downwind air and ground deposition sampling grid for measuring resuspended particles. Not shown are the meteorological instrumentation and the real-time samplers (1) for measuring airborne ZnS.

The meteorological instrumentation located at the asphalt edge were a vector-vane at the 4 ft height and two separate three cup-anemometers at the 1 ft height. At the 10 ft distance, integrating anemometers were located at heights of 13 in., 4 ft, and 7 ft to determine the average stress.

Particle resuspension rates were determined by using a mass balance around the initial ZnS placed on the asphalt, the air and ground exposures, and the sampling time. Average particle resuspension rates were determined to be from $5 \times 10^{-9}$ to $6 \times 10^{-8}$ fraction resuspended/sec for average wind speeds from 2 to 9 mph. The dependency upon windspeed is yet to be determined. Nevertheless, a very

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**FIGURE 1.** Schematic Showing Experimental Design for Particle Resuspension from an Asphalt Surface Caused by Wind Stresses
rough windspeed effect was suggested by an apparent correlation between wind gusts and the response of the real-time sampler. There is some indication that wind gusts greater than about 15 mph rapidly resuspended particles. This apparent correlation will be investigated more fully by magnetic tape recording of the signal outputs and determining the correlations.

Forces in addition to wind stresses can increase the particle resuspension rate. One such force is the mechanical disturbance of a man walking down the 50 ft length of tracer. In this case, the resuspension rate is the fraction of particles resuspended each time the person walks through the tracer. For windspeeds of 3 to 14 mph, these resuspension "rates" were determined to be from $1 \times 10^{-5}$ to $7 \times 10^{-4}$ fraction resuspended per walk through. This addition of mechanical force appears to cause more rapid particle resuspension.

**REFERENCE**


### Soil Resuspension by Surface "Creep"

G. A. Sehmei and F. D. Lloyd

A sampler was tested for measuring particle resuspension by surface creep. The sampler collects particles from at least 50 to 205 um in diameter.

Soil erosion by wind forces has been classified\(^1\) as soil particle movement by suspension, saltation and surface creep. Particles moving in surface creep are large in diameter and move on or very close to the ground surface. A surface creep sampler has been designed and tested for measuring this surface creep.

Bagnold's\(^1\) original design for measuring surface creep was a small bottle recessed into the ground. The bottle cap had a small hole through which the surface creep particles fell and were collected in the bottle. This bottle design was satisfactory for measuring gross soil and sand movement since the collected sand and
soil were easily analyzed by gravimetric methods. In contrast, the problem of analysis and representative sampling of radioactivity resuspension by surface creep is more difficult. Firstly, a larger ground surface should be measured for surface creep in order to obtain the average surface creep from any "hot spots" of contamination. Secondly, a sufficiently large mass of sand moving by surface creep must be collected in order that the minute quantities of radioactivity in the sample can be analyzed to be greater than background.

A surface creep sampler was constructed which is a 2 ft x 2 ft x 1 in. deep (61 cm x 61 cm x 2.5 cm) open topped box. Dividers are placed in the box on 1 in. centers in order to form a gridwork of 1 in. x 1 in. open topped squares within the larger box. The sampler bottom was closed with masking tape which was cut away from each square in order to remove the particles within each square.

The sampler was tested in a clear area near the meteorology building. It was placed in the sand so that the top of the box was approximately at ground level. The sampler collected particles for several days.

After particle collection, the sampler was removed and analyzed for the relative mass loading in each small square and for the particle size distribution in the squares along the upwind edge of the sampler and at the middle of the sampler.

The mass collected in each small square has been normalized to the mass collected along the upwind edge. These relative particle collections in each individual square are shown in Figure 1. The collection is seen to decrease

![Data Plotted at Center of Each Square](image.png)

**FIGURE 1.** Surface Sampler Total Mass Collection Profile
rapidly with downwind distance. Apparently, the large decrease must be caused by many particles actually "rolling" into the edge squares and fewer particles being airborne for short "jumps" into the downwind squares.

The particle diameters collected by the surface sampler were determined with a series of screens in a sonic sifter (Allen Bradley Co.). The particle size distributions from this sieving is shown in Figure 2. The mass median particle diameter along the upwind edge is 136 μm while the mass median particle diameters is 108 μm at the center of the sampler. Obviously, the sampler does show a size bias as well as mass sampling bias from the upwind edge. This bias is considered to be negligible compared to the more difficult problem of closing a mass balance around resuspending radioactive particles.

This surface sampler does collect particles at least from 20 to 25 μm in diameter. The smaller respirable particles which are the initial does-to-man hazard may be collected, but too little sand mass was collected for analysis.

![Image](Neg 715857-4)

**Figure 2.** Particle size distribution of sand collected in surface sampler

**Reference**

SOIL RESUSPENSION BY "SALTATION"
G. A. Sehmel and F. D. Lloyd

A sampler for collecting particles moving by saltation was constructed and its performance measured in the field. Most soil movement occurs within 1 ft of the ground surface. Particle exposures as a function of height are interpreted to show that the concentration of respirable size particles decreases 1 to 1.5 order of magnitude within 1 ft.

Soil erosion by wind forces has been classified(1) as soil particle movement by suspension, saltation, and surface creep. Particles moving in saltation are initially lifted almost vertically into the air before the gravitational force causes deposition. Saltation is by a series of "jumps" or "hops" to various heights and downwind distances.

An omni-directional saltation sampler has been tested in the field for total mass collection and particle size distribution as a function of height. The sampler is shown schematically in Figure 1. The sampler is constructed from metal funnels with a maximum diameter of 16 cm. The funnel outlet has been cut-off, inverted, and resoldered to form collection cups for particles. Typical sand collection pathways are shown by the dotted lines. The particles enter into each section and are collected as the sand accumulation shown in the figure. The present assumption is that collection efficiency is 100 percent.

The particles collected in the sampler after several days in the field were analyzed for the vertical profile of mass collected and also the particle size distributions. The particle mass collection has been normalized in

FIGURE 1. Schematic of Funnel Collector
Figure 2 to show the mass fraction collected in the funnel sampler as a function of height. The curve shows that most of the particle mass is collected close to the ground level. This is consistent with Bagnold(1) and Chepil(2) who showed that over 90 percent of soil movement occurs below a height of 12 in. (30 cm).

The particle size distributions have been determined by sieving (sonic sifter, Allen Bradley Co.) the particles collected on each funnel or series of funnels. These distributions are shown in Figure 3. The mass medium particle diameters are from 140 to 200 μm, and are a function of height.

The exposure (particle collection per unit height) is shown in Figure 4 for several particle size ranges. The exposures decrease rapidly with height and show a 1 to about 1.5 order of magnitude decrease to a height of 30 cm. Although similar rapid decreases have been reported by Chepil for total soil exposure, these are the first data that show the effect of particle diameter on exposure. If the assumption is made that these curves can be extrapolated to smaller respirable particle diameters, the conclusion is that respirable particles also show a rapid decrease with height in the first 12 in.

**FIGURE 2. Funnel Sampler Particle Collection Profile**

**FIGURE 3. Particle Size Distribution of Sand Collected as a Function of Height Above Ground**
(30 cm). This rapid decrease is unexpected since particle concentrations of 20 to 60 μm in diameter have been shown to decrease less than one order of magnitude from 1 to 400 ft.

REFERENCES


AIRBORNE DUST CONCENTRATIONS

G. A. Sehmel and F. D. Lloyd

An airborne dust concentration of 189 mg/m³ occurred during the high winds on January 11, 1972. This dust concentration at an average wind speed of 29 mph is used to extrapolate airborne dust loadings measured in 1950 for wind speeds less than 18 mph.

The resuspension of radioactive particulate from soil surfaces is intimately related to the airborne soil particle concentrations. Airborne soil particle concentrations were measured during the severe wind storm of January 11, 1972.
A 20 cfm high-volume air sampler with a 1" in. diameter-particle impactor was placed with the impactor facing into the wind. The lower impactor edge was about 4" in. above ground level. Particles were sampled for two hours, which resulted in 12.88 g of soil being collected. This quantity of material overloaded the impactor and consequently a particle size distribution of the airborne soil cannot be presented. This dust loading corresponds to an average dust concentration of 180 mg/m$^3$.

The wind velocities during the test period are shown in Table 1. The integrated average wind velocity is 29 mph.

<table>
<thead>
<tr>
<th>Time</th>
<th>Average</th>
<th>Maximum Dust</th>
</tr>
</thead>
<tbody>
<tr>
<td>1535</td>
<td>32</td>
<td>50</td>
</tr>
<tr>
<td>1405</td>
<td>34</td>
<td>44</td>
</tr>
<tr>
<td>1435</td>
<td>30</td>
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<td>1505</td>
<td>21</td>
<td>27</td>
</tr>
<tr>
<td>1535</td>
<td>27</td>
<td>32</td>
</tr>
</tbody>
</table>

For comparison purposes, airborne dust concentrations recorded at the 4 ft elevation during 1950 are shown in Figure 1. The dust concentrations

![Graph showing airborne dust concentration as a function of air velocity.](Neg 720207-2)
do show a considerable scatter, nevertheless an exponential dependency upon wind velocity is suggested. Consequently, the smoothed curve is redrawn in Figure 2 on log-log coordinates. Also shown is the 189 mg/m$^3$ dust concentration during the January 11, 1972 dust storm. Based upon current knowledge, it appears reasonable to extend the 1950 curve to the current data point.

Dust concentrations are also a function of height. Thus, the comparison of concentrations at the 1 and 4 ft level may be biased. Nevertheless, it would appear reasonable to expect similar rapid increases of dust concentrations with wind velocity for any height. These dust concentrations as a function of height and particle size will be determined.

FIGURE 2. Airborne Dust Concentration as a Function of Average Air Velocity
Particle resuspension from an asphalt road caused by vehicular traffic

G. A. Sehmel

Particle resuspension rates were determined for ZnS* particles which were placed onto one lane of a two lane, seasoned asphalt road. Either a car or a 5/4 ton truck was driven through the deposited material or in the adjacent lane. The fraction of particles resuspended for one vehicle pass was determined for vehicle speeds of 5, 15, 30, and 50 mph. The fraction resuspended per vehicle pass ranged from 10^-6 to 10^-2 with the higher resuspension corresponding to higher vehicle speeds and driving directly through the deposited ZnS.

Particle resuspension from roads caused by vehicular traffic can produce downwind health or environmental hazards. These hazards include those due to the dose-to-man inhalation hazard of radioactive particles as well as airborne salt and lead damage on trees and vegetation near roads.

Particle resuspension from a dry seasoned asphalt road was determined\(^\text{(1)}\) using the field grid schematically shown in Figure 1. The important features are a wind blowing across the road into the particle sampling grid consisting of filters on air sampling towers and ground deposition samplers. These samplers collected particles after the vehicles were driven through the lane onto which ZnS was placed or after the vehicles were driven by on the adjacent lane. Concurrently, the air velocity profile was measured with the meteorological tower.

The tracer particle was phosphorescent ZnS* which has a theoretical density of 4.1 g/cc, a mass medium diameter of about 5 \(\mu\)m, and a number medium diameter of about 2 \(\mu\)m. This material was selected since ZnS can be non-destructively and rapidly analyzed\(^\text{(2)}\) on the filters using a phosphorescent technique.

The fraction of particles resuspended per vehicle passage is shown in Figure 2 for data obtained on the same day that the ZnS was placed on the road. The fraction resuspended increases with vehicle speed. Of particular interest is that the slope of

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* This related resuspension research was supported in part by the Atlantic Richfield Hanford Company's Environmental and Regulatory Technology Program.

* ZnS Type 2210, United States Radium Corporation.
has been treated with a thin layer of petroleum jelly to minimize particle re-entrainment. Inspection of the two solid curves at 30 mph, shows that particle re-entrainment begins at about 9 μm for the dry surface and at about 14 μm for the coated surface. The curve for the sub-micron particles is presently being better defined with additional data points. Once defined, all the experimental data will be processed to better predict eddy diffusivities (1) and deposition velocities.

The particle deposition flux is described by

\[ N = \left( \varepsilon \tau / \nu + D/\nu \right) u_* \frac{dC}{dz} - v_c C, \]

in which \( \varepsilon \) is the particle diffusivity, \( D \) is the Brownian diffusivity, \( \nu \) is the kinematic viscosity of air, \( z^* = z u_* / \nu \) is the dimensionless distance above the surface, and \( v_c \) is the terminal settling velocity. Previously, the assumption has been made that the particle eddy diffusivity is independent of gravity. This independence is being experimentally determined.

The deposition velocity of particles depositing onto smooth brass placed on the wind tunnel ceiling is shown in Figure 2. These data will be used in conjunction with the completion of data for Figure 1 to predict the effect of gravity on particle eddy diffusivities near surfaces. This gravitational effect is important in predicting particle deposition to the many geometries of environmental surfaces.

**FIGURE 2.** Deposition Velocities onto the Bottom of a Smooth Surface (Gravity Opposing Deposition)

Particle deposition velocities to the ceiling show the same functional increase with particle diameter as for deposition on the floor. The principle differences are that the deposition velocities are less and that particle re-entrainment occurs more readily from the ceiling. Particle re-entrainment occurs for 4 μm diameter particles (and smaller) at a wind speed of 30 mph.

Particle deposition to the ceiling shows the same maximum at about 9 μm as for deposition for the floor. These maxima for a 30 mph wind speed show that the gravitational force
can have a minor effect on particle deposition for some circumstances.

A gravel surface is the next surface to be experimentally studied in the wind tunnel. This surface is chosen, (1) to more realistically model the rougher environmental surfaces, and (2) to develop deposition velocity data for a "true suspension" deposition sampler which will be used in closing the airborne particle mass balance for plutonium resuspension rate experiments.

REFERENCE


DEPOSITION VELOCITIES AS A FUNCTION OF PARTICLE CONCENTRATION
REFERENCE HEIGHT AND ATMOSPHERIC STABILITY

G. A. Sehmel and T. W. Horst

Deposition velocities were calculated from a model based upon particle effective eddy diffusivities being greater than the eddy diffusivities of air momentum in the region closer than 1 cm to the deposition surface. These predicted deposition velocities are used to determine the relative constancy of deposition velocity when the height for defining the particle concentration is increased from 1 cm ($z_1$) to 10 m ($z_2$). The ratio $k_2/k_1$ is nearly unity for stable, unstable, and adiabatic atmospheres.

A particle eddy diffusivity model has been developed\(^1\) to predict particle deposition velocities for the lower limit of deposition velocities which might be expected in the atmosphere. This is the case of deposition onto a smooth surface. The model shows that the effective particle eddy diffusivity becomes increasingly greater than that for air momentum as the particle approaches the surface from a $z$ of about 20. That model has been used to predict deposition velocities, $K_1$, when the particle concentration reference height is 1 cm above the deposition surface. The purpose here is to take the results of that model and to investigate the
behavior of deposition velocities when the particle concentration reference height increases to 10 m and the particle eddy diffusivity is equal to that of air between 1 cm and 10 m height for stable, unstable, and adiabatic atmospheres.

The deposition velocity, $K_z$, is defined as the deposition flux, $N$, divided by the particle concentration, $C$, at a reference height. Thus, $K_z$ at any height can be calculated from $K_1$ by the relationship

$$\frac{K_z}{K_1} = \frac{N/C_z}{N/C_1} = \frac{C_1}{C_z}, \quad (1)$$

where $N$ is constant.

The ratio $K_z/K_1$ was determined from the model

$$N = - (\varepsilon/\nu + D/\nu) u_m \frac{dC}{dz} + v_t C, \quad (2)$$

in which $\varepsilon$ is the particle diffusivity, $D$ is the Brownian diffusivity, $\nu$ is the kinematic viscosity of air, $z = z/(\nu/\nu)$ is the dimensionless distance above the surface, and $v_t$ is the terminal settling velocity. Values of the turbulent diffusivity in the atmosphere were obtained from a generalized correlation (2) for momentum transport as a function of thermal stability, i.e., for representative values of $z/\nu$ for stable, unstable, as well as adiabatic atmospheres.

The ratio, $K_z/K_1$, is a function of $K_1$ and $v_t$ or only $K_1$ if $K_1 \gg v_t$. Thus, we must first define a series of $K_1$ so that we can see the effect of height and stability on $K_z$. The deposition velocities chosen for comparison are those in Figure 1, for a particle density of 1.5 g/cm$^3$. Deposition velocities are seen to vary several orders of magnitude.

The large variations in $K_1$ are reflected in the $K_z/K_1$ ratios shown in Figure 2 for a reference height of 10 m. The $K_z/K_1$ ratio is nearly unity for all atmospheric stabilities for particle diameters from about $10^{-2}$ to 5 $\mu$m. This constancy reflects the fact that the controlling resistance to particle mass transfer is below 1 cm. From 1 cm to 10 m, the airborne particulate concentration is nearly uniform.

The ratio $K_z/K_1$ is a minor function of stability and decreases to about 0.5 to 0.7 for particle diameters near $10^{-3}$ and 10 $\mu$m. These decreases are caused by the relatively high deposition velocities, $K_1$, and the resulting particle concentration profiles which must develop above 1 cm.
to supply the steady flux of particles to the deposition surface. From this comparison in Figure 2, we conclude that the minimum deposition velocities are nearly independent of atmospheric stability and have been adequately predicted. This means that wind tunnel generated deposition data can be applied to the atmospheric case.

Real deposition surfaces are usually rougher than the smooth surfaces experimentally studied to date in the wind tunnel. Consequently, we must also show that deposition velocities for rough surfaces are also nearly independent of particle concentration reference height. Deposition concentration ratios, $K_2/K_1$, are shown in Figure 3 as a function of friction.
FIGURE 1. Schematic Showing Experimental Design for Particle Resuspension from an Asphalt Road Caused by Vehicle Passage

The curve for a car driven through the tracer is approximately two. That is, the fraction resuspended is proportional to the car generated turbulence.

The fraction resuspended is from $10^{-5}$ to $10^{-2}$ and shows that resuspension decreased nearly an order of magnitude when the vehicle is driven by the tracer as compared to the vehicle driven through the tracer.

REFERENCES


Deposition velocities in a wind tunnel

G. A. Sehmel and S. L. Sutter

Deposition velocities for monodispersed particles were measured for deposition onto a smooth electrically grounded brass surface located on the floor and ceiling of a wind tunnel. The deposition on both the floor and ceiling shows that re-entrainment from a coated surface occurs at about 9 μm for a 30 mph windspeed. For the ceiling, re-entrainment of 4 μm particles occurs from a dry surface. A model will be developed from the deposition velocity data to predict the effect of gravity on observed deposition.

Wind tunnel studies were conducted to determine the deposition velocities of uniform diameter particles from turbulent air flow onto a smooth surface. The deposition velocity, \( K_1 \), is defined as the particle deposition flux divided by the airborne particle concentration at 1 cm distance from the surface. This small distance is used to better define the controlling mass transfer resistance at the surface. The current deposition surface is an electrically grounded, smooth, brass shim stock which is placed either on the floor or the ceiling of the 2 ft \( \times \) 2 ft cross section wind tunnel.

Particle deposition onto the dry surface has been extended to larger particles for the 15 mph wind velocity and to sub-micron particles for the 30 mph wind velocity. The smoothed curves are shown in Figure 1. Also shown is a curve at 30 mph for deposition onto a brass surface which

\[ \text{NEG 715856-1} \]

**FIGURE 1.** Deposition Velocities onto the Top of a Smooth Surface
velocity and reference height for assumed $K_1$ values from $10^{-1}$ to $10$ cm/sec. We see that at a $K_1$ of $10^{-1}$ cm/sec, that the $K_z/K_1$ ratio is nearly unity. As the deposition velocity $K_1$ increases to 1 cm/sec, the ratio $K_z/K_1$ decreases to about 0.4 to 0.8. These $K_z/K_1$ values suggest that the controlling particle mass transfer resistance is still within 1 cm of the deposition surface. Consequently, particle deposition velocity data from a wind tunnel can be used to realistically predict particle deposition to many atmospheric surfaces.

NOMENCLATURE

- $z$ = vertical coordinate
- $z^*$ = vertical coordinate $z^* = z u_*/\nu$
- $\Theta = \text{potential temperature}$
- $u_*$ = friction velocity $[(\tau_0/\rho)^{1/2}]$
- $L = \text{Obukhov Length } [-\Theta u_*^3/(\kappa g \omega \Theta^2)]$
- $\nu = \text{kinematic viscosity}$
- $\rho = \text{air density}$
- $\tau_0 = \text{surface shearing stress}$

An overbar denotes a time average and a prime the deviation from the average.

REFERENCES


DEPOSITION VELOCITIES OF RESUSPENDED PARTICLES

G. A. Sehmel

Particle deposition velocities were calculated from field experiments for the determination of ZnS-particle resuspension from asphalt surfaces. The deposition velocity range was from 0.4 to 27 cm/sec with a medium of 3.8 cm/sec.

The determination of particle resuspension rates is important for predicting the downwind dose-to-man inhalation hazard. These resuspension rates are the source term for models of the downwind diffusion and particle
deposition. Particle deposition velocities have been determined from results of field experiments for the resuspension of ZnS particles from asphalt surfaces.*

The particle airborne exposure profile as a function of height and the total ground deposition as a function of downwind distance were determined in the resuspension experiments. In the experiments, air filters were used to determine airborne exposure and similar filters were pressed into the soil surface adjacent to the asphalt in order to estimate ground deposition. These profiles were used to determine the average airborne exposure, \( M_a \), at ground level (\( z = 0 \)) and the deposition, \( M_g \), resulting from that exposure.

The deposition velocity, \( K \), is defined as the deposition flux, \( N \), divided by airborne concentration, \( C \). The deposition flux is

\[
N = \frac{M_g}{A} \tag{1}
\]

and the airborne concentration is

\[
C = \frac{(M_a)_{z=0}}{F_0} \tag{2}
\]

where \( A \) is the filter cross sectional area, \( F \) is the air sampling rate in volume/time, and \( \theta \) is the time length of the experiment. Since the time cancels, the deposition velocity is

\[
K = \frac{N}{C} = \frac{M_g}{(M_a)_{z=0}} \tag{3}
\]

That is, \( K \) can be calculated from the airborne exposure at \( z = 0 \) and the ground deposition exposure.

The experimental deposition velocities ranged from a minimum of 0.4 cm/sec to a maximum of 17 cm/sec for the resuspended ZnS.* Although the ZnS has a mass medium diameter of about 5 \( \mu \)m and a number medium diameter of about 2 \( \mu \)m, the particle sizes characteristic of the deposition velocities is unknown. For some experiments, the deposition velocity decreased as a function of distance from 0 to 30 ft downwind from the asphalt. In other experiments the trend was not definable. The median deposition velocity was 1.7 cm/sec and the medium was 3.8 cm/sec.

* See contributions "Tracer Particle Resuspension Caused by Wind Forces Upon an Asphalt Road," and "Particle Resuspension from an Asphalt Road Caused by Vehicular Traffic," in this report, same author.

ZnS type 5010, United States Radium Corporation
MEASUREMENTS AND GRAPHS OF TURBULENCE AUTOCORRELATIONS
IN SPACE AND TIME*
C. E. Elderkin and D. C. Powell

A series of turbulence measurements has been taken at heights of 16 and 30 meters at Hanford from a horizontal line of seven logarithmically spaced sensors. When the wind direction was essentially along-tower-line, the autocorrelation function in space and time for each wind component has been graphed by entering correlation isopleths on the space-lag versus time-lag plane. The results illustrate (1) the degree of similarity between the autocorrelation function in time and that in space (Taylor's hypothesis), (2) the relationship between the mean wind speed and an effective eddy translation speed, (3) the persistence of high correlation following advection (frozen field hypothesis), (4) the autocorrelation function at a horizontally moving point, and (5) the relationship between Eulerian and Lagrangian time-correlations.

INTRODUCTION

In 1967 Gifford\textsuperscript{(1)} elucidated a point of view holding that there are four distinct coordinate reference systems in which turbulence description is pertinent. The Eulerian fixed-point system (I) refers to turbulence as experienced by fixed-point measuring instruments. The Eulerian space system (II) is a "snapshot" of turbulence. The quasi-Lagrangian system (III) refers to turbulence as measured at a point moving with the mean motion. The Lagrangian system (IV) refers to the motion of a single fluid particle. Gifford also identified several writers who, from an intuitive basis, consider systems (III) and (IV) to be equivalent. Csanady\textsuperscript{(2)} also accepts this as being approximately true.

Because system (I) is a convenient frame of reference for measurement, inferences about turbulence in the other reference frames are often made from Eulerian fixed-point description. By extension of Eulerian fixed-point measurement to include a line of sensors logarithmically spaced in the direction of the mean motion, a description of turbulence in system (III) may be simulated. The primary purpose of this paper is to describe this experimental approach and indicate some initial results.

Relating systems (I) and (IV), we have the Hay-Pasquill beta hypothesis

* This work performed under United States Atomic Energy Commission Contract No. AT(45-1)-1880 and United States Air Force Contract No. F18616-M-5096, "Take-Off and Landing Critical Atmospheric Turbulence (TOLCAT)."
\( R(\tau_E) = R(\tau_L); \quad \tau_L = 8\tau_E \) \hspace{1cm} (1)

In Equation (1), \( R \) is a correlation function for any of the three turbulence components, \( \tau \) is time lag, the subscripts \( E \) and \( L \) indicate Eulerian and Lagrangian, respectively, and \( 8 \) is assumed to be a constant.

The assumption expressed by Equation (1) is more restrictive than the following expression given by Gifford.

\( \varepsilon = n_T/n_L \) \hspace{1cm} (2)

where \( n_T \) and \( n_L \) are characteristic frequencies in systems (1) and (IV). Equation (1) states that the spectra for a given turbulence component in systems (1) and (IV) are the same function distinguished only by transposition. Equation (2) does not involve this assumption. The \( \varepsilon \) defined therein can be an average value of a proportionality factor, i.e., a function of frequency. Neither Equations (1) or (2) preclude the likelihood expressed by P. B. Smith (3) that \( \varepsilon \) is different for each turbulence component.

The assumption that systems (III) and (IV) are approximately equivalent may be expressed by

\( R(\tau_L) = R(\tau, \xi), \quad \xi = \bar{U} \tau \) \hspace{1cm} (3)

The space lag, \( \xi \), is measured in the longitudinal direction. By combining Equations (1) and (3), we get another expression relating systems (I) and (III) contending that the Eulerian correlation function differs from the quasi-Lagrangian correlation function by only the scale factor, \( 8 \). This corresponds to the investigation that is of interest in the present paper and is expressed by

\( R(\tau_E) = R(\tau, \xi), \text{ requiring } \xi = \bar{U} \tau \)

assuming \( \tau = 8\tau_E \) \hspace{1cm} (4)

To initiate this investigation, an experiment was conducted which utilized the turbulence measurement and analysis system.*

For this experiment, data from seven three-component turbulence sensors were utilized in the analysis. The sensors were located on the expandable towers adjusted to 15 m height and logarithmically spaced along the mean wind direction at 4, 8, 16, 32, 64, and 128 m separations between consecutive sensors. Sonic anemometers were located at the shortest intervals - 4 m, separating the first and second, and 8 m between the second and third. Gill anemometers were located at the remaining four sampling positions.

**ESTIMATION AND CALCULATION OF THE EULERIAN SPACE COVARIANCES**

Each set of two towers in the logarithmic array is separated by a unique distance. Therefore, genuine spatial covariances, computed by averaging over space, are not available from this data. However, by cross-analyzing parallel components measured at distinct separation distances,

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* See the contribution "Turbulence Measurement and Analytic Capabilities," in this report.
a pseudo-spatial covariance may be defined as given below

\[ R_{\alpha\alpha}(\xi) = E[u_\alpha(t,x_0) u_\alpha(t,x_0+\xi)] \]

\[ \alpha = 1,2,3 \text{ no summation.} \quad (5) \]

Here the expectation is a time average; and \( x_0 \) and \( x_0 + \xi \) are the locations of two measuring points. Pseudo-spatial correlations computed in this manner will be referred to simply as spatial correlations hereafter. From a line of 7 positions, 21 distinct separation distances are available for calculating spatial correlations. The correlations for both space and time may be defined by extending the same principle to a correlation function of two arguments. The result, normalized, may be written

\[ R_{\alpha\alpha}(t,\xi) = E[u_\alpha(t,x_0) u_\alpha(t+\tau, x_0 + \xi)]/E[u_\alpha u_\alpha]. \quad (6) \]

An example of this type of turbulence analysis is given by Lumley and Panofsky\(^4\) in their Figure 5.22. Such two dimensional correlation function analyses have been calculated for each turbulence wind component for the one experiment discussed here where the array of sensors was at 15 m height, the wind direction was 9° to the line of the sensors at 6.4 mps, and slightly stable conditions prevailed. The analysis was for a 28-minute data sampling period with each variable sampled at 0.2 second intervals.

**PROPERTIES OF TURBULENCE ILLUSTRATED IN DIAGRAMS OF TIME SPACE CORRELATIONS**

The time-space correlation functions for the three turbulence wind components: \( u_\alpha' \) the horizontal component along the tower line; \( v_\alpha' \) the horizontal component normal to the tower line; and \( w' \), the vertical component, are given in Figures 1, 2, and 3, respectively. These diagrams may be interpreted in several interesting ways. First, it will be noted that the function marked off by the isopleths on the abscissa is a pure time correlation, and is necessarily symmetrical. The function marked off by the isopleths on the ordinate is the spatial correlation. The scales of the two axes are adjusted to be equal when \( \xi = \bar{U}t \). The correlation at a point moving with the mean motion is the function marked off by the isopleths on the 45° line labeled \( \xi = \bar{U}t \).

Also, Taylor's hypothesis predicts that the same identical function is marked off on the abscissa and the ordinate. The isopleths of correlation in Figures 1, 2, and 3 may be approximated by ellipses with their major axes along the line \( \xi = \bar{U}t \). If the major axes of the ellipses fall, for instance, above the 45° line, an eddy transport speed greater than \( \bar{U} \) is indicated.

Assuming that the quasi-Lagrangian correlation function is a good approximation to the true Lagrangian correlation function, Equation (4) predicts that the function marked off on the
a $45^\circ$ line will differ from that marked off on the abscissa by no more than a proportionality factor, $\delta$. Comparing lag times, $\tau$, for the equal values of correlation, the first of which is on the $45^\circ$ line and the second of which is on the abscissa, provides a ratio which is an estimate $\delta$. If Equation (4) holds for system (III), the value of $\delta$, so estimated, should be approximately the same regardless of where on the correlation diagram it is determined. A large volume of these graphs should show reliably whether or not $\delta$ varies systematically with correlation length, from one turbulence component to another and as a function of stability or turbulence intensity.

The eccentricity of the ellipses indicates to what extent the frozen field hypothesis holds. The strict frozen field hypothesis requires that the isopleths in the diagrams are straight lines parallel to the line, $\xi = \bar{U} \tau$, and symmetrically located on
each side of that line. The eccentricity should, in general, be inversely proportional to the relative turbulence intensity and directly proportional to the value of $\delta$.

The principal axes of the correlation contours appear to be slightly above the $\xi = \nu t$ line for all three components but is most clearly shown for $w'$ in Figure 3, indicating an effective eddy translation speed slightly exceeding the mean wind speed.

From Figures 1, 2, and 3, we see that the eccentricity (and thus $\delta$) is least for the $u'$ component graph and the greatest for $v'$. This does not correspond to an inverse ranking of the relative turbulence intensities since $u'^2 \delta = 1.28 \text{ m}^2/\text{sec}^2$, $v'^2 \delta = 0.74 \text{ m}^2/\text{sec}^2$ and $w'^2 \delta = 0.31 \text{ m}^2/\text{sec}^2$. This indicates that $\delta$ is not the same for each component is more clearly shown in Figure 4. It is also seen that some tendency toward smaller $\delta$ at larger $t$ is suggested. However, this suggests that the quasi-Lagrangian and Eulerian fixed-point correlation functions are not truly similar is premature since

---

**FIGURE 2.** Autocorrelation Function of $v'_a$ in Time and Space
FIGURE 3. Autocorrelation Function of $v'$ in Time and Space

FIGURE 4. Estimated $\beta$ for the Three Wind Components as a Function of Eulerian Time Lag
experimental factors could account for the small variations (smaller than between components) observed. The \(B\) values of about 5.3 for the lateral wind component and 3.9 for the vertical component are consistent with other evaluations of \(B\) and suggest that the technique for studying \(B\) described here is a useful approach.

REFERENCES


PROFILE-DERIVED ESTIMATION OF CROSSWIND INTEGRATED EXPOSURE FROM ELEVATED RELEASES: INITIAL MODEL

D. C. Powell and C. E. Elderkin

The crosswind integrated exposures measured during nineteen tests (Richardson number varying from 0.005 to 0.15) at nine arcs, 400 to 12000 meters downwind of an elevated release point 28 meters above ground, are compared with estimations calculated from a model. When suitable values are used for various input parameters to the model, the calculated exposures are within a factor of two of the measured exposures 63% of the time and within a factor of four 90% of the time. The model is successful only with large variations of the Ray-Pasquill \(B\) parameter with stability. The calculations also change sharply as the coefficient in the deposition velocity dependence on drag coefficient is changed.

DESCRIPTION OF FACILITY AND DATA

The data used in this analysis are from 19 experiments on diffusion of zinc sulfide tracer released at an elevation of 26 meters from the 125 meter Hanford meteorological tower during neutral and stable conditions, i.e., Ri positive. The sampling is from nine arcs located at distances from the tower of 400, 800, 1200, 1600, 2200, 3200, 5000, 7000,
and 11,800 meters, respectively. The data elements fed into the analysis are integrations of exposure along each arc for each experiment, thus approximating crosswind integrations. This procedure makes 171 data elements possible, however, only 116 could be used. The most common reason for disqualification of a data element was incomplete sampling of the plume by a given arc during a particular experiment, making crosswind integration impossible in that case.

The original data contained more than 19 experiments. However, for all experiments with Ri greater than 0.15 the distribution was so ill-defined by the sampling arcs that estimates were available on only one or two arcs, and these data were not used. Two experiments exhibited Ri above the 0.215 value which is the upper asymptotic limit according to most recent turbulence similarity theory. These data were not analyzed.

For the 19 tests the mean wind speed at 15 meters varied from 2.8 to 7.2 mps.

**Mathematical Description of the Analysis**

The exposure estimation model consisted of four adjustable parameters, which is mathematically equivalent to a four dimensional domain. The four parameters are:

2. Basic value of Hay-Pasquill B for neutral conditions.
3. Linear variation of beta with Richardson number at 5.5 meters.
4. Deposition velocity.

(The model also included originally a settling velocity but when the analysis showed that a zero value sufficed, the parameter was dropped.) These quantities are all defined in a companion paper in this report.*

For an arbitrary specification of the domain values, the element of the analysis is an individual ratio, \( r_{ij} \) for each experiment, i, and each arc, j, where

\[
\frac{r_{ij}}{ij} = \frac{\text{calculated estimate}}{\text{measured estimate}}, \quad ij.
\]

The primary analysis presented in Table 1 for particular specifications of domain values is obtained by integrating all values of \( r_{ij} \) over all arcs and over all experiments for each average value (to be described below) computed. Also, a few calculations are made with the experiments divided into three stability classifications.

(a) \( 0 < Ri \leq 0.05 \).
(b) \( 0.05 < Ri \leq 0.12 \).
(c) \( 0.12 < Ri \leq 0.15 \).

The evaluation averages constitute a vector valued function of the four-dimensional domain. There are five elements in the evaluation function.

1. The mean ratio of calculated to measured crosswind integrated concentration:

\[ M = \exp \left( \frac{1}{L_A(t_{ij})} \right) \]  
(1)

Since a few of the \( r_{ij} \) were zero, these values had to be adjusted upward to an arbitrarily specified minimum value before \( M \) was computed. For these computations, the value of \( M \) is, of course, a function of this minimum value which was selected at 0.06.

2. The mean geometric deviation of individual \( r_{ij} \) from \( M \). To calculate this the individual quantities, \( d_{ij} \) are formed for each \( i \) and \( j \) from the larger of the two quantities:

\[ d_{ij} = r_{ij}/M; \text{ or } d_{ij} = M/r_{ij} \]  
(2)

Therefore, none of the \( d_{ij} \) are less than unity. The final measure of the scatter is the average of the \( d_{ij} \):

\[ s = \frac{\sum d_{ij}}{n} \]  
(3)

3. A dissemination of the 116 ratios, \( r_{ij} \), into 10 categories. These were

\[ r_{ij} < 2^{-4} \]

\[ 2^n \leq r_{ij} < 2^{n+1} \text{ } n = -4, -3, \ldots, 2, 3. \]  
(4)

\[ 2^4 \leq r_{ij} \]

4. An evaluation of the above distribution that is best described in a later section of the report where tabled results are presented.

5. A count, \( A_{ij} \) of the \( r_{ij} \) values adjusted upward for step 1 to prevent \( M \) from becoming zero.

**Physical Discussion of Analysis Parameters**

The best analyses to date have been obtained with the release height entered at its actual value into the computations. Lower values simulating initial fall of the tracer spray were used at times during the analysis when they seemed to produce better results which compensated for poor adjustment of some other parameter. But for purposes of this report the release height will be considered non-adjustable.

Since the model requires calculation of \( \sigma_z \) from application of the Hay-Fasquill equation, and since the vertical velocity spectrum in this equation is a function of height; either the variation of the turbulence with height must be part of the model or a characteristic height, \( H_c \), within the diffusion layer must be arbitrarily selected for calculation of spectral values and mean wind speed values which are assumed to apply to the whole diffusion process. Physical intuition suggested that this height should be less than the release height. In the analysis, heights that were multiples of three were tested, and best results were obtained with 15 or 18 meters for the

*See Equation 5 in "Profile-Derived Estimation of Crosswind Integrated Exposure from Elevated Releases: Method of Investigation," in this report."
characteristic height, a slight edge being apparent for the 15 meter figure.

For each experiment the value of \( \delta \) varied as a linear function

\[
\delta = B + C_1 R_i \text{ (5.5 meters).} \tag{5}
\]

Better results were achieved using the two degrees of freedom than using \( \delta \) equal 0.44 (turbulence intensity) as suggested by Pasquill\(^1\). However, more study would be required to firmly establish the dependence.

Results varied only slightly with varying values of \( \delta \), consequently, the commonly accepted value for neutral \( \delta \) of 4 was chosen for further analyses. As \( C_1 \) was varied, the mean ratio was optimized with \( C_1 \), at a value of 200 but \( C_1 \) had to be extended to a value of 900 for the scatter of the ratios to each a minimum. A value of about 600 appears at the present to provide the most reasonable results. With \( B = 4 \) and \( C_1 = 900 \), values of \( \delta \) ranged from 7 to 94 for the individual experiments. These values of \( \delta \) were used with the Hanford turbulence spectrum model,\(^*\) in the Hay-Pasquill equation.

For each experiment a separate deposition velocity of the form

\[
V_d = C_2 \frac{u_*^2}{U(2)} \tag{5}
\]

was used, where \( u_* \) is the friction velocity, computed as a function of Richardson number and mean wind speed,\(^*\) and \( U(2) \) is the measured wind speed at 2 meters. Judging from an incomplete computation sample the variable deposition velocity yielded better results than use of a constant deposition velocity for all experiments. Although several constants near the average value of \( V_d \) in Equation (6) were tried and found to be less effective than the variable \( V_d \), more evaluation is needed to definitely establish the optimum form for the deposition velocity. The variation of analysis results with variation of \( C_2 \) was quite pronounced more so than with variation of \( B \) in Equation (5). The mean ratio is best for the lower values of \( C_2 \) while the scatter is minimized for the larger values of \( C_2 \). For the present the best selection appears to be about \( C_2 = 5.0 \).

Because \( V_d \) is used in the theoretical model only in the form, \( \frac{V_d}{U_c} \), where \( U_c \) is the mean wind speed at the characteristic height, using this model of deposition velocity amounts to replacing the deposition velocity with a measure of the drag coefficient.

\[
\frac{V_d}{U_c} = \alpha \frac{u_*^2}{U(2)U_c} = C \frac{u_*^2}{U_c^2}
\]

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FIGURE 3. Correlation Between Real Plume Characteristics and the Centroid of the Real Plume

of the parameters from the 200 m data, a search was made for an alternative explanation for the correlations exhibited at 800 m. Such an alternative was found. It would appear that these correlations can be explained as an artifact of the sample duration. The amplitude of the meander of the real plume is essentially the same at 200 and 800 m in angular measurement. As a result, the linear amplitude at 800 m is 4 times greater than that at 200 m. The speed of crosswind movement of the real plume at 800 m must, therefore, appear to be 4 times that at 200 m since the period of oscillation is about the same at the two distances. The movement of the real plume during a 38.4 sec period may not be significant at 200 m, but, because of the increase in the apparent speed of the crosswind movement of the plume between 200 and 800 m, both the shape and size of the real plume may be significantly distorted at 800 m during the same period. The amount of alteration would be a function of the position of the real plume axis. The distortion of the plume would be minimum, at the extreme displacements, and it would be a maximum near the mean plume axis. Reduction of the sampling time would have reduced the apparent motion and resulting distortion.

Having found a suitable hypothesis to explain the behavior exhibited by the correlograms between the real plume position and its size and shape, it is tentatively concluded that the assumption that the real plume size and shape are independent of position is correct. At the same time, it is concluded that a sampling increment shorter than 38.4 sec is necessary for examination of the relationships between the real and apparent plumes at 800 m in order to reduce distortion caused by meandering. It is suspected that the duration of the sampling period must be decreased as the distance from the source is increased, although the distances with which this applies are yet to be determined.
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ABSOLUTE PEAK-TO-MEAN CONCENTRATION RATIOS

J. V. Ramsdell

Absolute peak-to-mean concentration ratios are defined as the ratio between a maximum short-term concentration at any given position and the time-average concentration at the mean plume axis. They are examined as a function of both position in the plume and the time ratio. A comparison between conventional and absolute peak-to-mean ratios is shown.

In discussions of peak-to-mean concentration ratios, both the peak and mean concentrations are usually assumed to apply to the same position. As a result, there are some problems differentiating between actual peak concentrations at different positions when the conventional peak-to-mean ratios are used. To alleviate this problem, another type P/M ratio is needed. A peak-to-centerline-mean ratio was suggested in the 1968 annual report.\(^{(1)}\) During the past year, further analysis of the \(^{85}\)Kr data has been directed along these lines.

In many ways, it seems advantageous to consider peak concentrations normalized to a common concentration rather than the local mean concentration. The obvious concentration for use in the normalization
of peak concentrations would appear to be the average concentration at mean plume axis. To prevent confusion, peak concentrations normalized to this axial mean concentration will be referred to as absolute P/M ratios \(P/M_L\), as opposed to conventional P/M ratios \(P/M_a\) in which peak concentrations are normalized to the local mean concentration.

An absolute peak-to-mean concentration ratio might be thought of as the product of a conventional P/M ratio and the ratio between the local and axial mean concentrations. Thus, it is to be expected that \(P/M_L\) are a function of the same variables as \(P/M_a\). A summary of absolute peak-to-mean ratios for the \(^85\)Krypton tests is presented in Table 1 by relative crosswind position and time ratio. The central value in each group is the geometric mean \(P/M_a\) for the class and the upper and the lower values are one geometric standard deviation removed from their respective \(P/M_a\). The range of values of \(T/t\) and \(y/\sigma_y\) for each group is indicated by the arithmetic mean ± one standard deviation.

The relationship between \(P/M_a\) and normalized crosswind positions is shown in Figure 1. The most striking feature of this figure is that the maximum \(P/M_a\) ratios occur at the mean plume axis. The relatively flat portion of the curves near the center of the mean plume is the result of plume meander and its extent essentially defines the limit of meandering of the axis of the instantaneous plume. As the edge of the mean plume is approached, the rapid decrease in \(P/M_a\)

<table>
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<tr>
<th>(T/t)</th>
<th>0.21 ± 0.12</th>
<th>0.70 ± 0.14</th>
<th>1.23 ± 0.16</th>
<th>1.75 ± 0.16</th>
<th>2.31 ± 0.16</th>
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<tr>
<td>3.65</td>
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<td>3.96</td>
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<tr>
<td>20.2 ± 3.9</td>
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**FIGURE 1.** Absolute Peak-to-Mean Concentration Ratios as a Function of Normalized Crosswind Position

is caused by the comparison of peak concentrations at the extreme edge of the meandering plume with the axial mean concentrations. Figure 2 shows the scatter of the actual data points about the curve for $T/t$ about 20. This scatter is representative of the scatter about each of the curves.

The differences in the relationships between conventional and absolute peak-to-mean ratios and the crosswind position are obvious in Figure 3. In this figure, the lines which increase toward the right represent conventional $P/M$, while those decreasing to the right represent absolute $P/M_a$. As can be seen, the two peak-to-mean ratios are the same at

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**FIGURE 2.** Typical Scatter of Absolute Peak-to-Mean Concentration Ratios

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**FIGURE 3.** Comparison Between Absolute and Conventional Peak-to-Mean Ratios
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Volume I of this report to the USAEC Division of Biology and Medicine covers work in the life sciences and is issued as BNWL-1650 in two parts: Biological Sciences, Part I and Ecological Sciences, Part 2. Volume II of this report covers work in the physical sciences and is issued as BNWL-1651 also in two parts: Atmospheric Sciences, Part 1; Radiological Sciences, Part 2.
FOREWORD

This report to the USAEC Division of Biology and Medicine covers work done in the fields of environmental and radiological chemistry, radiation instrumentation, radiation dosimetry, and radiation and health physics. It continues a series of reports on work initiated in 1959. Special effort has been made as last year to make the report a brief and concise technical summary of the accomplishments. For details of the work, references are given to the pertinent publications and presentations resulting from the studies.
Reports previously issued in this series are as follows:

HW-63824                  July 1, 1959 - December 31, 1959
HW-70050                  Period Ending December 31, 1960
HW-73337                  Period Ending December 31, 1961
HW-77609                  Period Ending December 31, 1962
HW-81746                  Period Ending December 31, 1963
BNWL-36 III                Period Ending December 31, 1964
BNWL-235 Part 2           Period Ending December 31, 1965
BNWL-481 Part 2           Period Ending December 31, 1966
BNWL-715 Part 2           Period Ending December 31, 1967
BNWL-1051 Part 2          Period Ending December 31, 1968
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Publications and Presentations
RADIOLOGICAL CHEMISTRY

INHALATION HAZARDS TO URANIUM MINERS

The high incidence of lung cancer and other respiratory ailments in uranium miners requires accurate characterization of the radionuclides, elemental and organic content of the uranium mine atmosphere and their correlations to lung exposure.

Elemental Content of Uranium Mine Atmosphere as a Function of Particle Size
J. A. Cooper, P. O. Jackson, T. M. Tanner and L. A. Rancitelli

Atmospheric contaminants encountered in underground uranium mines are known to present serious occupational health hazards. Evaluation of the detrimental effects of the aerosol components requires that their elemental composition be known as a function of particle size.

Airborne particulates in several locations of an operating uranium mine were sampled with both Lundgren and Andersen impactors. The impactor stages on which particles of a specific size range collected were analyzed for their elemental content by both neutron activation analysis and isotopic X-ray fluorescence. Seventeen elements were measured and include Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Mn, Fe, Zn, Se, Sr, Pb and U. Additional analysis using long-lived neutron activation products are expected to yield results for another 15 elements.
The amount of material collected ranged from a few hundredths of a microgram of Mn to several hundred micrograms of Al. The weights of most of the elements were highest on the second stage of the Lundgren impactor (3 micron median diameter particles) and lowest on the fourth stage (0.3 micron median diameter particles). The Pb concentration, however, peaked on the first Lundgren stage (greater than 10 micron diameter particles) while the Zn concentration peaked on the fourth stage along with the carbon from diesel exhaust. This agreed well with the visual observation that most of the ore dust was deposited on the first two Lundgren stages while the diesel exhaust was deposited mostly on the fourth stage and backup filter. This suggests that the Zn may be associated with the diesel exhaust.

Inter-element ratios and correlations are being examined to determine whether specific elements are associated with a particle of a particular size and to relate the mine conditions to those in the animal test chamber. Initial comparisons of elemental ratios showed some of them to vary considerably from one impactor stage to the next. Many of the elements varied by as much as two- or threefold relative to Al with the Zn to Al ratio increasing 25 times over the four stages of the Lundgren impactor. On the other hand, the Mg to Al ratio remained about constant over all four stages of the impactor and the backup filter. The Cl to Al ratio was also constant over the first three stages but increased by two- and tenfold on the fourth stage and backup filter, respectively. The Na to Al ratio followed a similar pattern but showed a smaller increase on the backup filter. The additional neutron activation analysis data and a complete correlation study of all of the elements will provide a greatly improved understanding of the contributions of various sources of uranium mine aerosols.
Photograph of the Lundgren Impactor Stages and Backup Filter After Sampling About $1.8 \times 10^4$ Liters of Ambient Air and About $2.4 \times 10^3$ Liters of Exhaled Breath from Air in an Operating Uranium Mine

Neg 715759-1
Determination of Uranium Mine Aerosol Constituents Retained in Human Lungs
P. O. Jackson and J. A. Cooper

The inhalation of dust particles by mine workers is a contributing factor to pulmonary disorders. The potential hazards associated with dust inhalation is greater in uranium mines because of radon daughters which attach themselves to dust and the radiation from uranium and its long-lived daughters. The pulmonary deposition, location and retention of aerosol particles is a function of their size, density and associated aerodynamic properties.

The objectives of this study were to determine the fraction of the various types and sizes of aerosol particles which are retained in the respiratory system.

An instrument for collecting and sorting exhaled particulates has been developed and used in an operating uranium mine to determine pulmonary tract retention of stable and radio-element constituents of a uranium mine aerosol. It consists of an "air purification train" into which the subject exhales, and this air is rapidly carried through a Lundgren cascade impactor which separates particulates into 10, 3, 1 and 0.3 micron diameter groups. The impactor is followed by a backup filter which removes smaller particles. Sampling periods of two man-hours were employed for collecting sufficient material for elemental analyses while five minute sampling periods in a rather poorly ventilated mine area were employed for short-lived radon daughter studies. The stages of the cascade impactor and the backup filter are analyzed for elemental content by neutron activation analysis and isotopic X-ray fluorescence. In the case of sampled mine air, the exhalates consisted of light-colored ore dusts observed on the first stages (larger sizes) and black carbon from diesel exhaust deposited mostly on the fourth stage and backup filter. Elemental analyses are not yet complete; however, the aluminum and vanadium, which were major components of uranium mine dust, and chlorine which was primarily associated with carbon loaded (diesel) stages in mine air samples are complete. The respiratory tract retention is higher for these elements when associated with particles 0.3 micron or greater than for those less than 0.3 micron. A higher fraction of aluminum was retained on the 1-3 micron particulates than on either 0.5 or 10 micron sizes. Chlorine was significant only on particulates less than 1 micron. Since it was higher in exhaled air than in normal mine air, it was apparently associated with biological components of breath. Analyses in progress will provide data for about 20 additional elements and permit a fuller evaluation of the respiratory tract fixation of dust constituents.
In the radon daughter experiments, the impactor stages and backup filter were counted with an alpha spectrometer a few minutes after sampling both exhaled breath and mine air. In both mine air and expired air the fraction of the radon daughter activity on particles larger than 0.3 micron was less than 0.3%. For both samples the radioactive equilibrium was essentially complete for particles collected on the backup filter, which was to be expected in the poorly ventilated study area.
Characterization of Radon Daughter Components of Uranium Mine and Animal Test Chamber Aerosols

J. A. Cooper, P. O. Jackson, H. G. Rieck and R. W. Perkins

The major radiation hazard associated with uranium mining results from pulmonary deposition of radon daughters. The radon daughter concentration must be known as a function of particle size and time to relate the conditions in our animal test chambers to those in a uranium mine and to evaluate the relative dose to the lung. A sensitive research instrument based on simultaneous alpha and gamma-ray spectrometry was developed...
and applied to the continuous measurement of the radon daughter concentrations as a function of particle size in the atmosphere of animal test chambers and in different locations of an operating uranium mine. Different Andersen impactor stages were used as pre-separators to select a given size particle which was then collected on a membrane filter and the activity measured. The pre-separation of large particles allowed continuous operation with a minimum of self-absorption effects by limiting the material collected on the filter to the fine particles with which essentially all of the radon daughter activity was associated. The amount of alpha self-absorption was determined by comparing the intensity of the 7.69 MeV alpha peak of $^{214}\text{Po}$ with that of the 0.609 MeV gamma-ray peak from $^{214}\text{Bi}$. The self absorption correction was less than 10% for collection periods of up to two hours in a dusty mine atmosphere.

The $\text{RaA} (^{218}\text{Po})$ and $\text{RaC} (^{214}\text{Po})$ were measured directly from their 6.00 MeV and 7.69 MeV alpha peaks while the $\text{RaB} (^{214}\text{Pb})$ and $\text{RaC} (^{214}\text{Bi})$ were measured from their gamma-ray peaks at 0.395 and 0.352 MeV and 0.609 MeV, respectively. Equations were derived to relate a series of these measurements to the radionuclide concentration in the mine air as a function of time. The radon daughters associated with different sized particles were studied by using different Andersen impactor stages as pre-separators. The elemental content of the separated particles is being measured to establish the correlation with the radionuclide measurements and the distribution of elements on the stages of a Lundgren impactor.
Rates of Deposition of Airborne Radionuclides
In the Alaskan Environment

The atmospheric deposition rates, routes, biological uptake rates and concentrations of fallout radionuclides and their stable element counterparts are being investigated in the terrestrial and marine environments, with emphasis on the food chains that consistently lead to high radionuclide concentrations.

Radionuclides in the Alaskan Arctic Regions
C. E. Jenkins and James C. Langford

Relatively few studies have been made of the behavior of radionuclides other than \(^{55}\text{Fe}\) and \(^{137}\text{Cs}\) in the Arctic regions. This program includes a study of the movement, uptake and relative behavior of 18 radionuclides from both natural and man-made sources. In addition to investigating the well known lichen-caribou-man (wolf) food web other possible avenues for uptake of radioelements by man are being studied to determine their relative importance. These measurements provide a basis for estimating the radiation dose received by resident Eskimos. These studies also provide basic information which will be extremely valuable in understanding deposition rates, movement, biospheric uptake and ultimate fate of toxic trace elements since many of the radioelements which are being investigated, e.g., \(^{65}\text{Zn}, \, ^{110}\text{Ag}, \, ^{125}\text{Sb}\) and \(^{210}\text{Pb}\), have stable toxic elemental counterparts from a wide spectrum of anthropogenic sources.

One important route by which radioelements may enter man is through his water supply. This can occur to a limited degree from drinking water but the major route is through aquatic organism concentration processes and the subsequent use of aquatic organisms as food. Measurements of the summer water supply of the Anaktuvuk Pass Eskimos, which consisted of a nearby creek (headwaters of the John River) showed radionuclide concentrations which were somewhat higher than those in the Columbia River upstream from the Hanford Plant. This pathway may be of greater importance in the Arctic region where lake and stream water are used directly for drinking purposes and local aquatic organisms are consumed.
Animals are the important source of radionuclides in the Alaska natives' diet. This is particularly evident from $^{55}$Fe concentrations which have been measured. The yearly changes in $^{55}$Fe concentrations since the 1961-1962 nuclear test series in various Arctic plants and animals are shown in the accompanying figures. Slow growing lichens show concentrations relatively constant since 1966 which may be partly due to additions from the Chinese nuclear testing while fast growing seasonal willows and grasses show sharp decreases through 1966.

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**Anaktuvuk Pass Water Sample**

**August 1, 1970**

**dpm $10^3$**

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Filters</th>
<th>Suspended</th>
<th>Cation (b) Bed</th>
<th>Anion (c) Bed</th>
<th>Al Oxide Bed</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{22}$Na</td>
<td>---</td>
<td>1.9 ± 0.35</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>3160 : 200</td>
<td>204 : 10</td>
<td>---</td>
<td>88.8 ± 16</td>
<td>---</td>
</tr>
<tr>
<td>$^{54}$Mn</td>
<td>14.9 : 3</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>8.73 : 1</td>
<td>4.74 ± 0.45</td>
<td>---</td>
<td>1.30 ± 0.3</td>
<td>---</td>
</tr>
<tr>
<td>$^{65}$Zn</td>
<td>45.2 : 6</td>
<td>67.2 ± 5</td>
<td>---</td>
<td>15.8 ± 3</td>
<td>---</td>
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<tr>
<td>$^{95}$Zr-Nb</td>
<td>108 : 14</td>
<td>18.7 ± 1.8</td>
<td>29.3 ± 1.8</td>
<td>81.8 ± 14</td>
<td>12</td>
</tr>
<tr>
<td>$^{106}$Ru</td>
<td>---</td>
<td>43.1 ± 7</td>
<td>147 ± 6.4</td>
<td>446 ± 12</td>
<td>---</td>
</tr>
<tr>
<td>$^{125}$Sb</td>
<td>99 : 5</td>
<td>34.9 ± 5</td>
<td>194 ± 4.7</td>
<td>577 ± 6</td>
<td>---</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>22.0 : 2</td>
<td>16.0 ± 2.1</td>
<td>---</td>
<td>7.67 ± 2</td>
<td>---</td>
</tr>
<tr>
<td>$^{208}$Tl</td>
<td>7.83 : 2</td>
<td>---</td>
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<tr>
<td>$^{214}$Bi</td>
<td>72.6 : 6</td>
<td>53.9 ± 6</td>
<td>---</td>
<td>---</td>
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</tr>
</tbody>
</table>

*a. 551 liters of creek water  
b. Two 1 in. thick cation beds  
c. One 1 in. thick anion bed*
55Fe Specific Activity in Caribou and Reindeer

55Fe Specific Activity in Arctic Plants
Radionuclide Distribution in an Alaskan Forest Environment
C. E. Jenkins

Information on radioelemental behavior in a forest system can add considerably to our knowledge of transfer and exchange processes. Preliminary investigations of radionuclide concentrations and the environmental factors involved in their large-scale movement were made in the Chugach Forest near Anchorage, Alaska. The upper Kenai River drainage basin has an annual precipitation rate of about 50 inches. This is about five times that at Anaktuvuk Pass. Since the deposition rates and subsequent movement of radionuclides is known to be related to the hydrology of an area, major differences in concentration and biogeospheric uptake between the tundra and forest environs were expected. The distribution and concentration of radioelements suggest that retention times in the flora component of the Alaskan forest are short in comparison to those of the Arctic tundra. Kenai lichen concentrations contain less long-lived and considerably more short-lived man-made fallout radionuclides than equivalent Anaktuvuk Pass flora.

Investigations of the Kenai soil component in the muskeg show a distinctive behavioral pattern of various radionuclides. Concentrations of the natural radionuclides $^{40}$K, $^{232}$Th, and $^{226}$Ra are lower in the upper one inch of the heavy root zone than in the deeper zone. Other radionuclides have higher concentrations in the surface inch of soil. Thus, approximately 96% of the $^{60}$Co, 67% of the $^{95}$Zr-$^{95}$Rh, 100% of the $^{106}$Ru, 54% of the $^{125}$Sb, 96% of the $^{137}$Cs, and 100% of the $^{144}$Ce are found in the first inch of soil. For $^{54}$Mn the concentrations increase with soil depth for at least the first four inches. Zinc-65 concentrations are high at the surface, then drop but gradually increase again with depth.

River bottom sediments have predominantly those radioelements found in the upper inch of soil. The clay sediments have higher concentrations of $^{60}$Co, $^{125}$Sb and $^{137}$Cs than the sand sediments. Very little $^{54}$Mn was found in the river bottom sediments; however, it was shown to be present in the cationic fraction of water samples. These observations indicate that $^{54}$Mn is rather highly mobile and reaches the plant root zone rapidly. It would thus be expected to find its way into plants through the roots.
Radionuclide Concentrations in North Pacific Salmon as a Function of Latitude
C. E. Jenkins and James C. Langford

A study was initiated in 1967 to define the distribution of gamma-emitting radionuclides and $^{55}$Fe in five mature salmon species sampled at several locations along the western coast of the United States. Immature salmon have also been measured to provide a basis for comparing river, estuarine, and oceanic food webs. In addition, information regarding the uptake of these various radioelements relative to their stable elemental counterparts is being
obtained. Radionuclide concentration in salmon appears to be the result of a complex interplay of several parameters. Among the most important are the oceanic migratory pattern of the individual salmon species, their life span, food web, and the relative dependence of their food web on elements supplied by river waters. Past and present studies confirm that some fallout radionuclides are preferentially taken up by surface feeding organisms relative to their stable elemental counterpart by factors approaching $10^3$.

The group of fallout radionuclides most effectively concentrated by salmon on a long-term basis are the elements $^{54}$Mn, $^{55}$Fe, $^{60}$Co, and $^{65}$Zn. These radionuclides may be concentrated by factors of $10^3$ to $10^5$ over seawater concentrations. The highest $^{55}$Fe and $^{54}$Mn concentrations observed in salmon are in the northern latitudes. Fe-$^{55}$ concentrations are 10 to 30 times higher in northern compared to southern salmon. This physical or chemical phenomenon appears related to the food web and for its relative dependence on fallout. Plots of $^{54}$Mn, $^{60}$Co, and especially $^{65}$Zn concentrations in salmon versus latitude show the increase in concentration approaching the mouth of the Columbia. In 1968, Columbia River waters were approximately one, three, and several orders of magnitude higher in $^{54}$Mn, $^{60}$Co, and $^{65}$Zn, respectively, than oceanic waters. Since that time all of the Hanford reactors which discharge their cooling water directly to the Columbia River have been shut down and the concentration of the Columbia River radionuclides has steadily declined.

A very slight dip in $^{55}$Fe concentration and its specific activity which occurs in salmon caught near the Columbia River may be associated with the low $^{55}$Fe specific activity of its waters. This is verified by the low $^{55}$Fe specific activities in immature salmon which spent their lives in the river. The high concentrations of $^{60}$Co and $^{65}$Zn indicate the extent of the influence of the Columbia River waters. Ra-$^{226}$ concentrations in immature Columbia River salmon show a dip similar to $^{55}$Fe concentrations and specific activities. We have observed that $^{226}$Ra which originates from soil or sediments is enriched in areas of high river output, in areas of ocean upwelling, and in areas where the seas are shallow. The Columbia River and the spring and summer upwelling along the Oregon coast may be responsible for high $^{226}$Ra levels in salmon from the southern regions. High $^{226}$Ra levels in Bering Sea salmon may be a result of shallow seas which enable $^{226}$Ra from the sediments to reach the euphotic levels of the ocean more quickly. The concentrations of $^{137}$Cs in salmon peak at the mid-latitudes where the fallout rate is the greatest and drop rapidly.
southward, closely following concentrations of 137Cs in the seawater.

Annual measurements of radio-nuclides in salmon from Kotzebue and the Columbia River have shown rapid declines. Kotzebue salmon show 55Fe and 54Mn declines that approximate the stratospheric input rate. Cs-137 levels are still rising, apparently due to its long half-life and resident time in the oceans. Co-60 levels appear to be leveling off as would be expected for this intermediate half-life radionuclide. Columbia River salmon are showing rapid yearly declines in 54Mn, 55Fe, 60Co and 65Zn. The 55Fe decline is similar to that observed for fallout. Mn-54 60Co and 65Zn are falling at a much more rapid rate, indicative of the Hanford facility shutdowns.

Neg 711283-2
Trace Element Content of Alaskan Caribou and Lichen
L. A. Rancitelli

The transport and distribution of radionuclides in the Alaskan environment has received a great deal of attention due to the unusually high radionuclide concentrations in certain principal food chains. While significant advances have been made in understanding the transport of fallout radionuclides from the air through the marine and terrestrial food chains to the native population, the effects of dilution of these radionuclides by their stable element counterparts have received very
little attention. Thus, to help elucidate the mechanisms responsible for radionuclide transport through the Alaskan food chain, a study is being conducted of 15 elements: Ag, As, Br, Co, Cr, Cs, Fe, Hg, K, Na, Rb, Sb, Sc, Se and Zn, in lichen and caribou, important links in a major chain leading to man. The measurements are made by instrumental neutron activation analysis using 200 mg aliquots of caribou liver, muscle and kidney tissue and lichen samples which are simultaneously analyzed for their radionuclide content. The specific activity data obtained from the trace element study and the radionuclide analysis permit the interpretation of the radionuclides in the food web to be characterized.

Preliminary results of the trace element analyses of caribou reveal some trace element patterns similar to those observed in marine and aquatic organisms. The majority of the silver is contained in the liver. However, the high silver content of liver was not accompanied by high Se concentrations as previously noted in salmon. The caribou kidney and liver contained far higher cobalt concentrations than the muscle tissue. The muscle tissue contained the highest levels of zinc, in marked contrast to marine organisms where the highest concentrations are found in the liver and kidney. The mercury content of the caribou tissue samples was below 0.5 ppm, and in muscle tissue was below 0.02 ppm on a fresh weight basis. The interpretation of the elemental analyses of lichen samples is difficult, since it is obvious from the Sc, Sb and Fe concentrations that a significant amount of soil material was contained in the samples. However, several important observations are apparent in spite of the soil component: the Se content (0.03 to 0.08 ppm) is below the range of 0.2 to 0.8 ppm recommended by the Research Council's Agricultural Board as necessary for domestic livestock food. Selenium is suspected to be an essential element and the effects of Se deficiencies on livestock are well documented. The mercury and arsenic contents of lichen are also low, 0.05 ppm and <0.5, respectively.
<table>
<thead>
<tr>
<th>Tissue</th>
<th>Au</th>
<th>Be</th>
<th>Br</th>
<th>Ca</th>
<th>Cr</th>
<th>Cs</th>
<th>Fe</th>
<th>Mg</th>
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The rates and mechanisms of the chemical and mixing processes in the ocean are being investigated by use of the ocean's natural elemental and radionuclide tracers. These studies as a function of depth over wide areas of the ocean are allowing the determination of factors governing vertical diffusion and the determination of oceanographic pollutant concentrations and their pathways to man.

Latitudinal Variation of \(^{7}\)Be Fallout in the Pacific Ocean
W. B. Silker

Data presented in another section of this report show a relationship between the total ocean water column inventory of \(^{7}\)Be and its surface concentration. A similar relationship was found for \(^{90}\)Sr-Nh. Thus the surface water concentrations of these radionuclides can be used to evaluate both their transfer rate across the air-sea interface and the total input from recent fallout.

The accompanying figure shows the surface water concentrations of \(^{7}\)Be in samples collected on three north-south transects of the Pacific. The samples collected two years apart along the eastern Pacific Ocean boundary show remarkable consistency in their latitudinal \(^{7}\)Be radionuclide distribution. Of particular interest are the very high concentrations found at about 10°N latitude along the lines and the minor maximum at the same latitude in the mid-ocean track. The air-to-sea transfer rate near the eastern Pacific Ocean boundary is some 50-fold greater at 10°N latitude than at the equator. This latitudinal distribution differs drastically from those found in the Health and Safety Laboratory 80th Meridian studies for \(^{90}\)Sr or \(^{137}\)Cs, which both increase regularly from minimum equatorial values to a maximum at 40-50°N. The \(^{7}\)Be maxima observed at 10°N are probably at least partially due to wet deposition by precipitation. Total annual rainfall, both along the 180th meridian and at Pacific Coast stations, exhibits a very pronounced maximum between 5-10°N, decreasing to a minimum at both the equator and mid-latitudes. Weather satellite data reveal persistent and highly developed cloud cells over the eastern Pacific with maximum cloudiness about 8°N and decreasing cloudiness both north and south. The southern hemispheric data also are interesting because of the much higher concentrations shown for mid-ocean samples when compared with those taken along the eastern margin. Surface samples taken on a track between 25-30°S showed quite high concentrations.
west of 130°W longitude with decreasing values to the eastward. This also is the western boundary of the cold Peru Current. Almost desert conditions of low rainfall prevail in the eastern equatorial south Pacific because of the lack of water transfer from the cold ocean surface to the atmosphere.

The excellent agreement between surface water radionuclides and the rainfall and cloud distributions strongly suggests wet deposition to be an important mode of transfer of radionuclides from the atmosphere in the regions of high 7Be concentration. The important fact from these data is that certain areas of the tropical ocean receive much higher quantities of fallout, at least at certain times of the year, than had previously been recognized.

Neg 715485-1

7Be Concentrations in Pacific Ocean Surface Water
The Physical Form of Radionuclides in Pacific Ocean Water
W. R. Silber

The application of $^7$Be and fission products as tracers of oceanographic mixing processes requires that they act conservatively, being distributed within the water column only by the physical forces acting on the water mass. Many investigators have either theorized or found by measurement that many fission products, including $^{95}$Tc-Nb, $^{106}$Ru and $^{144}$Ce, were largely associated with oceanic particulates, while others, conversely, found large percentages to be "soluble."

In a study of this question, data from 120 samples of North Pacific Ocean water taken from surface to 100 meter depths were analyzed as a function of depth with respect to the radionuclide fraction associated with the particulates. These samples were taken over a large oceanic area extending from the equator to the Gulf of Alaska and the continental coastline to approximately 180°W during 1968-1971. In the following table the percentages of $^7$Be, $^{95}$Tc-Nb, $^{106}$Ru and $^{144}$Ce which are retained on 0.3μm pore size filters are presented as a function of depth. The percentages associated with particulates are given in terms of the median, the average and the maximum.

These data show that the fraction of these radionuclides associated with particulate material is generally 10% or less. They also show that a rather constant fraction of these radionuclides is associated with the particulate phase throughout the top 100 meters of the water column. Therefore, the particulate fraction, regardless of physical form, is apparently behaving within this 100 meter deep water column in the same manner as a "soluble" radionuclide. The radionuclides of this group therefore, are excellent tracers for measurement of vertical diffusion and advective transport in the open oceans.

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</table>
The Relationship of $^7$Be in Surface Water to That in the Water Column

W. B. Silker

Beryllium-7, produced in the atmosphere by cosmic-ray spallation of nitrogen and oxygen, may serve as a tracer for measurement of vertical eddy diffusion rates within the upper mixing layer of the oceans. Owing to its relatively short half life ($T_{1/2} = 53$ days), it is possible to obtain the inventory of $^7$Be by sampling to limited depth, and this inventory represents the steady state addition by transfer from the atmosphere to the sea surface. Thus, by determination of the concentrations contained by a number of water columns, representative of the total ocean, the cumulative input of atmospheric aerosols to the ocean may be estimated. Limitations of time and logistics have precluded such an ambitious project. If, however, a relationship exists between the concentration of $^7$Be in surface water and the water column inventory, the estimation of the transfer across the air-sea interface will be greatly simplified.

The $^7$Be production rate varies with both latitude and altitude. Surface air concentrations vary seasonally about a factor of three with the northern hemisphere maximum occurring during June and the minimum in December. If it is assumed that the transfer of $^7$Be across the air-sea interface is proportional to the atmospheric concentration, then its concentration in surface water should closely parallel the seasonal fluctuations in the atmosphere, and the water column inventory would be a function of the total transfer during the preceding several months.

Data derived from twenty-one profiles sampled from 1968-71 north of the equator in both the Atlantic and Pacific oceans are shown in an accompanying figure. These data are presented as the variation with time of the ratio of the surface concentration of $^7$Be to its water column inventory. A strong seasonal change of this ratio is apparent. Highest ratios occur during July and August following the period of highest air concentrations, and hence the highest input, when the total water column inventory of $^7$Be is at its highest value. Low ratios occurring during the early spring months reflect the combination of both minimum airborne radionuclide concentration and the high $^7$Be inventory accumulated during preceding, higher input periods.

The deviation of the individual values from the curve in the figure was $11.2\%$ at the 95\% confidence level. From these data it appears that the input rate of $^7$Be, and presumably of other atmospheric contaminants, can be determined within about 11\% simply by monitoring the surface water concentration. This is remarkable, considering that the surface concentrations ranged from 200-900 dpm/m$^3$, and that
the profiles were sampled over a three-year period across a wide range of longitude and from 8°-45° north latitude. Future work is required to determine the shape of the curve during months that have not yet been sampled, and perhaps more detailed depth sampling is needed to define any fine structure in the depth distribution of \(^{7}\text{Be}\) which would permit a more accurate assay of the water column inventory.

Seasonal Change of the Ratio \(^{7}\text{Be}\) Water Column Inventory to Its Concentration in Surface Water
Determination of Mercury in Seawater
D. E. Robertson

Because of the great concern for mercury contamination of the marine environment by man's activities, it is essential that the mercury concentrations and distribution in the ocean be determined. Since the mercury levels in the oceans appear to range between 0.01 to 0.3 µg Hg/1, it is necessary to preconcentrate the mercury to meet instrument sensitivity and to minimize interferences from other elements. A rapid, simple procedure for quantitatively measuring mercury in 100-200 ml of seawater has been developed, based on its amalgamation with copper metal and subsequent analysis by flameless atomic absorption spectrophotometry. Seawater (usually acidified to pH 1.5 with high purity hydrochloric acid) is passed through a 1.0 cm diameter quartz tube containing a 1.5 cm long column of 50 mesh, granular, copper metal* at a flow rate of 5 ml per minute. The copper is rinsed with a few ml of purified water and a few ml of acetone, then thoroughly dried by passage of a stream of clean air. The quartz tube is placed in a furnace preheated to 250°C.

The volatilized mercury is swept, with a stream of N2 gas, through an optical cell placed in the light beam of an atomic absorption spectrophotometer. Mercury is measured by absorbance at the 2536.5 Ångstrom resonance line. The yield and precision of this method, determined by six replicate analyses, with radioactive 205Hg tracer, was 96.7 ± 0.9%. Preliminary analysis of deep Pacific Ocean water collected several hundred miles off San Diego, California gave mercury values ranging between 0.17-0.20 µg Hg/1.

When methyl 205Hg chloride tracers were used to evaluate the procedure, a yield of only 4.0 ± 0.2% was recorded. Thus, this analytical method is almost completely insensitive to methyl mercury. It has been shown, however, that methyl mercury can be readily photo-oxidized and subsequently retained on the granular copper metal column. Inorganic and methyl mercury can thus be separately measured by first extracting and measuring the inorganic mercury. The effluent seawater is then subjected to ultraviolet irradiation to oxidize the methyl mercury. Then the seawater sample is reanalyzed for mercury. Mercury observed in the second analysis is thus a measure of the Hg in the methyl mercury form and possibly in other organo-metallic compounds.

* Minco copper metal, fine granular; Midwest Instrument Co., Delafield, Wisconsin.
Storage of seawater samples presents numerous problems due to losses of certain elements by adsorption onto container surfaces. Storage conditions for seawater collected for mercury analysis were evaluated by spiking seawater with $^{203}$Hg$^{++}$ and methyl $^{203}$Hg chloride tracers and storing at pH 1.6 and 8.0 in polyethylene bottles. At pH 8.0 both the $^{203}$Hg$^{++}$ and methyl $^{203}$Hg chloride are rapidly and nearly quantitatively lost from solution by adsorption onto the polyethylene containers. However, when the pH is reduced to 1.6 with hydrochloric acid no serious adsorption of these tracers occurs. Our standard method of handling seawater samples consists of acidifying the seawater to pH 1.5 to 1.6 at collection time with high purity hydrochloric acid and storing in acid-cleaned polyethylene bottles; thus the adsorption problem is minimized.

![Graph](Neg 713989-7)

**Adsorption of $^{203}$Hg$^{++}$ and Methyl $^{203}$Hg Chloride From Sea Water at pH 1.6 and 8.0 onto Polyethylene Container Surfaces**
Multielement Studies of Micro and Macro Marine Organisms
L. A. Rancitelli

The trace element content of Pacific salmon, phytoplankton, zooplankton and macro algae has been extensively characterized in an attempt to understand the pathways and biological uptake of radionuclides and pollutants in the marine environment. Samples of muscle and liver from five species of Pacific salmon have been analyzed by a multielement technique of instrumental neutron activation analysis. The concentrations of 14 elements--Na, K, Cs, Rb, Hg, Cr, Fe, Zn, Co, Sc, Sb, Se, Ag and Br--have been characterized in thirty-five salmon. Relationships between the alkali metals K, Rb and Cs were established which indicate that the radiologically important element Cs is preferentially assimilated relative to K and Rb. The Rb to K ratios in salmon tissue and seawater are identical. The simultaneous determination of 14 elements in each tissue specimen permitted a statistical analysis of the data which revealed other trace element correlations.

A normal bivariate distribution was assumed for pairs of elements, and the null hypothesis that the correlation coefficient is different from zero was tested. The statistical analysis revealed several interesting interelemental relationships. The correlation coefficient for the Se-Ag content of liver of 0.823 is different from zero at a significance level of $10^{-6}$ for the number of samples considered, indicating only one chance in a million of a random correlation. A number of other radiologically important elements were found to have correlations at the 0.01 level of significance in salmon liver and muscle. In addition to the Rb-K relationship noted previously, these include Fe-Na, Zn-K, Fe-Cs, Zn-Br, Br-Co, Co-Zn, and Co-Fe in muscle and Fe-Na, and Ag-Na in liver. A comparison of the elemental abundances in phytoplankton, zooplankton, macro algae and salmon muscle indicates none of the elements considered in this work are substantially concentrated in the higher trophic levels of the marine food chain.
The multi-institutional GEOSECS program is designed to measure a large group of ocean constituents and hydrographic parameters during transects of the world's major oceans, with the aim of better understanding the geochemistry and mixing dynamics of the oceans. Two preliminary intercalibration and testing cruises, one in the N.E. Pacific and another in the N.W. Atlantic, have been completed in preparation for the main GEOSECS expeditions which will begin this summer in the N.W. Atlantic Ocean. Battelle-Northwest has participated in both of these intercalibration cruises, measuring trace element, $^7$Be and fallout radionuclide concentrations in depth profiles. During the GEOSECS program these measurements will help characterise the ocean’s geochemistry, determine the worldwide oceanic deposition of radionuclides and define the vertical mixing rates above, through and below the main thermocline. The results of these preliminary studies of trace element and radionuclide measurements are summarized.

Northeast Pacific Ocean Station
D. E. Robertson


Northwest Atlantic Ocean Station
D. E. Robertson and W. B. Silker

In August, 1970 a second pre-GEOSECS (Geochemical Ocean Sections Study) intercalibration and testing cruise was conducted in the Northwest Atlantic Ocean aboard the R/V Knorr. A station at 36°N latitude, 68°W longitude was occupied during which vertical sampling was conducted to determine the depth distribution of various ocean constituents. The vertical distributions of $^7$Be, $^{95}$Zr-Nb, $^{106}$Ru and $^{144}$Ce were measured utilizing large-volume sampling techniques and high sensitivity gamma-ray spectrometry. These measurements have been summarized in the 1970 Annual Report BNWL-1551, Vol. II, Part 2.

Sea water samples were also collected for interlaboratory comparisons of trace element concentrations at 29 depths between the surface and the ocean floor in modified 30 liter Niskin bottles. The unfiltered seawater samples were acidified to about pH 1.5 with high purity HCl and stored in polyethylene bottles. Aliquots of the seawater samples were divided among Battelle-Northwest Laboratory, Woods Hole Oceanographic Institution and Yale University.
Instrumental neutron activation analysis was employed to measure the concentrations and depth distributions of Zn, U, Co, Cs, Sb and Sc. This method consists of evaporating 25 ml aliquots of seawater and neutron activating the dried salts, together with appropriate standards, to an integral thermal neutron exposure of about $10^{18}$ n/cm$^2$. The samples were stored for a month and then counted on a multidimensional gamma-ray spectrometer and Ge(Li) diode detector. The samples were analyzed in duplicate about one month apart. The U, Sb and Cs were very homogeneously distributed throughout the entire water column. Unlike the Pacific pre-GEOSECS station, the Zn, Co and Sc distributions showed no systematic variation with depth, even though large scattered variations were observed. It appeared that several of the Co values represented contamination introduced during the collection or storage of the samples. The results of the Woods Hole and Yale groups are not yet available, so no interlaboratory comparisons of the Atlantic Ocean samples can be reported at this time.
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*Sea water samples obviously contaminated during collection and storage.*
IN-CLOUD SCAVENGING

Several inert tracers were released into precipitating clouds near Quillayute, Washington and St. Louis, Missouri in order to determine precipitation scavenging efficiencies and aerosol and hydrometeor trajectories. Rain samples obtained from networks of rain collectors are being analyzed for the tracer materials by neutron activation techniques. Concentrations of the cosmogenic radionuclides $^{24}$Na, $^{38}$Cl, and $^{39}$Cl were measured in large volume rain samples to determine scavenging efficiencies and elapsed times between scavenging and arrival of the precipitation at the ground. The concentrations of these cosmogenic radionuclides in argon and in air filter samples are also being measured to determine atmospheric production rates.

Precipitation Scavenging of Tracers Released into Rainstorms Near Quillayute, Washington

J. A. Young, W. E. Davis,
B. C. Scott, W. G. N. Slinn,
T. M. Tanner, C. W. Thomas,
J. M. Thorp and N. A. Wogman

Indium and lithium tracers were released from an aircraft at 5400 feet into precipitating clouds southeast of the Quillayute airport on March 11, 1971 in order to determine precipitation scavenging efficiencies and also aerosol and hydrometeor trajectories. Samples of rain were collected from the grid of 31 rain samplers, were frozen using dry ice and returned to the Richland laboratory for analysis.

Samples 15 through 38 and four reagent blanks were analyzed for lithium by atomic absorption spectroscopy. The concentrations were all below the detection limit (0.01 ppm Li), so none of the remaining samples were analyzed for lithium.

Indium concentrations in the samples were determined by neutron activation analysis. Samples and standards were irradiated at the Washington State University 1 megawatt TRIGA III-fueled, water moderated, research reactor at a position with a neutron flux of approximately $8 \times 10^{12}$ neutrons per cm$^2$ per second. Most of the samples contained measurable amounts of indium.

Large volume rain samples were collected at the Quillayute airport and at Rayonier's lumber camp, Camp Hoko, throughout the rainstorm for analysis for the cosmogenic radionuclides $^{24}$Na (15 hr), $^{38}$Cl (37 min), and $^{39}$Cl (55 min). There did not appear to be any detectable systematic difference in the concentrations of the cosmogenic radionuclides between Quillayute and
Hoko. When a water droplet or ice crystal begins to grow primarily by coalescence, its radius to volume ratio decreases and the rate of capture of newly formed cosmogenic radionuclides by the growing hydrometeor should rapidly become negligible compared to the cosmogenic radionuclide activity of the hydrometeor. The cosmogenic radionuclides on the hydrometeor should then begin to decay away and the $^{39}\text{Cl} /^{38}\text{Cl}$ ratio should increase with time at a rate determined by the $^{38}\text{Cl}$ and $^{39}\text{Cl}$ half-lives. Therefore, the hydrometeor lifetime can be calculated from the $^{39}\text{Cl} /^{38}\text{Cl}$ ratio of the precipitation, assuming that the $^{39}\text{Cl} /^{38}\text{Cl}$ activity ratio on the droplet or ice crystal was at the equilibrium value of 1.5 at the beginning of growth by coalescence. The hydrometeor lifetimes calculated in this manner varied from nearly zero to 99 minutes, and did not appear to be significantly different between Quillayute and Hoko. The time showed a tendency to decrease throughout the storm.

On April 16, 1971 indium, gold, iridium, and silver iodide were released into precipitating clouds near the Quillayute airport. Indium and silver iodide were released by means of pyrotechnic flares, while the iridium and gold were released by burning heptane solutions of iridium and gold in aerosol generators. Rain samples were collected in the same manner as in the previous tracer release of March 11, 1971, except that a slightly larger network of 90 samplers was used. The samples have been analyzed by neutron activation for indium and gold. The samples are being analyzed at the present time for iridium and silver.

Large volume rain samplers were also collected for cosmogenic radionuclide analysis at Quillayute and Camp Hoko on April 16, 1971. Both the $^{38}\text{Cl}$ and $^{39}\text{Cl}$ concentrations and the $^{38}\text{Cl}$ to $^{39}\text{Cl}$ ratios were appreciably higher at Hoko than at Quillayute. The hydrometeor lifetimes, calculated in the manner described above varied from nearly 0 to 58 minutes at Quillayute and from 41 to 94 minutes at Hoko. Of four rainstorms studied in early 1971, this was the only one in which there were significant differences in the cosmogenic radionuclide concentrations between Quillayute and Hoko.
Rain Collection Network on the Olympic Peninsula of Washington State
Cosmogenic Radionuclide Production Rates in Argon from 3 to 19 km
J. A. Young, C. W. Thomas and N. A. Wogman

For the past four years the concentrations of the cosmogenic radionuclides $^{24}$Na (19 hr), $^{38}$Cl (37 min) and $^{39}$Cl (55 min) have been measured in air filter samples collected by aircraft. These radionuclides are produced at a relatively constant rate in the atmosphere by spallation reactions of cosmic-rays with atmospheric argon. In order to determine whether the measured cosmogenic radionuclide disintegration rates corresponded to the actual production rates, metal spheres were lined with rubber meteorological balloons, filled with argon under pressure, and flown at altitudes ranging from 3 to 19 km. At the end of each flight the argon was vented through a charcoal impregnated filter. The filters and rubber liners were then composited and the cosmogenic radionuclide concentrations measured by direct analysis on a NaI(Tl) multidimensional gamma-ray spectrometer. In addition to $^{24}$Na, $^{38}$Cl and $^{39}$Cl, the cosmogenic radionuclides $^{18}$F, $^{26}$Mg, $^{34}$Ar Cl and $^{38}$S were measured on some of the 15 to 19 km flights.

The measured $^{24}$Na production rates were almost identical to the measured atmospheric disintegration rates, indicating that the air filters are removing essentially all of the $^{24}$Na from the air. The $^{38}$Cl and $^{39}$Cl production rates, however, were much higher than their measured atmospheric disintegration rates, probably indicating either that the lifetime of the $^{38}$Cl and $^{39}$Cl atoms is too short for them to become quantitatively attached to filterable atmospheric aerosol particles or that their production rate is enhanced in the argon spheres by low energy radiation produced by the reaction of cosmic rays with the aircraft and the metal spheres. Higher energies are required for $^{24}$Na production, so the $^{24}$Na production rate would not necessarily be enhanced by the latter effect.

On March 11, 1971, several spheres were filled with a total of 60 M$^3$(STP) of argon and flown for 350 minutes at 15.5 km. The largest amount of argon that had been flown on any previous flight was 42 M$^3$. These measurements permitted a significant improvement in the accuracy of cosmogenic radionuclide production rates; however, an attempt to measure $^{32}$P (14.3 d) and $^{33}$P (25 d) was unsuccessful even with very nearly quantitative chemical separations. Production rate measurement of $^{32}$P and $^{33}$P will require a long exposure of the argon-filled spheres at some high altitude location.
Radioanalytical procedure development

Radiochemical separation procedures which are compatible with and augment advanced instrumental analysis technology are required to provide a complete quantitative analysis of a radionuclide mixture. Although some of the developed methods are designed to separate a specific radionuclide, the major effort is directed toward group separation procedures.

A $^{85}$Kr Field Monitoring System and Its Application to Maximum Air Dose Measurements of a Radioactive Gas

J. D. Ludwick and P. W. Nickola

Our $^{85}$Kr field monitoring system has been expanded by doubling the number of detector stations from 64 to 128 positions and by installing automatic cover plates on each detector which can be adapted to energy discriminate between two tracers.

An improved plume definition was sought by selective positioning of the added sensors although little, if any, improvement in sampling density was accomplished by the additional 64 stations. Monitoring stations were extended to the 1600 meter arc on the meteorological grid. Definition of the upper extent of typical plumes will be accomplished through measurements now available from sensors located up to 70 meters above ground elevation.

Modifications of the data collection center include the expansion of the programmer memory to identify information from 256 separate locations rather than the previous 64. In this way the instrumentation is ready for any further expansion of the field locations beyond the existing 128. The real-time monitoring information collected on two 4096 address memory banks is also handled in a new manner. An electronic coupler was built to facilitate transfer of the 4096 information pieces to DEC-tape transports such as used in PDP-15 data processing computers.

There are several important advantages of this new system. The rapid information transfer ability of the coupler-tape system conserves real-time monitoring time, allowing continuous measurements of plume characteristics in time increments as small as 0.1 second. In addition, the computer compatibility feature of the new tape system allows data readout in a form designed to significantly reduce the man-hours previously needed for necessary information presentation.

The first problem investigated with the expanded $^{85}$Kr monitoring system was one concerning the need for a more precise estimation of the maximum exposure near a nuclear installation in the event of a serious accident. For this purpose, a series
of 10 Ci $^{85}$Kr sources were prepared in quartz vials. These vials, when crushed, simulate the rapid release of a dangerous radioactive cloud. The atmospheric transport of this puff-plume was then determined with the overall electronic system. In order to measure the consequences of accidental releases of this nature during unfavorable atmospheric conditions; releases were planned during relatively stagnant atmospheric conditions. To date, three releases have been made in the middle of the sampling grid, under rather light wind conditions, and during stable temperature-inversion characteristics. True stagnation conditions have not, as yet, been encountered; however, their seasonal appearance is anticipated and puff releases during these periods are planned.

Analytical Procedure for Measuring Copper in Biological Material
C. W. Thomas and L. A. Rancitelli

A technique has been developed to measure copper in biological material using neutron activation and electrodeposition. Samples are weighed under ultra-clean conditions in a laminar flow work station and placed in tared polyethylene irradiation containers which have been previously washed in doubly distilled nitric acid and rinsed with doubly distilled water. After weighing, the samples are freeze-dried to a constant weight, reweighed and the container heat sealed, and irradiated in a nuclear reactor together with standard solutions containing known amounts of copper. The irradiated samples are transferred from the activation capsules to Erlenmeyer flasks containing 10 ml of fuming nitric acid, 50 mg of copper carrier and 20 mg of Na, K, and Br hold-back carriers. Each sample is digested for 2 hours, reduced to a final volume of 2 ml, transferred to a tared platinum evaporating dish, and diluted with 50 ml of water and 1 ml of concentrated sulfuric acid. The copper is electroplated onto the sides of the platinum evaporating dish by applying an electrical potential of 4 volts for one hour. After electrodeposition, the solution is discarded and the dish is washed several times with distilled water and finally with acetone. After drying under a heat lamp, the dish is reweighed to determine the chemical yield. The copper is dissolved with 2 ml of nitric acid, diluted to a constant volume and transferred to a standard counting geometry. The $^{64}$Cu activity in the samples is measured by counting its annihilation radiation (a pair of 511 keV photons) with an anticoincidence shielded 9 in. diameter NaI(Tl) well crystal detector. The copper concentration in each sample is determined by direct comparison of the sample activity with that observed in known amounts of copper standards. The accuracy and precision of the technique was estimated to be 5% determined from replicate measurements of the copper content of standard reference materials (orchard leaves and beef liver) obtained from the National Bureau of Standards.
Analytical Procedure for Measurement of $^{214}\text{Pb}$ and $^{214}\text{Bi}$ in Rainwater

C. W. Thomas, J. A. Young and N. A. Wogman

The short-lived daughters of radon, $^{214}\text{Pb}$ (26.8 min) and $^{214}\text{Bi}$ (19.7 min), are potentially useful tracers of precipitation scavenging processes because they are attached to the natural aerosols, they are present in easily measurable concentration in rainwater, and their half-lives are of the same order of magnitude as the time scale on which these processes occur. The major problem in using the radon daughters as tracers is the difficulty of rapidly separating them from large volumes of rainwater after short consecutive collection periods. A technique was recently developed to continuously separate $^{214}\text{Pb}$ and $^{214}\text{Bi}$ from rainwater at flow rates of up to 30 liters per minute and consecutive sampling periods of 5-10 minutes. Rainwater from a 3000 ft$^2$ polyethylene surface is collected in a polyethylene 55 gallon drum and pumped through a fiberglass filter followed by an anion exchange bed (0.64 cm thick by 28 cm diameter of Dowex 1×8, Cl$^-$ form, 200-400 mesh), and a cation exchange bed (0.64 cm thick by 28 cm diameter of Dowex 50WX8, H$^+$ form, 200-400 mesh). Bismuth-214 is removed at greater than 90 percent efficiency by the anion exchange bed. Bismuth-214 is measured immediately by counting the coincident photons emitted by the sample with a pair of 13 1/2 inch diameter by 6 inch thick NaI(Tl) detectors. No measurable amount of $^{214}\text{Pb}$ is removed by the anion exchange bed; however, greater than 90 percent of the $^{214}\text{Pb}$ is retained on the cation exchange bed which in turn does not retain $^{214}\text{Bi}$. After the collection period the cation bed is set aside for about 1 hour to allow ingrowth of $^{214}\text{Bi}$ which is then measured using the detector system described and the $^{214}\text{Pb}$ concentration is calculated from this ingrowth.
Techniques Used to Stabilize Tracer Solution for Aerosol Generation
C. W. Thomas

Finely divided aerosols of several rare elements are generated by acetone-burner techniques on aircraft and serve as tracers in studying precipitation scavenging rates and mechanisms. Techniques were developed to put tracers of gold, iridium, ruthenium, tellurium and selenium either into solution or into a colloidal suspension in a solvent mixture that had excellent ignition and burning characteristics. A solution of acetone containing about 20 percent by volume of n-heptane was found to be compatible with almost any tracer that was moderately soluble in acetone and/or alcohol and had ignition and burning qualities that were compatible with a forced air aerosol generator. Laboratory and field studies were conducted and the following recipes were formulated and successfully demonstrated for both ground level and aircraft releases from acetone-burner generators of the types used in AgI releases.

Gold
175 grams HAuCl₄·3H₂O was dissolved in 13 liters of acetone in a 50 liter polyethylene carboy. 3.2 liters of pyridine was added to prohibit reduction of gold by the metal surfaces of the generator. 6.5 liters of n-heptane was added. This solution was stable for two weeks after which the gold solution was slowly reduced to the metal.

Iridium
32 grams of IrCl₃·3H₂O was dissolved (with difficulty) in 10.5 liters of acetone by the following technique: the iridium chloride salt was stirred into a liter of acetone and the mixture was filtered. The filtrate was placed in a 50 liter polyethylene carboy. The residue was dried and fumed with concentrated hydrochloric acid and taken to just dryness. The residual salts were added to a liter of acetone and stirred and the mixture was filtered. The filtrate was added to the polyethylene carboy. This process was repeated until no appreciable amounts of residual salts remained after filtration. Additional acetone was added to make up to 16.5 liter volume and 4.5 liters of n-heptane was added. The solution was stable for weeks.

Ruthenium
150 grams of RuCl₃·3H₂O was dissolved in acetone by the method used for iridium and made up to 16.5 liters with acetone. 4.5 liters of n-heptane was added. The resulting solution was colloidal but stable for one week after which slow coagulation occurred over a period of several weeks.

Tellurium
300 grams of TeCl₄ was dissolved in 16.5 liters of acetone and 4.5 liters of n-heptane was added. This solution was stable for weeks.
Selenium

Selenium tetrachloride was not compatible with either acetone or n-heptane, both of which resulted in a rapid reduction to the metal. Alcohol was a suitable solvent for selenium, thus, 450 grams of SeCl$_4$ was dissolved in 4 liters of absolute ethanol. This solution was metered simultaneously with n-heptane to the burner compartment of the generator. Since this required separate reservoirs for each solution the method has not been so suitable as others for aircraft release.
The concentrations of trace elements, cosmogenic radionuclides, and nuclear-weapons-produced radionuclides were measured in the air from ground level to 19 km and in rain and seawater in order to determine the origin and chemistry of the atmospheric aerosol and to study the rates of atmospheric and oceanic mixing, the rates of air-sea interchange, and the rates and mechanisms of precipitation scavenging.

Air Concentrations of $^{90}$Sr and $^{55}$Fe at Richland, Washington from 1963 to 1970
C. W. Thomas and J. C. Langford

The atmospheric concentrations of $^{90}$Sr and $^{55}$Fe were measured in ground level air samples collected at Richland, Washington from 1963 to 1970 as part of a program to define the rates of long-term stratospheric processes in the northern hemisphere. Seasonal variations in the concentrations of $^{90}$Sr were similar to those of radionuclides of stratospheric origin, decreasing from 1963 to 1966 as a result of decay and deposition on the earth's surface. The Chinese thermonuclear tests at Lop Nor (45°N) beginning in 1967 maintained the surface air $^{90}$Sr concentration at about the same level from 1967 to 1970.

The seasonal variations in the concentrations of $^{55}$Fe were not as pronounced as those of radionuclides of stratospheric origin suggesting that a portion of the $^{55}$Fe in the air at Richland, Washington came from the Hanford Project. This has previously been reported to be the case for $^{60}$Co, $^{46}$Sc, $^{65}$In and $^{134}$Cs. Discharge of reactor effluent water to the Columbia River causes buildup of radionuclides in river bottom materials and as the river level changes, some of the radionuclides in the dry areas become airborne.
The Concentration of $^{55}$Fe in Ground Level Air at Richland, Washington from 1964-1968

The Concentration of $^{90}$Sr in Ground Level Air at Richland, Washington from 1963-1970
The Determination of Air-Sea Exchange and Oceanic Mixing Rates Using \(^7\text{Be}\) During the BOMEX Experiment

J. A. Young and W. R. Silker

The concentrations of \(^7\text{Be}\) and other radionuclides were measured in air, rain, and seawater in a region east of Barbados, British West Indies, in the summers of 1968 and 1969 during the BOMEX experiment. The deposition of \(^7\text{Be}\) on the sea surface by rainfall in August 1968 and May through July 1969 was only 15 and 26 percent, respectively, of that necessary to maintain the measured seawater inventories. The calculated deposition velocity of \(^7\text{Be}\) on the sea surface by wet and dry deposition was 1.0 cm sec\(^{-1}\). During May through June of 1969 the calculated flux of \(^7\text{Be}\) across the sea surface averaged about 5.8 \(\times\) \(10^{-2}\) atom cm\(^{-2}\) sec\(^{-1}\) and the inventories were increasing at a rate of 1.0 \(\times\) \(10^{-2}\) atom cm\(^{-2}\) sec\(^{-1}\). Due to seasonal variations in the atmospheric \(^7\text{Be}\) concentrations the \(^7\text{Be}\) seawater inventories should vary seasonally with maximum inventories occurring around July and minimum inventories around February. The yearly average of the flux of \(^7\text{Be}\) across the sea surface was calculated to be 1.6 \(\times\) \(10^{-2}\) atom cm\(^{-2}\) sec\(^{-1}\). The vertical eddy diffusion coefficient, \(K_z\), in the sea decreased from greater than 7 cm\(^2\) sec\(^{-1}\) at the surface to 0.25 to 0.85 cm\(^2\) sec\(^{-1}\) at 30 to 40 meters and then possibly increased somewhat from 40 to 100 meters. The \(^7\text{Be}\) inventory in the sea varied rapidly due to the horizontal motion of seawater through the sampling region. The \(^{103}\text{Ru}\) to \(^{95}\text{Zr}\) ratio in the top 15 meters of the sea indicated that large amounts of \(^{103}\text{Ru}\) and \(^{95}\text{Zr}\) from the French nuclear test series at 23\(^\circ\)N beginning 7-7-68 entered the sampling region on 8-15-68 and also was present in June, but not in May 1969. The debris probably was carried into the sampling region by the Guiana Current which flows northward along the coast of South America.

Trace Element Characterization of High Altitude Aerosol

T. M. Tanner and L. A. Rancitelli

In a continuing effort to measure the trace element content of atmospheric aerosols and to establish the origin, history and movements of various air masses, sampling missions were flown over the Pacific Ocean off the Oregon coast. Previous measurements of the trace element content of aerosols at altitudes of 3 to 15 km between Albuquerque, New Mexico (34\(^\circ\)05'N, 106\(^\circ\)45'W) and Spokane, Washington (45\(^\circ\)56'N, 117\(^\circ\)58'W) during 1967 and 1968 have been reported. It was shown that the concentrations of cobalt, iron and scandium could be explained by injection of material with the composition of the earth's crust into the atmosphere. It was also shown that the elements Sb, Ag, Zn, and probably Cr were far too high in concentration to have originated from the earth's crust. Sb and Zn were attributed to pollutant sources and Ag to extensive worldwide cloud seeding.

Measurement of the trace element profiles of the marine aerosol was essential to provide additional information on maritime air masses for our precipitation scavenging studies on the Washington coast. Missions were
flown during July, 1970 at altitudes of 5 to 12 km at a distance of 40 km off the Oregon coast (45°0'N, 124°34'W). The samples were taken and processed in the usual manner for analysis by neutron activation techniques. The aerosol samples taken at 9 and 12 km on July 27 and 28, 1970 have been analyzed and the results for eight elements, Ag, Br, Co, Cr, Fe, Sb, Sc and Zn, are presented in the accompanying table. Also presented for comparison are the concentrations of these elements at comparable altitudes in the samples taken over the continent. In the samples taken over the ocean, the concentrations were lower than over the continent. The trace element concentrations of these aerosols, when normalized to their iron concentration, show that with the exception of Ag and Br the ratios are nearly the same for both samples.

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**Comparison of the Concentrations of Several Elements in the "Maritime" Aerosol with "Terrestrial" Atmospheric Aerosols, Urban Ground Level Aerosol, Seawater and the Earth's Crust.**

(All values normalized to Fe)

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<th>Urban Ground Level</th>
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*a. Average of two samples taken at 9 km and 12 km on 7-27-70 and 7-28-70, respectively.

b. Average of nine samples taken between 3 km and 15 km from August 1987 to October 1988.*
### Trace Element Concentrations Measured over the Pacific Ocean and Western U.S. ng/scm of Air

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<th>Co</th>
<th>Cr</th>
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<td>0.002±0.001</td>
<td>0.001±0.0001</td>
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<td>4±1</td>
<td>45±8</td>
<td>0.05±0.004</td>
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<tr>
<td>12.3 (Maritime) (7-27-70)</td>
<td>7.64</td>
<td>0.018±0.005</td>
<td>4.4±0.3</td>
<td>0.06±0.002</td>
<td>≥4</td>
<td>4±1</td>
<td>0.08±0.002</td>
<td>0.006±0.0007</td>
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**Comparison of Atmospheric Radionuclide Concentrations at Neah Bay and Richland, Washington**

C. W. Thomas

The atmospheric concentrations of several radionuclides were measured continuously in ground level air from 1968 through 1970 at Richland, Washington (46°N, 118°W) and west of Neah Bay, Washington on the Makah Indian Reservation (49°N, 125°W) near the Pacific coast. Separating these two sites are the coastal and Cascade mountain ranges. Storm systems originating in the Aleutians pass over the Makah sampling site and then are orographically lifted several thousand feet before descending to the Richland sampling site.

At both Richland and Makah, large 350 cubic feet per minute air pumps are mounted several feet above the ground and draw air through membrane filters. The radionuclide concentrations on the filters are measured using large volume NaI(Tl) gamma-ray spectrometers. The concentrations of $^7$Be, $^{95}$Zr, $^{106}$Ru, $^{137}$Cs, and $^{144}$Ce at the two sites show the seasonal variation that is characteristic of radionuclides of stratospheric origin. However, the concentrations are generally higher in the Richland samples, especially during the summer when the concentrations are at a maximum.

The lower average concentrations at the Makah site may be at least partially due to precipitation scavenging, since the rainfall rate is at least tenfold higher at that site. However, the difference in concentration between Makah and Richland is greatest in the summer when rainfall rates at both locations are relatively low, and is smallest in the winter when precipitation is heavy. It seems more likely that the high concentrations at Richland result primarily from vertical mixing associated with passage of air over the coastal and Cascade mountain ranges. Radionuclide concentrations are much higher at higher altitudes, so vertical mixing would result in higher...
ground level concentrations. Supporting this explanation is the fact that $^{24}$Na concentrations measured at ground level at Richland were equal to the production rate at 7000 feet.

Comparison of Radionuclide Concentrations in Air at Richland (46°N, 117°W) and Makah (49°N, 125°W), Washington as a Function of Time.
Vertical Profiles of Radionuclide Concentrations in the Atmosphere
J. A. Young and N. A. Wogman

Vertical profiles of atmospheric radionuclide concentrations have been determined on a monthly basis since 1967 from air filter samples collected by RB-57 aircraft at 1.5 to 3.0 km altitude intervals from 0.5 km to an altitude of over 18 km. Most of the vertical profiles were measured either south of Spokane, Washington at 47°N, 117°W, or east of Barbados, British West Indies from 12 to 18°N and 54 to 59°W. The profiles at Barbados were taken during the summer of 1969 as part of the BOMEX project. The concentrations of cosmogenic radionuclides, nuclear weapons produced radionuclides, and radon and thoron daughters were determined using NaI(Tl), multidimensional gamma ray spectrometers and, beginning in late 1971, a dual, anticoincidence shielded, Ge(Li) gamma-ray spectrometer.

At Barbados the radionuclide concentrations generally did not increase systematically with altitude from ground level to 9 to 12 km, but then increased rapidly at higher altitudes. At higher latitudes in the northern hemisphere the concentrations generally increased continuously with increasing altitude at all altitudes. Apparently radionuclides which have been transferred from the stratosphere into the troposphere through the tropopause discontinuity at mid-latitudes have time to become fairly well mixed vertically before they reach Barbados. A surprising feature of the profiles at all latitudes has been the fairly common occurrence of very pronounced concentration minima at altitudes from 6 to 9 km. Sometimes the concentrations at these altitudes are orders of magnitude lower than the concentrations at lower elevations. The only commonly measured radionuclide which does not show these concentration minima is cosmogenic $^{24}$Na (15 hr). Na-24 is produced in the atmosphere at a relatively constant rate by spallation reactions of cosmic rays with atmospheric argon. Its half-life is short enough so that its atmospheric concentrations are relatively constant. Apparently, whatever process causes the concentration minima occurs long enough before sampling to allow $^{24}$Na to grow back to nearly equilibrium. The presence of $^{24}$Na at normal concentrations in the samples where minimum concentrations of the other radionuclides occur eliminates the possibility of sampling error as a cause of the minima. Most of the concentration minima probably result from precipitation scavenging, however the minima occur at altitudes above the most common altitudes of rain formation. It may be that the air is scavenged and then rises to 6 to 9 km before being sampled. The minima in the Barbados region may be due to air from the giant cumulonimbus of the intertropical convergence zone south of Barbados which reach altitudes of nearly 15 km. Another possibility is that the minima result
from scavenging by cirrus clouds. The cirrus ice particles may scavenge radionuclide bearing aerosol particles, either during or subsequent to nucleation and then settle, leaving a region of low radionuclide concentration behind.

In the past, efforts have been made to calculate back trajectories of the air containing minimum radionuclide concentrations in an attempt to determine the cause of the minima. However, the lack of meteorological stations on the ocean upwind of the location of the profile measurement has thwarted these efforts. Therefore, vertical profiles have been measured in 1971 farther inland over the continental United States. A profile measured on February 24, 1971 near Albuquerque at 35°N, 106°W showed concentration minima at 9.1 km, and a profile measured on November 16, 1971 near Denver at 39°N, 103°W showed a concentration minimum at 6.1 km. In both cases the $^{24}$Na concentration was only slightly below normal. The concentration of a few radionuclides on November 16 are plotted as a function of altitude in the figure. Back trajectories are being calculated for both cases in an attempt to identify the cause of the minima.

![Radionuclide Disintegration Rates on November 16, 1971 at 39°N, 103°W](image-url)
Atmospheric Fallout During 1971

C. W. Thomas and J. A. Young

The atmospheric concentrations of radionuclides in ground level air were measured continuously during 1971 at Point Barrow, Alaska (71°N) and Richland, Washington (46°N) by filtering large volumes of air through membrane filters followed by gamma-ray spectrometric analysis. The concentration of nuclear-weapons-produced radionuclides, including fission products and neutron activation products, increased steadily from 1967 through 1971 because of the high yield thermonuclear tests conducted by the Chinese at Lop Nor (45°N) during this period with a total yield of over ten megatons. During 1971 essentially all of the nuclear-weapons-produced radionuclides present in northern hemispheric air originated from these Chinese tests. The concentration of $^{89}Y$ (107 day half-life) increased 100-fold over 1968 peak levels and during 1971 reached peak concentrations comparable with peak concentrations of 1962-1963.

The Chinese nuclear test of 11-19-71 was detected in low concentrations in ground level air at Richland, Washington five days after detonation. During 1970 the nuclear reactors, which are a part of the Richland Atomic Energy complex and use Columbia River water as a straight through coolant, were shut down. The effect of this shutdown was noted by a decrease in air concentration of several radionuclides that were known to be associated with the Richland operations. Scandium-46 concentrations in air were down by two orders of magnitude, $^{60}Co$ was down one order of magnitude, and $^{124}Sb$ is now below detection levels. Also affected was $^{65}Zn$ air concentration which now shows an excellent seasonal variation and is down by an order of magnitude.
IRON-55 AND LEAD IN THE HUMAN POPULATION OF THE WORLD

This study investigates the transfer of $^{55}$Fe from fallout, with special emphasis on the concentrations in man and in food chains leading to human consumption. Transfer is being measured in the general population as a function of age, sex, and diet, including the dilution effect of stable iron. Although $^{55}$Fe is being emphasized, the same samples are used to measure Pb, $^{210}$Pb, and $^{220}$Po in the biosphere.


C. E. Jenkins and J. C. Langford

Iron-55 and stable iron measurements are being made to help characterize the $^{55}$Fe uptake and movement through the marine biosphere. Particular attention is being focused on salmon and their food chain since salmon as a food source are responsible for the highest $^{55}$Fe concentrations observed in man. Results of this study show the following: $^{55}$Fe concentrations are more than an order of magnitude higher in salmon from the Bering Sea than in those from waters off the contiguous United States; $^{55}$Fe in salmon has an apparent effective half-life of 9 to 10 months; $^{55}$Fe entering the ocean from the atmosphere is rapidly assimilated by the oceanic biomass with little apparent recycling; $^{55}$Fe concentrations in the oceanic organisms are thus declining about 2 times faster than in the terrestrial herbivores; Kotzebue chum salmon are 3- to 10-fold higher in $^{55}$Fe than Alaskan caribou, and 300 to 1000 times higher than cattle from Columbia Basin, Washington; salmon-eating Eskimos have 20- and 100-fold higher $^{55}$Fe body burdens than those of caribou-eating Alaskan Eskimos and Richland, Washington cattle-eating residents, respectively.

This study indicates that high $^{55}$Fe concentration in marine organisms from Alaskan waters is not the result of a high initial fallout from Russian thermonuclear testing and recycling, as has been suggested by some authors. The phenomenon of increasingly higher radionuclide concentrations in salmon from more northerly waters is not confined to $^{55}$Fe. Both $^{54}$Mn and $^{60}$Co appear to behave similarly. Cesium-137 levels in seawater confirm land measurements of fallout which indicate that fallout is less in the northern regions than at the mid-latitudes. Since fallout rates are indeed less at northern latitudes, while concentrations in ocean organisms are higher, this suggests that a very efficient mechanism exists in northern oceanic waters for concentrating
fallout elements. This has extremely important implications with regard to all anthropogenic toxic trace elements and suggests that a most critical area in the marine biosphere is in the northern regions of our oceans.

The cause of this phenomenon is not yet understood; however, it is being seriously investigated. The implications are, of course, that not only radionuclides but many toxic trace elements that are currently entering the oceans from the atmosphere will be most highly concentrated in the marine biosphere in the northern regions of the ocean.
55Fe Concentrations in Pacific Salmon
These studies are concerned with the development of radiochemical instrumentation and analytical technology, their utilization in radioisotope studies of environmental samples, their application to neutron activation analysis to define stable element versus radionuclide behavior, and their use in the elucidation of the routes described by natural, tracer, or waste radioisotopes in the environment.

The Performance and Application of a Versatile Ge(Li)-NaI(Tl) Coincidence-Anticoincidence Gamma-Ray Spectrometer for Environmental and Biological Problems

J. A. Cooper and R. W. Perkins

A high-sensitivity gamma-ray spectrometer incorporating a high-performance Ge(Li) detector and a NaI(Tl) well crystal has been developed, which provides improved sensitivities for coincident and noncoincident gamma-ray emitting radionuclides commonly found in environmental and biological samples. As an anticoincidence spectrometer it provides the highest peak-to-Compton edge ratio ever observed, 780:1, over 3 times greater than any other known system. Interference from such coincident gamma-ray emitting radionuclides as $^{60}$Co is reduced by 80-fold in the anticoincidence spectrum. This high efficiency for removing Compton interference, the 10-fold suppression in natural background, and the high absolute efficiency of the Ge(Li) detector (27% for a 122 keV point source at the cap), provides the most sensitive system yet described for measuring radionuclides such as $^{137}$Cs in environmental samples.

Normal plastic shielded anticoincidence spectrometers have not been able to significantly improve the sensitivity for the measurement of coincidence gamma-ray emitting radionuclides. However, because of the resolving capabilities and the high efficiency of the NaI(Tl) well crystal used in this spectrometer, it is possible to select specific-energy coincidence events occurring in the NaI(Tl) and Ge(Li) detectors with about the same counting efficiency as with a normal Ge(Li) detector and to obtain tremendous improvements in the sensitivity for the measurement of coincident gamma-ray emitting radionuclides. Background interferences have been reduced by as much as 400 times. In the analysis of caribou liver samples, the peak to background ratio for the 434 keV peak of $^{109m}$Ag was improved by about 350-fold. This improvement was in a coincidence spectrum which was gated on the
the background in non-hydrogen containing systems are the gamma-rays emitted by members of the $^{238}\text{U}$ and $^{232}\text{Th}$ decay chains and $^{40}\text{K}$. Alpha particles from the decay of $^{238}\text{U}$ and $^{232}\text{Th}$ also produce background peaks from 3 to 8 MeV. These alpha particles were observed in 10,000 minute backgrounds in NaI(Tl) crystal systems, but not in the Ge(Li) spectrometers. The cosmic-ray induced background results from the passage of primary or secondary particles through the detector and from the reaction of these particles with surrounding materials producing neutrons which may interact with the system materials to produce radioactive contaminants. In those systems containing hydrogen; e.g., plastic phosphors, a very apparent background photopeak results from the prompt gamma ray from cosmic-ray neutron interactions with hydrogen [$^1\text{H}_2$, $^1\text{H}_4$, 2.225 MeV $\gamma$ $^2\text{H}_2$]. In addition, these neutrons interact with 1 of the NaI(Tl) detector and with the Pb or Fe construction materials to produce prompt gamma-rays of 6.8, 7.4, and 7.6 MeV, respectively. Cosmic-ray particles deposit energy in the NaI(Tl) and Ge(Li) crystals directly. For example, the most probable energy loss of a 1 GeV muon in NaI is 1.47 MeV cm$^2$/g. This process creates a high energy peak and an energy continuum throughout the range of 0-10 MeV.

A threefold background reduction of the neutron induced peak was achieved by lining the shield with four inches of borated (5% by weight) paraffin to thermalize and capture all neutrons produced in the external shields. A concurrent background reduction of 15% occurs over the energy range of 0 to 3 MeV. The internal neutron component was not eliminated. The lowest overall backgrounds were obtained by a combination of methods to reduce the individual background sources: (1) eliminating external natural radiation by use of uncontaminated construction materials, (2) borated paraffin shielding to moderate and capture externally-produced neutrons, and (3) installation of the instrument deep underground where the massive overburden attenuates the major portion of the cosmic ray muons.
Natural Radioactivity in NaI(Tl) and Ge(Li) Gamma-Ray Spectrometer Construction Material Often Precludes Low-Background Utilization and Its Variance from Sample to Sample Requires Analysis for 232Th, 238U, and 40K Prior to Use

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<tr>
<th>Spectrometer Assembly Material</th>
<th>232\text{Th}</th>
<th>238\text{U}</th>
<th>40\text{K}</th>
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Improved X-Ray Fluorescence Analysis of Environmental and Biological Samples Using a High-Intensity Radioisotope Excitation

J. A. Cooper

The elucidation of the role of beneficial and detrimental trace elements in environmental and biological systems is basic to our understanding of their environmental balance. Because studies of behavior of trace elements in environmental and biological systems often require the analysis of a large number of samples, many of these studies have been conducted only in a superficial manner because of the lack of sensitive, rapid, and practical techniques of analysis.

Improved X-ray fluorescence techniques which employ radioisotopic sources and which should provide sensitivities comparable to those from machine excitation are being developed for environmental and biological specimens. A high-intensity radioactive source of photons is used to excite the elemental X-rays which are analyzed with a Si(Li) diode. The radioactive sources (\(^{55}\)Fe, \(^{109m}\)Cd and \(^{241}\)Am) are about ten times more intense than previously available sources and the holders are doubly collimated to minimize nonsample scattering. A large vacuum chamber is also used to eliminate the air scattering, one of the major sources of interference remaining for thin samples. The technique is capable of providing sensitivities in the ppm range for most environmental matrices.

A large variety of environmental samples have been analyzed for trace elements. Sixteen elements (S, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ge, Br, Sr, Zr and Pb), many of which (S, Ca, Ti, V, Ge, Sr and Pb) are not readily analyzable by other techniques such as neutron activation analysis, are measured in many types of samples in just 30 minutes using 100 mCi \(^{109m}\)Cd source. Samples of suspended particulates from a uranium mine and animal test chambers have been analyzed for over a dozen elements (Si, K, Sc, Ti, V, Mn, Fe, Cu, Zn, Pb, Se, U, Sr, Y and Zr) many of which would have been quite difficult by neutron activation analysis because of the large interference from U fission products. Other samples such as sea flounder muscle tissue, seawater, urine, finger nails and geological samples have also been examined. Techniques for preparation of thin environmental and biological samples, for obtaining more precise quantitative results, and for increased automation of the analysis are being developed.
Elemental Content of Biological and Environmental Samples Such as Air Filters are Measured Rapidly and Nondestructively with X-ray Fluorescent Techniques and High-Intensity Radioactive Sources. This Technique Provides the Capability of Studying Large Numbers of Samples Required in Many Environmental and Biological Investigations.

X-ray fluorescence determination of the zinc to copper ratios in fingernails for the identification of Cystic Fibrosis

J. A. Cooper, C. E. Jenkins, J. C. Langford and N. A. Wogman

Cystic Fibrosis, which has an incidence of approximately one per thousand births, must be diagnosed early in order that permanent irreversible damage to the lungs and other body organs be prevented. Early detection permits the initiation of treatment which helps keep the effects of this disease in check.

The analysis of fingernails or sweat for sodium has been the method most frequently suggested as a possible means for mass screening of children for the disease; however, the reliability of this method is not very satisfactory.

A rapid and inexpensive technique for the multielement analysis of trace elements in nail clippings has been developed, based on excitation of the characteristic X-rays of the elements in the nail with a high-intensity radioisotopic source and their analysis using a Si(Li) diode. The sensitivities for many elements
are in the parts-per-million (ppm) range and the procedure could be easily automated, making it a very useful tool for scanning the trace element distribution in a large number of individuals. The elements S, Cl, K, Ca, Fe, Co, Ni, Cu, Zn, Pb, Br and Sr have been measured in fingernails from about 6 confirmed cystic fibrosis cases and 50 normal individuals. The Cu content of fingernails from individuals with cystic fibrosis is usually higher than normal while the Zn content is usually lower than normal. Thus, the ratio may be a sensitive indicator and free from many of the normal errors associated with an absolute concentration determination.

Preliminary results indicate the Zn:Cu ratio of normal individuals is about 7:1 while the ratio for those with cystic fibrosis is about 5:1. Unfortunately, the spread in the distribution is quite high, which may be due to physical differences in the nail clippings analyzed or the small number of samples used to define the distribution. A technique to provide sample uniformity is being developed, which consists of a quick dissolution of a weighed nail sample followed by evaporation on a thin Mylar film. This should allow precise determination of the Zn:Cu ratio under controllable conditions. A large number of nail clippings from both normal and cystic fibrosis individuals have been obtained from the University of Washington and will be used to accurately define the variation in the Zn:Cu ratio in the two groups.

Transmission Properties of Specific Scintillator Reflective Coatings
R. L. Brodzinski

Four 40-inch diameter by 12-inch thick sections of plastic scintillator, which were to make up an anticoincidence shield, were found to be inefficient in transmitting light to their phototubes. Test sections of these ingots demonstrated satisfactory scintillation properties; therefore, it was recognized that the difficulty was related to the reflective coatings applied to the phosphors' external surface. Tests were conducted on a small core of scintillator material (Pilot Y) with the criterion for good performance being a high efficiency for detection of $^{137}$Cs gamma rays. The effect of reflective coating thickness on photon detection efficiency was evaluated for three different formulations supplied by Pilot Chemical Company. Maximum photon counting efficiency was observed for coatings between 0.0025 and 0.0030 inch thick. With optimum coating thicknesses it was found that all coating formulations yielded approximately the same efficiency. On studying light-tight materials such as aluminum foil, aluminized Mylar, and black plastic, it was found that they greatly affected the transmission losses from the reflective coatings. Though the transmission losses were small in all cases, there were two to threefold differences in the light output.
Optically coupling the reflective coating to the light-tight wrapping was found to enhance the transmission of light through the coating. Therefore, optical coupling should be avoided, and the light seal material should be wrapped over a clean dry surface. The reflective properties of aluminum foil and aluminized Mylar are very low compared to those of the spray-on reflective coatings and should not be used as a primary reflector.

Computer System for Radionuclide Analysis
J. R. Kosorok

Standard procedures in instrumental neutron activation analysis require the simultaneous irradiation of standards and samples. After irradiation, the gamma-ray spectra of the radionuclides in the samples and standards are acquired and compared to determine element content. Quantitative analysis requires the spectral evaluation together with the weights of standards and samples, radionuclide half-lives, shape, and location of the characteristic photopeaks of the radionuclides, irradiation and counting times and counting intervals. When large numbers of routine samples are involved, as much of the analysis procedure as possible should be automated to reduce the time required to perform the analysis. A small computer system has been programmed for routine use in these evaluations.

In using the system, the operator directs the computer system to compose and maintain a library of constant, characteristic data for a maximum of 40 radionuclides; acquire, display and store gamma-ray spectra; and perform the necessary computations to determine element concentrations. The computer automatically collects spectra and calculates element concentrations with little direction from the operator. When not acquiring spectra or processing acquired data, the system can be used by the operator to initialize, expand or modify the radionuclide constants library. A researcher may start with a library containing data on a few isotopes and expand it as his samples become more complex.

Any pulse-height-to-digital converter which generates a live/clock pulse in channel zero can be interfaced to the computer system to acquire spectra for time intervals set by the operator. In addition to measuring the elapsed counting interval, the system has a real-time clock and stores the time and date at which a spectrum was collected. The time of counts is used internally to correct for radionuclide decay between the time of irradiation and time at which the gamma-ray spectrum was acquired. A scope display of the spectrum may be requested during or after data acquisition. Live time is not appreciably affected by the display, because the display has direct access to the computer memory rather than being entirely program controlled. The spectrum
for a particular standard or sample along with enough data to identify it for further analysis may be listed on a printer for the operator's study as well as stored on magnetic tape for automatic processing.

A line in the report printed by the system identifies each element, the decay corrected gamma-ray count of its activation product, and its concentration in the sample. The computer system was developed by integrating commercially available hardware and software with interfaces and software developed at Battelle-Northwest. The hardware for the system includes a Digital Equipment Corporation PDP-15/50 computer, a pulse-height-converter, a display unit, and a fast printer. All programs performing operator requested functions operate under control of the monitor provided by the computer manufacturer. Programs communicating between the monitor and the interfaces for the pulse-height-converter, display unit and fast printer were developed in Battelle-Northwest.

The computer system for instrumental neutron activation analysis described here has been routinely used for determining trace amounts of elements in environmental studies. The software design has allowed modification and expansion to fulfill the changing needs of research and has provided a flexible and powerful tool for scientists.

Multielement Analysis of Biological Reference Materials
L. A. Rancitelli

The elemental content of biological materials, particularly the trace constituents, has received a great deal of interest in the last few years. This recent interest has been spawned by the realization that inorganic pollutants such as mercury have permeated the biosphere and invaded the food chain of man. Additional interest in the trace inorganic constituents of the biosphere has been generated in studies of the radiologically important fallout and reactor associated radionuclides in the environment. Recognizing the need for biological standards both for the comparison of analytical techniques and for providing inter-laboratory comparisons, the Division of Biology and Medicine of the Atomic Energy Commission supported the National Bureau of Standards efforts to prepare and certify botanical and animal tissue reference materials. This effort has included their certification of a standard orchard leaf, SRM 1571. The analysis of the orchard leaf and two other botanical materials, under consideration as possible botanical standards, tomato leaves and citrus leaves, was conducted at our laboratory using a highly sensitive and precise technique of instrumental neutron activation developed at the Pacific Northwest Laboratories.
Briefly, the method consisted of irradiating 100 mg samples of freeze-dried material in a Hanford production reactor to an integrated neutron exposure of $10^{18}$ n/cm$^2$. A gamma-ray analysis of the activated samples, after appropriate decay intervals, was made with both a Ge(Li) detector system and an anticoincidence shielded multidimensional analyzer to measure the neutron-induced activities $^{24}$Na, $^{42}$K, $^{46}$Sc, $^{51}$Cr, $^{59}$Fe, $^{60}$Co, $^{65}$Zn, $^{75}$Se, $^{76}$As, $^{82}$Br, $^{86}$Rb, $^{110}$mAg, $^{124}$Sb, $^{134}$Cs, $^{140}$La, and $^{203}$Hg. A comparison of these activities with those induced in known amounts of each element irradiated with the samples permitted the determination of 16 elemental concentrations in each sample.

Seven samples of orchard leaves, and 4 samples each of tomato leaves and citrus leaves were analyzed by neutron activation analysis. The results are presented in the table together with the mean concentration and standard deviation. In the case of orchard leaves, the agreement between the results of this work and those elements certified by NBS, sodium, potassium, arsenic, rubidium, iron, mercury, selenium and zinc, are generally good. In addition to those elements certified by NBS, we also report in the table the concentrations of bromine, chromium, cobalt, silver, antimony, and cesium. The utility of the citrus leaves as a standard is doubtful due to its gross inhomogeneity as indicated by the high variability in the iron and cobalt content. The tomato leaves have a uniform trace element content much like that of the orchard leaves. However, it is questionable if the differences in elemental composition between the certified orchard leaves and the tomato leaves justifies the adoption of the latter as a second botanical standard.
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<th>Element</th>
<th>Orchard Leaves**</th>
<th>Citrus Leaves</th>
<th>Tomato Leaves</th>
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<td>20,300 ± 1400</td>
<td>44,600 ± 2000</td>
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<td>Na</td>
<td>78 ± 3</td>
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<td>Fe</td>
<td>300 ± 45</td>
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<td>As</td>
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<td>0.46 ± 0.05</td>
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No. of Analyses

* Tentative Values

** National Bureau of Standards, SRM-1571
Characterization of Columbia River Basalt Flows From Major, Minor and Trace Element Considerations*
L. A. Rancitelli

The safe storage of radioactive wastes at Hanford requires a comprehensive knowledge of the local geological structure. For example, faults in the bedrock as well as contact zones between geological strata can be potential avenues for the transport of liquid wastes into the ground water. The bedrock underlying the Hanford project is composed of more than 20 independent basalt flows ranging in thickness from a few feet to several hundred feet. Although these flows were deposited over a long period of time, they are very similar in their major element content, petrology and the physical properties usually employed to characterize them. These similarities make it very difficult to identify or recognize an individual basalt flow over distances of a few miles, an important point in understanding the geological characteristics of the region. One technique which has not been extensively investigated in the study of the local stratigraphy has been the trace element content of the Columbia River Basalts. In an effort to characterize individual basalt flows from the elemental composition, a multielement technique of comparative instrumental neutron activation analysis capable of simultaneously measuring 17 elements with a high degree of sensitivity and precision was developed to support the AEC's waste storage programs. This approach has proved highly successful in characterizing the local stratigraphy of the Columbia River Basalts using samples taken from test wells.

Briefly, the technique involves a 10 second irradiation of a 100 mg basalt sample together with appropriate standards at a neutron flux of $10^{15}$ n/cm$^2$-sec. After a 2 minute decay interval to reduce the interference from $^{26}$Al, the sample was counted on a Ge(Li) spectrometer for 2 minutes. This irradiation permits the simultaneous measurement of Al, Mg, Ti, Na, V, and Mn. The sample and appropriate standards are then re-irradiated to an integral neutron flux of $10^{17}$ n/cm$^2$, decayed for 5 days and 20 days and counted again on a Ge(Li) spectrometer for periods of 10 and 100 minutes, respectively. The 10 minute count permits the measurement of La, Sm and Nd, while the later 100 minute measurement provides the concentrations of Fe, Co, Sc, Eu, Th, Cr, Hf, Ta, and Tb. It will also be possible to measure Ca from the $^{47}$Sc activity; however, the necessary calibrations are not yet complete. The technique provides a high degree of precision, on the order of 3% except when the detection limit of an element is approached, as demonstrated by 18 analyses of the U. S. Geological Reference Material BCR-1. This high precision is of paramount
importance to the study since the
concentration of some elements varies
by only a few percent between basalt
flows. As an example, the Sc content
 ranged from 36 to 47 ppm in the
Atlantic Richfield Hanford Company
test well DDH-3 from depths of
1680 feet to 2700 feet. However,
other elements show significant dif-
f erences between adjacent flows which
have been successfully employed to
characterize a flow uniquely. For ex-
ample, Cr was noted to vary by 2
orders of magnitude between flows
while varying only 10% within a flow,
while Eu, La, Sm, Tb, and Ta varied
two-to-four-fold. A complete major,
minor, and trace element characteriz-
ation of the Columbia River Basalt
flows promises not only to provide
the necessary information to uniquely
characterize each flow but may also
provide valuable geochemical evidence
bearing on the conditions which ex-
isted at the time of their formation.

Trace Element Content of Aquatic
Organisms from the Columbia River

L. A. Rancitelli and
C. E. Cushing, Jr.

Stable element concentrations in
the environment are an important
parameter in our understanding of the
transport and distribution of radio-
u nuclides throughout an ecological
system. The stable element coun-
terparts of radionuclides not only serve
as carriers and diluents but in some
cases such as Co, Fe, and Zn partici-
 pate in biologically essential func-
tions within an organism. Other ele-
ments such as As, Cd, and Hg, which
are known to be toxic to biological
systems, can produce detrimental
effects in the environment which can
be serious. Thus, it is important
to establish the present levels of
the toxic heavy metals, biologically
essential elements and radiologically
important elements in the environ-
ment. In an effort to accomplish
this end for the Columbia River sys-
tem, an extensive study of 17 ele-
ments in seven Columbia River organ-
isms has been made by instrumental
neutron activation analysis. Dorsal
muscle, liver and kidney samples from
the 6 fish, chiselmouth, carp, squaw
fish, coarse and fine scale suckers,
white fish, and sturgeon, as well as
caddis fly larvae were analyzed for
the following elements: K, Na, Ag,
As, Au, Br, Cd, Co, Cr, Cs, Fe, Hg,
Rb, Sb, Sc, Se and Zn.

As was the case for marine organ-
isms, silver was found primarily in
the liver of the Columbia River fish.
Cadmium levels of 1 to 10 ppm were
found in the kidney, while mercury
levels as high as 6 ppm in the kidney
of a white fish were observed. Since
the sturgeon has a much longer life
span than other fish, a more exten-
sive survey was performed which
included heart muscle, notochord,
gonads, pectoral ray and cartilage
in addition to the dorsal muscle,
liver and kidney. An arsenic con-
centration of 140 ppm (dry basis)
was found in the gonads. The mer-
cury content of all sturgeon samples
was below 0.2 ppm on a fresh weight
basis.

The caddis fly larvae had much
higher levels of Fe (2000 ppm),
Sb (0.11 ppm) and Sc (0.5 ppm) than is normally encountered in biological tissue, probably reflecting the fact that a significant amount of inorganic detrital material was present in the organism.

Radionuclide Distribution in Olympic National Park, Washington C. E. Jenkins, N. A. Wogman and H. G. Rieck

A survey of the radionuclide distribution in precipitation, river water, river sediments, soils, plants, and animals was conducted in the Olympic National Park of Northwestern Washington which has an annual precipitation ranging from 60 to 200 inches in less than 50 miles distance. Because of the high precipitation regime and geographic location, the total fallout radionuclide concentrations per unit area are relatively high. These relatively high concentrations permit the tracing of many of the less abundant and shorter-lived radionuclides through the biosphere.

Thirty-one cosmogenic, naturally occurring, and fallout radioelements were measured in air, rainwater, stream water, sediments, flora, and fauna samples to establish concentration levels and provide data on the relative behavior and fate of radioelements in a forest ecosystem. It was found that the total fallout concentrations per unit area in the Olympic National Forest were the highest observed for any non-nuclear testing area in the world and generally were some three- to four-fold higher than for the Alaskan Arctic region. However, the radionuclide concentrations in an Olympic elk and deer appear 1.2- and 4-fold lower, respectively, than for Arctic Alaskan caribou. Olympic Peninsula residents who consume local produce and wild game have slightly increased levels of $^{137}$Cs over Richland, Washington residents, whose levels are nearly 50- to 100-fold less than in caribou eating Alaskan Eskimos. Two distinct seasonal patterns emerge from Olympic flora fallout concentrations. The first pattern, which is typical of most Olympic flora, shows seasonal variations which are somewhat greater than seasonal variations in air concentrations. The second pattern, which is typical of slow growing mosses and lichens, shows less variation than fallout concentrations in the atmosphere.

A rough approximation of the radionuclide concentration factors from the troposphere through this forest biome are: air to rainwater, $10^6$ to $10^7$; rainwater to flora, $10^2$ to $10^3$; and flora to fauna, 10 to $10^2$. The input of radionuclides to the forest biome is much greater than loss from the system by runoff which indicates a net accumulation. This indicates that pollutant trace elements would also be accumulated. In view of present atmospheric levels of many hazardous materials, detailed studies of the biogeochemical behavior of these materials would be desirable.
### Radionuclide Concentrations for Air and Rainwater
1966-1967 Olympic Peninsula, d/min/liter

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1. Air is sampled bimonthly. Table values represent the average during the rain and/or snow collection period.
2. C is cosmic ray produced radionuclide
3. N is natural radionuclide
4. A is activation produced radionuclide
5. F is fission produced radionuclide
Radionuclide Concentration Factors in Air, Rainwater, and Flora
March 1967 - Upper Hoh River

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1. Creanoea Oregana - Epiphyte
2. Selaginella Serpens - Epiphyte
3. Ephagnum Nitz - Log Top
4. Ephagnum Nitz - Ground Cover
5. Meadow Grass - Mixture
6. Grass - Mixture
7. Polycotomous Nuitum - Sumpi Fum
8. Silica-80 cloth
9. Source of Radionuclide
10. Cosmic Ray + C
11. Naturally Occurring + n
12. Neutron Activated + A
13. Fission Product + F

Subcritical Multiplication of $^{252}$Cf Neutrons and Its Application

N. A. Wogman and R. W. Perkins

Subcritical multiplication of $^{252}$Cf neutrons has been successfully demonstrated to show that it can be accomplished in a straightforward and safe manner. This accomplishment provides solutions to two problems which presently hinder some of the most useful potential applications of $^{252}$Cf. First, it permits high neutron fluxes to be achieved without resorting to extremely large and expensive $^{252}$Cf sources; and second, it permits a 10- to 100-fold reduction in radiation levels for a given shield during the transportation of portable sources. Neutron flux amplifications of some 40-fold have been readily achieved and with proper source sizes will provide fluxes which are adequate for many applications of neutron activation, neutron radiography, and other uses normally considered the domain of reactors. A subcritical demonstration facility consisting of PuO$_2$-polystyrene having a k$_{eff}$ of 0.978, was assembled. At distances of 15 cm and greater where most neutron exposures would be made...
(areas of fairly uniform flux), the flux was enhanced by more than
35-fold over that of an unmultiplied \(^{252}\text{Cf}\) source. Many neutron activation analysis techniques use a thermal neutron flux of \(10^{13}\, \text{n}\cdot\text{cm}^{-2}\cdot\text{sec}^{-1}\) on a 100 mg sample. Comparable induced radioactivity could be produced with \(10^{10}\, \text{n}\cdot\text{cm}^{-2}\cdot\text{sec}^{-1}\) and 100 g samples and this is entirely feasible for many sample types. Utilizing a subcritical multiplier design with a \(k_{\text{eff}} = 0.975\) and optimum flux trap techniques, a 100 mg \(^{252}\text{Cf}\) source would provide 2 to 3 \(\times 10^{10}\) thermal neutrons cm\(^{-2}\) sec\(^{-1}\).

To demonstrate the feasibility and application of subcritical multiplication, a 200 \(\mu\text{g}\) \(^{252}\text{Cf}\) source was employed in the above PuO\(_2\) - polystyrene assembly. Samples of molybdenum ore concentrate, coal and several biological materials were irradiated. A one hour irradiation of a 66 g specimen of molybdenum concentrate and subsequent gamma-ray analysis of the resultant radionuclides with a 4.2% Ge(Li) diode, allowed 16 elements to be analyzed. Some elements such as rhenium were present only at the ppm levels. A much larger primary source of neutrons (10 to 100 mg of \(^{252}\text{Cf}\)) would be adequate for analysis of trace constituents in many large biological specimens. The high flux would also be necessary to analyze trace constituents such as mercury, selenium, antimony and others at the ppm to ppb level in coal.

Additional important applications of neutron multiplication are in producing short-lived isotopes for industry, research and medicine. Neutron multipliers could also provide the necessary thermal flux for a variety of industrial process control applications where conventional methods are not feasible.
Gamma-ray Spectra of Neutron Activated Stibnite

Subcritical Multiplying Demonstration Assembly
The central objective of the METROMEX project is to study convective storms in the St. Louis metropolitan area. During August of 1971 Battelle-Northwest personnel released ruthenium, selenium and gold tracers into the updrafts of convective storms from a ground-based aerosol generator. Samples of rain from these storms, collected using a network of over 100 samplers, are being analyzed for these tracers by neutron activation to determine aerosol scavenging efficiencies and aerosol and hydrometer trajectories. Large volume rain samples were analyzed for the short-lived cosmogenic radionuclides $^{36}$Ar, $^{38}$Cl, and $^{39}$Cl in order to determine aerosol scavenging efficiencies and the elapsed times between scavenging and the arrival of the precipitation at ground level. Air filter samples collected from ground level to 45,000 feet both upwind and downwind of St. Louis are being analyzed to determine the amounts of contaminating materials added to the atmosphere by the St. Louis metropolitan area.

Preliminary Report on Experiments Conducted by Battelle-Northwest During METROMEX

J. A. Young, T. M. Tanner, C. W. Thomas and N. A. Wogman

Field experiments in the METROMEX project were conducted by investigators from several organizations during the summer of 1971 in a 2000 square mile area centered around St. Louis, Missouri. It is planned to continue the experiments for the next four summer seasons. The central objective of this project is to study convective storms, including their dynamics and microphysics. Special attention is being given to determining the effects of the heat and contaminating materials added to the atmosphere from the St. Louis area on precipitation processes and aerosol scavenging in convective storms. The participation of the Radiological Chemistry Section of Battelle-Northwest in the METROMEX experiment has three main objectives. The first is to determine by means of neutron activation and X-ray fluorescence analysis the contribution of the St. Louis metropolitan area to the trace element composition of air and rain. A second objective is to add six inert tracers to convective storms in order to determine scavenging efficiencies and aerosol particle and hydrometeor trajectories by means of neutron activation analysis of precipitation samples. The third objective is to determine aerosol scavenging efficiencies and times between scavenging and arrival of the precipitation at ground level by measurements of cosmogenic radionuclide concentrations in precipitation.
Battelle personnel conducted field experiments for the METROMEX project from August 1 through August 20 during 1971. RB-57 aircraft were used to collect air filter samples at 3, 6, 9, 12 and 15 thousand feet both upwind and downwind of St. Louis to determine the contribution of the St. Louis metropolitan area to the atmospheric aerosol. Samples were collected two days a week on each of the first three weeks of August. Air samples were also collected for 24 hour sampling periods using hurricane air samplers at Scott Air Force Base and at a farm southwest of St. Louis (see Figure). Normally the farm is upwind of St. Louis and Scott AFB is downwind. Anderson and Lundgren impactors were used to separate airborne particulates into different particle size ranges. These samples were brought back to the Richland laboratory for trace element analysis by neutron activation and X-ray fluorescence analysis.

Tracers were released into the atmosphere from ground level by means of an aerosol generator which produces submicron sized particles of a tracer material by burning acetone solutions of the tracer. The tracers were released for 30 or 40 minutes immediately preceding the arrival of a convective cell at the generator site, in an attempt to introduce the tracers into the updraft of the cell. Ruthenium, gold and selenium were released sequentially during the 40 minutes prior to the arrival of a cell soon after midnight on August 11. Ruthenium was released prior to the arrival of a convective cell on the afternoon of August 14, and gold was released during the evening of August 14. Following each tracer release, samples of rain were collected using a network of over 100 rain samplers in the St. Louis area. The samples were frozen and returned to the Richland laboratory where they are currently being analyzed in an attempt to determine the trajectories and precipitation scavenging efficiencies of the tracer materials.

Large volume rain samples were also collected for short-lived cosmogenic radionuclide analysis by means of a 1590 ft² tarp located at Scott AFB. Unfortunately, rain fell at Scott AFB during the experimental period only on August 14. The activities of the short-lived cosmogenic radionuclides in the rain were similar to those measured at Quillayute, Washington, suggesting that the cosmogenic radionuclides present in the rain came primarily from low altitudes and did not enter the convective cell at mid-tropospheric levels. Cosmogenic radionuclide activities are two orders of magnitude higher at mid-tropospheric levels than at ground level.
METROMEX Rain Collection Network
Chemical Characterization of Urban Aerosol
L. A. Rancitelli and T. M. Tanner

The atmospheric aerosol is derived from a variety of natural and man-made sources, each with a particular chemical composition. The major natural sources include the sea salts from the oceans and terrestrial dust derived from the contents. The anthropogenic sources of the atmospheric aerosol are too numerous to tabulate, but can in general be ascribed to major population centers. It has become evident through our studies of the chemical composition of the high altitude aerosol and rain water collected on the west coast of the State of Washington, that the northern hemisphere concentrations of a number of elements have been increased by orders of magnitude as a result of man's activities. Thus, any model describing the mechanisms responsible for the distribution, transport and deposition of aerosol must consider the material emanating from urban centers. In an effort to characterize the chemical composition of urban aerosols, a highly sensitive and precise technique of instrumental neutron activation analysis was developed that is capable of simultaneously measuring over 30 elements on air filters used to sample the urban aerosol at ground level.

The technique involves a one-minute irradiation of the filter samples together with the appropriate standards at a neutron flux of $1.3 \times 10^{13}$ n/cm$^2$ sec. The sample and standards are allowed a 10-minute decay interval to reduce interference from $^{28}\text{Al}$ and then counted for 5 minutes on a Ge(Li) spectrometer. This permits the simultaneous measurement of Al, Br, Cl, Mg, Mn, Na, Ti and V. The samples and standards are then re-irradiated to an integral neutron flux of $10^{17}$ n/cm$^2$, allowed to decay for 5 and 20 day periods, and counted on a Ge(Li) spectrometer for 30 and 100 minutes, respectively. The 50-minute count permits the measurement of Na, K, La, As, Br, Sm, U and Au, while the 100-minute count provides the concentration of Fe, Co, Sc, Eu, Th, Cr, Sb, Rb, Ag, Se, Cs, Hf, Tb, Te and Hg.

The filter samples which are placed in clean polyethylene capsules for the irradiation are 30 cm$^2$ sections taken from a total area of 400 cm$^2$ of a Whitman 41 filter paper. The volume of air passed through these filters is usually in the range of 1000-1500 m$^3$. While the results are preliminary, several important observations concerning the origin of elements in the urban aerosol can be inferred from the ratio of trace element concentrations. Our high altitude trace element studies indicate
that the Sc content is a valid baseline indicator of natural earth's crust material. When the trace element concentrations are normalized to the scandium content of a filter and compared with similar ratios in major earth's crust materials such as granite or diabase, it is evident that the As, Sb, Zn, Hg and Se levels have been increased by several orders of magnitude, while Fe, Mn and Cr show enrichments of an order of magnitude in urban centers. Elements such as La, Eu, Hg, Fe, Al and Co appear to exist at levels compatible with an earth's crust origin.
The radionuclide depletion processes in the Columbia River are being characterized following the closure of the Hanford reactors. Residual radioactivity is associated mainly with sedimentary deposits behind McNary Dam. The most abundant radionuclides are 55Fe, 60Co, 99Mo, 133Cs, 152Eu, 185Ta, and 189Re. These radionuclides are bound to the sediments and enter the river by sediment resuspension and dissolution into soluble forms. The main mechanism for depletion of radioactivity from the river, other than radioactive decay, is by scouring of surface sediments during high river flow, and their subsequent transport into the Pacific Ocean. It appears that eventually the radioactive sediment deposits behind McNary Dam will become covered by new, uncontaminated silt deposits. The high fraction of radionuclides in the particulate form in Columbia River water changes their availability for biological uptake.

Transport and Depletion of Radionuclides in the Columbia River
D. E. Robertson, W. B. Silker and J. C. Langford

With the closure of the KE plutonium production reactor in January, 1971 essentially no additional radioactivity has been directly discharged to the Columbia River in reactor effluent water. However, some residual radioactivity has remained in the river, being associated mainly with the sediments and the resuspended load. The Columbia River is, therefore, an extremely valuable natural laboratory for studying the release of radionuclides from river sediments, and for evaluating the natural cleansing processes of river systems following intentional or accidental releases of radioactivity. In addition, these radionuclides serve as excellent tracers for characterizing the rates and mechanisms of sediment transport in the Columbia River system.

In April, 1971 sampling programs were initiated to study the distribution and behavior of radionuclides in Columbia River sediments, suspended matter, and water between Hanford and Bonneville Dams.

Sediment cores were taken behind McNary and The Dalles Dams using a 6 in. diameter gravity corer. No large sediment deposits could be located behind John Day and Bonneville Dams. The cores were frozen immediately after collection and then sliced into 2.5 cm thick slabs, which were trimmed, oven-dried, homogenized and placed in 4 in. diameter by 1/2 in. thick plastic counting dishes. The sediments were counted on a multidimensional gamma-ray spectrometer and an anticoincidence shielded Ge(Li) diode detector.
A typical gamma-ray spectrum of McNary Dam surface sediments, as counted on an anticoincidence shielded Ge(Li) detector, is shown in the accompanying figure. Ten radionuclides of Hanford origin are present in the sediments including $^{46}\text{Sc}$, $^{54}\text{Mn}$, $^{60}\text{Co}$, $^{65}\text{Zn}$, $^{125}\text{Sb}$, $^{137}\text{Cs}$, $^{144}\text{Ce}$, $^{152}\text{Eu}$, $^{154}\text{Eu}$ and $^{155}\text{Eu}$. Iron-55 was measured in some sediments after radiochemical separations and was found to be the most abundant radionuclide. The typical radionuclide composition of surface sediments behind Mcnary Dam is shown in an accompanying illustration. Although $^{55}\text{Fe}$ is the most abundant radionuclide in the sediments, it emits only 5.9 keV X-rays and Auger electrons during its disintegration, and from external dose considerations is of minor importance. Analyses of selected core samples for plutonium radionuclides are in progress.

The depth distribution of seven radionuclides of Hanford origin in the sediments behind Mcnary Dam is of particular interest. The concentration of the shorter half-life radionuclides $^{46}\text{Sc}$, $^{54}\text{Mn}$ and $^{65}\text{Zn}$ decrease rapidly with depth in the core, whereas the long-lived components $^{60}\text{Co}$, $^{137}\text{Cs}$, $^{152}\text{Eu}$, $^{154}\text{Eu}$ and $^{155}\text{Eu}$ can be found at a depth of 50 cm at concentrations comparable to surface values. The peak in radionuclide concentrations at a core depth of about 28 cm corresponds to a maximum release of radioactivity to the river in June, 1967. The sediment deposition rate at this core location behind Mcnary Dam between June, 1967 and April, 1971 can then be extrapolated to be about 7 cm/year. Sediment deposition rates calculated from changes in the ratios of $^{46}\text{Sc}/^{65}\text{Zn}$ and $^{65}\text{Zn}/^{60}\text{Co}$ with depth in the sediment core give 7 cm/year and 10 cm/year, respectively.

Because of the very low concentrations of radionuclides in Columbia River water, Battelle Large Volume Water Samplers were used to remove particulate and soluble forms of radionuclides from 500 to 1000 liters of river water. Particulate material was removed on a series of 12 in. diameter fiberglass filters, whose retention efficiencies were shown to be comparable with 0.3 μm membrane filters. The filtered water was then passed through 12 in. diameter by 1 in. thick beds of cation and anion exchange resins to remove dissolved ionic species, and finally through a 12 in. diameter by 1/4 in. thick bed of activated aluminum oxide to remove non-ionic constituents. The flow rate through the filtration-sorption assembly was maintained at 9 liters/minute. The filters, aluminum oxide beds and aliquots of the cation and anion beds were counted directly on high sensitivity multi-dimensional gamma-ray spectrometers to measure the radionuclide concentrations contained on each material.

River water is periodically sampled from the generator cooling supply at Mcnary, John Day, The Dalles and Bonneville Dams. These sampling locations within the dams were chosen because the generator cooling waters represent vertically mixed water from behind the dams, and are therefore more characteristic of the dynamic flow of the river.
The relative distribution of radionuclides in the Columbia River between particulate matter (>0.3μ), cationic, anionic and non-ionic species indicates that nearly all of the radioactivity in the water is present in the form of resuspended sediments (see table). These radionuclides remain very tightly bound to the sediments, and any leaching of radionuclides from the sediments to form soluble species is very small. It is interesting to note that the 106Ru present in the river as fresh fallout is mainly in a non-filterable form, which is retained on the Al2O3 bed. This would indicate that the 106Ru is present in either a nonionic form or a colloidal species.

The transport of 65Zn on resuspended sediments between McNary and Bonneville Dams is shown in an accompanying illustration. Most of the sediments behind McNary Dam are of Snake River origin and are deposited mainly on the Oregon side of the river. Because of this influence of Snake River water and sediments, it was necessary to sample from both the Oregon and Washington sides at McNary Dam in order to differentiate the contribution of Snake River water to the Columbia River. The 65Zn concentrations in the suspended matter collected from both sides of the dam clearly show this influence. The Snake River flow began peaking around the middle of March and increased the scouring of the main deposits of radioactive sediments on the Oregon side of the river behind McNary Dam. The higher radionuclide concentrations in the resuspended sediments on the Oregon side of the river became less pronounced after the Columbia River flow had peaked during the first week in April. The transport of 65Zn (and other radionuclides) on resuspended sediments between McNary and Bonneville Dams indicates no major systematic redeposition of these suspended sediments in the slack water pools behind the dams downstream from McNary. It appears that once surface sediments have been resuspended during the spring freshet, the major portion of them is flushed completely out of the river and into the ocean. The absence of any major deposits of fine sediments between McNary and Bonneville Dams supports this observation. With the recent construction of John Day Dam some sedimentation must occur in the slack water behind this dam, but the transport rates of resuspended radioactive sediments indicates this is not a major sedimentation basin.

As indicated in the accompanying illustration, there is a direct relationship between river flow rate and transport rates of radionuclides past Bonneville Dam. The rapid increase in radionuclide transport correlated with the spring freshet, and then decreased during the period in which river flow remained at a maximum. Apparently the spring freshet immediately scour off a layer of the finest surface sediments and then resuspends the underlying sediments at a slower rate. The rapid decrease in the radionuclide transport rate closely followed the decline in river flow rate, and then leveled off after August when the low river flow stabilized. The total curies of radioactivity of Hanford origin (excluding
$^{55}\text{Fe}$ transported past Bonneville Dam and presumably into the Pacific Ocean between February and December, 1971 is estimated to be about 600 curies. This may be compared with annual discharges on the order of 350,000 curies/year during the early 1960's when eight production reactors were operating.

The main mechanism for depletion of radionuclides (other than radioactive decay) from the Columbia River is by the resuspension of surface sediments during the spring freshet and high water runoff, and the subsequent transport of the suspended sediments to the ocean. Since the net deposition rate of $^{134}$, nonradioactive sediments (mainly of Snake River origin) behind the dams is apparently greater than the scouring losses of old sediments by high runoff, it is probable that the radioactive deposits will eventually become buried by fresh layers of uncontaminated silt. The extent of this burial and the resulting decrease in river concentrations will be defined as this study progresses.

Iron-55 Concentrations in Columbia River Sediments, 1971 Units of D/M/g

<table>
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<th>Sediment Depth, cm</th>
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<tr>
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<tr>
<td>Surface</td>
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<td>178 ± 7</td>
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<td>366 ± 9</td>
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<td>McNary Dam - Washington Side</td>
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<tr>
<td>8</td>
<td>1250 ± 30</td>
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<tr>
<td>10</td>
<td>701 ± 18</td>
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<tr>
<td>13</td>
<td>460 ± 13</td>
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<tr>
<td>15</td>
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<td>18</td>
<td>385 ± 13</td>
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<tr>
<td>20</td>
<td>291 ± 11</td>
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<tr>
<td>The Dalles Dam</td>
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<tr>
<td>5</td>
<td>87 ± 3</td>
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<tr>
<td>8</td>
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<td>10</td>
<td>70 ± 3</td>
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Typical Composition of Photon Emitting Radionuclides in Surface Sediments Behind McNary Dam, April, 1971
Columbia River Surface Sediment—McNary Dam
Typical Distribution of Physico-Chemical Forms of Radionuclides in the Columbia River - McNary Dam, July 13, 1971

Percentage Distribution on Filters, Resins and $\text{Al}_2\text{O}_3$

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<tr>
<th>Unit 2 - Oregon Side</th>
<th>$^{46}\text{Sc}$</th>
<th>$^{60}\text{Co}$</th>
<th>$^{55}\text{Zn}$</th>
<th>$^{106}\text{Ru}$</th>
<th>$^{152}\text{Eu}$</th>
<th>$^{228}\text{Th}$</th>
<th>$^{226}\text{Ra}$</th>
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<td>98.4</td>
<td>70.0</td>
<td>99.9</td>
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<td>1.6</td>
<td>2.5</td>
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<tr>
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<td>&lt;0.1</td>
<td>0</td>
<td>0</td>
<td>0</td>
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<tr>
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<td>0.5</td>
<td>100.00</td>
<td>0</td>
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<tr>
<td>Particulate ($&gt;0.3\mu$)</td>
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<td>98.6</td>
<td>99.3</td>
<td>39.4</td>
<td>97.3</td>
<td>100.0</td>
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<td>9.7</td>
<td>0.4</td>
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<td>1.1</td>
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<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Nonionic</td>
<td>0.3</td>
<td>0.7</td>
<td>0.4</td>
<td>60.6</td>
<td>1.5</td>
<td>0</td>
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Transport of $^{65}\text{Zn}$ in the Columbia River Between McNary Dam and Bonneville Dam

Transport of Radionuclides Past Bonneville Dam April Through November, 1971
RADIOLOGICAL PHYSICS

DOSIMETRY OF RADIONUCLIDES IN MAN

These studies are concerned with the continued development and improvement of diagnostic techniques for the assessment of internal depositions of radiotoxic materials in man and the development and improvement in dosimetry associated with such depositions.

Promethium
D. B. Shiplet,* J. E. Ballou,** D. McConnon*** and I. C. Nelson

Efforts continued toward development of a method for diagnosing the retention of promethium in the bodies of workers following accidental inhalation of $^{147}$Pm$_2$O$_5$. Ultimately a quantitative diagnostic method is expected to be obtained as a result of human testing using promethium-143 in place of promethium-147 in an oxide aerosol. Promethium-143 facilitates in vivo detection with low doses to participating volunteers. However, the mass of promethium-143 based on detection capability and acceptable dose is inadequate for the formation of realistic aerosols. Since there is no stable isotope of promethium, samarium is under investigation as a carrier for promethium.

Experiments involving intravenously injected promethium-147 and samarium-145 chlorides in citrate buffer solution in rats disclosed that no gross differences occurred in the metabolism of the two radionuclides in that species. (1)

During the year efforts centered on assessing the feasibility of using samarium as a carrier for promethium-143 in inhalation studies using the rat. Thirty-six rats were exposed (nose only) to an aerosol consisting of particles of $^{145}$Sm$_2$O$_5$, $^{145}$Sm$_2$O$_5$ and Sm$_2$O$_5$ having an aerodynamic median activity diameter of 3.4 μm and a geometric standard deviation of 1.6. One group was sacrificed immediately and the lungs were analyzed to determine the initial lung burden.

Periodic collections of urine and feces were made for each rat. Groups of rats were sacrificed at 14 and 30 days following exposure and lungs, liver, kidneys, head, muscle, pelvis and skeleton were sampled at necropsy. The samples were analyzed using coincidence gamma scintillation counting. All samples for which analysis
was intended have been analyzed. Numerical analysis of the data continues. Preliminary results indicate that the ratio of samarium to promethium in tissue samples agrees favorably with the ratio of the nuclides in the aerosol and suggests that samarium will be usable as a carrier for promethium-145 in inhalation studies.

Plutonium
P. J. Dionne* and I. C. Nelson

A computer model predicting the excretion, translocation and retention of plutonium in the human body following intravenous injection of plutonium nitrate was developed through comparison of such data obtained by Langham, et al. (2) for plutonium citrate in humans, Stover, et al. (3) for plutonium citrate in beagle dogs and Bair, et al. (4) for plutonium nitrate in beagle dogs. Recognizing the possible limitations of the model due to differences in pH, chemical composition and amounts of the injected material, prediction factors were estimated relating results of the human and dog models for injected plutonium citrate. These prediction factors along with the dog model for plutonium injected in nitrate form were used to develop predictions of initial whole body, bone and liver burdens given the daily amount of plutonium excreted in urine for injection of plutonium nitrate in humans. Pres. / results, which are subject to revision as the model is refined, suggest that the total body burden of plutonium following intravenous injection of plutonium nitrate can be estimated from

\[ B_{TB} = 2.4 \times 10^3 \text{ U} \exp \left( \frac{0.693t}{1.3} \right) 1 \leq t \leq 8 \]

where \( B_{TB} \) is the total body burden in the same units as the daily urine excretion, \( U \), and \( t \) is time in days. Similarly, the bone burdens can be estimated from

\[ B_{Bone} = 1.9 \times 10^2 \text{ U} \exp \left( \frac{0.693t}{1.3} \right) 0 \leq t \leq 136 \]

For comparison, the data presented by Langham, et al. (2) for injection of plutonium citrate suggests a total burden of \( B = 4.5 \times 10^2 \text{ U} \exp 0.77 \)

\[ 0 \leq t \leq 136. \]

Further study and manipulation of the model is expected to yield estimates of the most probable organ burdens over longer time intervals.

REFERENCES CITED


* Systems Engineering Department, Battelle-Northwest.
CHARACTERIZATION OF RADIATION-INDUCED FREE RADICAL REACTIONS IN AQUEOUS SYSTEMS

This project is concerned with determining the chemical structures and reactivities of free radicals formed in irradiated model systems in order to understand and alleviate the development of radiobiological damage. Electron spin resonance spectroscopy and optical absorption spectrophotometry have been used to identify the radicals and determine the kinetics of their reactions with other biochemical substrates in aqueous solutions. The damage-propagating abilities of the radicals have been assessed by studying their reactivities toward diagnostic reagents, while methods for chemical protection from free-radical damage have been tested in various biochemical systems.

Chemical Reactivities of Radicals in Aqueous Solution
D. R. Kalkwarf, H. W. Fenrick*
and R. N. Diebel

A systematic study of relations between the structures and reactivities of radiation-induced biochemical radicals in aqueous solution was continued. Radicals were formed in irradiated crystals of biochemical compounds or irradiated samples of their solutions in ice since in these forms their concentrations and general structural features could be readily determined by electron spin resonance. Bleaching of eriocin at pH 6 and of p-nitrosodimethylaniline at pH 9 were again used to detect the presence of oxidizing radicals, while one-electron reduction of flavin mononucleotide was used to detect reducing species.

The most numerous class of radicals found in irradiated biochemical crystals is the carbon radical, i.e., a radical in which the unpaired electron is predominantly associated with a particular carbon atom in the molecule. Radicals containing a carboxylate group adjacent to this carbon atom were found to be particularly potent oxidizing agents. Examples include $\text{C}_2\text{H}_3\text{C}=\text{C}=\text{O}$ derived from malonic acid, $\text{C}_2\text{H}_3\text{C}=\text{C}=\text{O}$ derived from succinic acid, and $\text{C}_2\text{H}_3\text{CO}$ derived from glycine. When positively-charged or other electron-withdrawing groups were present in the radical, oxidizing ability was greatly decreased. This was illustrated by $\text{C}=\text{C}(\text{OH})(\text{COOH})\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\cdot\•...
comparing the ESR spectra and chemical reactivities of radicals formed in both dry crystals and in 1M ice solutions irradiated at -196°C. For many of the samples investigated, e.g., glycine, α-alanine, lactose, valine, ESR spectra of the radicals formed in ices were either very similar to those seen in irradiated crystals of the compound at room temperature or became similar as the ice was warmed. Exceptions were found, however, e.g., maltose and fructose. Whether formed in irradiated crystals or ices, radicals derived from α-alanine, galactose and maltose were found to be oxidizing agents while the radicals derived from ε-alanine were found unreactive toward the diagnostic reagents.

Until more extensive comparisons can be made, it was concluded that hydration generally does not alter the chemical structures of biochemical radicals and that their reactivities reported here and in past reports should be applicable in irradiated living tissue.

**Radical Reactions of Biochemical Membranes**

D. R. Kalkwarf

Research was begun to test whether free-radical attack at biochemical membranes could be a particularly significant means for amplifying radiation damage in living systems. A model membrane system was developed which can be made extremely sensitive to changes in chemical structure and should be a good indicator of free-radical or other types of radiation damage. This membrane consists of a protein-activated lipid bilayer separating two aqueous compartments, and the electrokinetic phenomena occurring at the interface can be made to closely resemble excitable cell membranes.

In preparation for these experiments, ESR spectrometry and diagnostic reagents were used to detect the presence and reactivities of any radicals formed in gamma-irradiated crystals of the membrane-forming lipids and related compounds. Irradiated glycerides, e.g., tripalmitin, the various dipalmitins and tristearin, were found to form R1-CH2-CH-COOR2 radicals.

In contrast, irradiated phospholipids such as α,γ-dipalmitoyl-α-lecithin and stearoylsphingomyelin gave no ESR signals. Steroids such as progesterone, cholesterol and cholesterol acetate gave complex spectra which have yet to be interpreted.

Radical-induced oxidation of diphenylpicrylhydrazyl in chloroform and polymerization of vitamin E in benzene were used to evaluate the reactivities of these radicals. All of the radicals detected by ESR spectrometry were found to bleach diphenylpicrylhydrazyl. Only radicals derived from dipalmitin and tripalmitin have been tested as yet with vitamin E, but both of these were found to dimerize the reagent as detected by the increased optical absorption at 305 nm. These experiments show that radicals derived from naturally occurring lipids can extend the effects of radiation damage and that their behavior in lipid membranes could be of great significance in the development of this damage.
Flash Photolysis of Nucleic Acid Components
W. D. Felix

The reaction kinetics of the radiolysis products of water, i.e., the hydroxyl radical (OH) and the hydrated electron (e\textsubscript{aq}) have been characterized for many substances of biological interest. In most cases, the products of these reactions are, themselves, highly reactive free radicals. For example, in the biological environment, it would be expected that the products of hydroxyl radical reaction with the chloride or carbonate ions of the body fluids will result in the formation of the free radicals Cl\textsuperscript{+} and CO\textsubscript{3}\textsuperscript{2-}. These radical ions are expected to be strong oxidizing agents and should be expected to significantly contribute to the general disruption of cellular processes as they interact with cellular components.

In this study the reactions of the CO\textsubscript{3}\textsuperscript{2-} radical ion are being investigated in order to determine its relative biological damage potential. In preparation for this study, the flash lamp assembly developed in this laboratory has been improved. Because of the complexity of the mechanisms involved, it has also been necessary to develop an effective data handling system. Flash photolysis data are collected directly with a digital memory oscilloscope and transferred to paper tape for analysis, or photographs of oscilloscope traces are analyzed with a semi-automatic reader for conversion of information to punched paper tape. Edit programs then convert the punched paper tape into a computer compatible format.

Simultaneously with this development work, programs were also written to synthesize ESR spectra whereby direct comparison with experimental data may be made for use in the identification and characterization of the radical products.

Reaction of the CO\textsubscript{3}\textsuperscript{2-} radical as formed during flash photolysis is indicated by a change in the absorption of light at the 600 nm absorbance peak. Rate constants determined for the decay of CO\textsubscript{3}\textsuperscript{2-} in the presence of uracil and cytosine (at 2 \( \times \) \( 10^{-4} M \) concentration) indicate that CO\textsubscript{3}\textsuperscript{2-} is only slightly less active than Cl\textsuperscript{+} in these two pyrimidine systems. The rate constants, 0.9 \( \times \) \( 10^{7} M^{-1} \) sec\textsuperscript{-1} and 2 \( \times \) \( 10^{7} M^{-1} \) sec\textsuperscript{-1} for reaction with uracil and cytosine respectively, are approximately one fourth of the magnitude of the constants for the respective reactions with Cl\textsuperscript{+}.\textsuperscript{(2)} These initial results indicate that the reactions of the secondary radicals, such as CO\textsubscript{3}\textsuperscript{2-}, can be significant in consideration of the overall radiation damage picture. However, three factors mitigate against the inclusion of CO\textsubscript{3}\textsuperscript{2-} radical reactions in most systems: 1), the rate constants for reaction of OH with Cl\textsuperscript{-} is 1.5 \( \times \) \( 10^{10} M^{-1} \) sec\textsuperscript{-1} \textsuperscript{(2)} compared with 1.1 \( \times \) \( 10^{6} \) to 3.7 \( \times \) \( 10^{5} \) (pH dependent) for CO\textsubscript{3}\textsuperscript{2-}; 2), the rate constants of reaction of Cl\textsuperscript{+} with the pyrimidines are four times larger than those of CO\textsubscript{3}\textsuperscript{2-}; and 3), the concentration of Cl\textsuperscript{-} in the usual cellular fluids is much higher than that of CO\textsubscript{3}\textsuperscript{2-}. Thus, in most biological systems, reaction of Cl\textsuperscript{+} with cytosine and uracil will be significantly more important than the reaction of CO\textsubscript{3}\textsuperscript{2-}.
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NUCLEAR TECHNIQUES IN MEDICAL SCIENCE

The purpose of this study is to perform research in the application of nuclear technology and radioisotopes to the fields of medicine and biology and to combine the advanced knowledge, techniques, and instrumentation of the nuclear field with the talent and knowledge of the medical profession to improve and increase the direct medical benefits to mankind. Advanced instrumentation concepts, detectors and circuitry are also developed for use in the fields of health physics and environmental studies.

Absolute Measurement of Total Body Calcium by In Vivo Neutron Activation Analysis
H. E. Palmer
with
W. B. Nelp, J. D. Denney, R. Murano,
T. G. Rudd, University of Washington
Medical School, Seattle, Washington.

A method for measuring serial changes in total body calcium in man has been in use for approximately two years. The uniformity of activation, precision, and reproducibility of the measurement are such that a 2% change in calcium content is detectable. (1,2) With variations among people in body geometry and the amount of tissue moderator surrounding the skeleton, absolute determinations of total body calcium were possible with an accuracy of ±8 to 10%. In order to improve the absolute accuracy of this method, five cadavers of various body sizes were accurately irradiated and counted several times. The complete skeletons were then carefully ashed by slow heating in a large muffle furnace after which the calcium content of the ash was accurately determined by an atomic absorption method. The milligrams of calcium per gram of bone ash were remarkably constant from skeleton to skeleton, averaging 366 mg ± 1.9 mg S.D.

As expected, it was found that for any given height, skeletal activation would be somewhat less in the heavier subject as compared to a leaner one. The skeletal activity of the $^{40}$Ca was correlated with an expression of "Body Form" relating the weight per unit height. This expression is weight in kilograms divided by height in meters, i.e., kg/m. There is a linear relationship over the range of cadaver body sizes studied. By using this graph, one can enter the body form, kg/m of any subject, determine the predicted specific activity based on the chemically analyzed series of cadaver standards and then calculate the body calcium. Statistical analysis of the data permits estimation of the accuracy for measuring body calcium in grams in any given individual. By incorporating the known counting errors and the previously determined precision of a single activation,
with the data in the coefficient of variability for estimating body calcium from a single-measurement is approximately 5.21.

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In Vivo Production of $^{37}$Ar and $^{41}$Ar From Fast Neutron Irradiation of Calcium

H. E. Palmer

$^{37}$Ar and $^{41}$Ar can be produced by an n,a reaction of fast neutrons with $^{40}$Ca and $^{44}$Ca respectively. In ani-
where both the Auger electrons and X rays from the $^{37}$Ar are counted with almost 100% efficiency. The background of the proportional counter under the 2.6 keV photopeak is less than 1 count per minute which allows very sensitive determination of the $^{37}$Ar.

Many more studies must be done to determine if expired $^{37}$Ar can be used as a measure of total body calcium. Preliminary studies provide the following results and conclusions.

The use of $^{37}$Ar as a measure of body calcium is much preferred over $^{41}$Ar because it has less interference from other elements and its overall counting efficiency is more than 100 times better than that for $^{41}$Ar. Therefore a much lower neutron radiation dose can be used.

Experiments involving the simultaneous irradiation of both a live and dead rat indicate that 85% of the $^{37}$Ar is exhaled within 30 minutes after irradiation.

Irradiation of $\text{Ca(NO}_3\text{)}_2$ solutions and subsequent counting of the $^{37}$Ar produced indicate that a 0.01 rad dose to an adult human would provide a count rate of 675 counts per hour over a background of 6 counts per hour. A 15 hour count would provide about 10,000 net counts over a background of 90 counts. The proportional counter is very stable and could be used to count the sample for several days in which case the dose could be reduced to 0.001 rad and the $^{37}$Ar counted for 6 days to provide a net count of 10,000 over a background of 900 counts.

If it is found that essentially all the $^{37}$Ar is expired from the body with in an hour, it is expected that this technique will provide an accurate measure of total body calcium. The technique would eliminate the need for a whole body counter and its associated errors due to different body geometries and self absorption. The radiation dose required by this technique could be only 1% of that currently used for the determination of total body calcium by in vivo neutron activation analysis.

The Determination of Cadmium In Vivo by Measurement of Prompt Gamma Rays Following Neutron Capture

H. E. Palmer, L. W. Brackenbush and L. G. Faust

Cadmium can constitute a serious health problem. Exposure to cadmium in industrial as well as general environments has caused serious intoxications in human beings. Cadmium tends to accumulate in the body and most of it becomes deposited in the kidneys and liver. Animal experiments show that excretion of cadmium via urine and the gastrointestinal tract is very low and related to body burden only in a minor degree. A method to measure existing levels in living humans is needed so that cadmium toxemia can be recognized, treated, or prevented.

Since cadmium has a relatively large cross section for thermal neutrons (20,000 barns for $^{113}$Cd), prompt gamma ray measurement during low level neutron irradiation of the liver or kidneys may provide a method for determining cadmium concentrations in these organs. The major prompt
gamma ray emitted during neutron capture has an energy of 559 keV. Preliminary studies were conducted using a collimated beam of neutrons from a 252Cf source which irradiated a water phantom simulating the human liver surrounded by body tissue. A 50cc volume Ge(Li) detector having a resolution of about 2 keV for the 559 keV photopeak was used to measure the prompt gamma rays to produce a spectrum obtained during a one hour irradiation of the phantom in which the liver contained a solution of 4 grams of cadmium per liter. Most of the interfering Compton continuum is produced from captured gamma rays from hydrogen and lead. The radiation dose to the liver during this measurement was approximately 10 millirem. As little as 20 mg cadmium per liter has been detected in the phantom and this is the lower limit of detection.

The normal content of cadmium in kidney and liver adult humans is about 10 and 5 mg respectively. Assuming that the masses of the liver and kidneys are 1700 and 300 grams, respectively, the normal concentrations are about 3 and 33 mg per kilogram of tissue. The method as described above will not detect these levels, but with improvements in shielding and collimation the sensitivity might be extended down to these concentrations. Toxicity effects of cadmium become manifest at kidney concentrations of about 140 mg per kilogram. This method in its present state of development could detect such levels.

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2. Ibid, p. 37-66

3. Ibid, p. 108
Radiation Instrumentation.

K. L. Swinth

Emphasis on the detection of plutonium translocated to the tracheobronchial lymph nodes continued with the evaluation of advanced probes and thorough analysis of counting geometries. Although advanced probes have displayed more uniform angular sensitivity, expected increases in counting efficiency have not been obtained. With the larger area diodes employed in these probes, one obtains large areas of the surface with poor counting efficiency. With a typical 2 cm long detector, only 1.2 cm of the length could be considered effective in counting the plutonium X rays. Because of this lack of uniformity, detection limits are higher than originally projected.

The minimum detectable amount (MDA) for the tracheobronchial nodes in the human phantom is 8 nCi under optimum counting conditions with a 20 minute counting period. This was with a single 1 cm long detector—not with the most recent three element probe which had a minimum detectable amount of 15 nCi under
similar conditions. To obtain the required uniformity of angular response a multi-element probe is necessary. The measured angular sensitivity of the three element probe is uniform to ±3% whereas the sensitivity of a single element probe drops to almost zero at some angles. It was found that a shaping time constant of 2 usec was optimum with our system and that anti-coincidence operation of the detectors in the three element probe did not significantly reduce the background count rate.

The tracheobronchial lymph nodes of dogs are smaller and more uniform than those of humans; however, dogs were used to aid in the evaluation of the probes. Analyzing experimental data obtained from dogs it was found that an average MDA of 30 nCi was obtained for the middle tracheobronchial node and an MDA of 16 nCi was obtained for the right tracheobronchial node when employing a one minute counting period. Counting sensitivity was consistently higher for the right tracheobronchial node which is apparently in a more favorable counting position. Because of the high plutonium concentrations in the lymph nodes of the dogs, self absorption of the X rays was considered; however, it was found by calculation that less than 10% of the X rays would be absorbed.

Calculations were performed and available materials investigated for optimization of a fiber optics coupled scintillator for the detection of 17 keV X rays from plutonium. Limiting the sensitive area of the photocathode of the phototube along with improvements in the light transmission of fiber optics should result in increased sensitivity and decreased background rate compared to previous prototype systems. MDA's would then be comparable to the avalanche probes; however, the counting would depend primarily on the detection of 60 keV gammas from 241-Am. The system is being developed for both wound counting and in vivo counting of lymph nodes.

Wound visualization studies have centered around optimization of an image intensifier system. NaI(Tl), CaWO4, and ZnS(Ag) scintillators have been used on the system with the clearest images resulting from the ZnS(Ag). Although the other scintillators are more efficient, internal reflections and light scattering tend to "wash out" the image. Because one must obtain a large number of events to produce a recognizable image, such a system will be limited to high activity contaminations. A matrix of detectors with larger "resolution elements" or a position-sensitive proportional counter that could be placed in contact with the wound would be more appropriate in most cases.

Measurements of X-ray-alpha ratios were compiled for a number of transuranic isotopes and provided to members of the "Intercalibration Committee for Low-Energy Photon Measurements." Reported intensities for X rays are rare and differences of greater than a factor of three are evident for some low-energy gamma rays that have been proposed for in
in vivo plutonium counting. Plutonium whole body counter intercalibration sources are being routed among interested laboratories; however, only a preliminary analysis of the data has been performed. When the routing of the sources is completed the data will be analyzed in detail.

**Neg 712954-2**

Scans Along Length of the Detectors in the Three Element Probe Showing Loss of Sensitivity Near Center of Probe. This is due to non-uniformities in the response of the avalanche detectors.

<table>
<thead>
<tr>
<th>Counting Sensitivity for Right and Left Tracheobronchial Lymph Nodes in Beagle Dogs Based on Post-mortem Analysis of Tissues</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dog</td>
</tr>
<tr>
<td>329</td>
</tr>
<tr>
<td>248</td>
</tr>
<tr>
<td>335(a)</td>
</tr>
<tr>
<td>344(a)</td>
</tr>
<tr>
<td>Unweighted Average</td>
</tr>
</tbody>
</table>

a. Post mortem experiment
b. Counts with probe D-1, but normalized to probe LA-3
Scans made in a Human Thorax Phantom Showing Increase in Count Rate Near Lymph Node Sources with an Optimum Detector Configuration. The inter-tracheobronchial source is at the 2 cm position and the laterotracheal sources are at 5 cm.
MECHANISMS OF ENVIRONMENTAL EXPOSURE

This project involves a continuation of the environmental study to measure the decline of radioactivity in various population groups and to reduce, compute, analyze and report the environmental information that can be extracted from these new studies and those parts of the completed research not adequately reported.

A Study of Seafood Consumption and \( ^{65} \text{Zn} \) Body Burden

T. H. Essig, G. W. R. Endres and J. F. Honstead*

One of the radionuclides formed by neutron activation in Hanford production reactors and released to the Columbia River in effluent cooling water prior to February, 1971 was Zn-65. This radionuclide has a relatively long radioactive half-life (245 days) and is an isotope of an element utilized in metabolic processes by most shellfish and other aquatic life forms. Consequently, Zn-65 is found in fish and shellfish in the Columbia River and in the Pacific Ocean near the mouth of the River. The transfer of \( ^{65} \text{Zn} \) through various food chains to man has been studied at Hanford for a number of years.\(^{(1,2,3,4)}\)

Since the half-life of \( ^{65} \text{Zn} \) is fairly long, it can enter into rather complex transport processes in a variety of food chains and can persist in measurable quantities in the environment—particularly in organisms that concentrate zinc by metabolic absorption. Shellfish such as oysters, clams and crabs concentrate zinc (and consequently \( ^{65} \text{Zn} \)) to a high degree and thus represent the dietary items of primary interest in this study.

When \( ^{65} \text{Zn} \) decays, a 1.11 MeV photon is emitted. The energy of this photon is high enough to be readily detected by a scintillation crystal external to the object being measured, e.g., environmental samples or the human body. Thus, scintillation counter techniques (such as those employed in a mobile whole-body counter), together with knowledge of an individual's dietary habits, can be used to study the environmental behavior of \( ^{65} \text{Zn} \), particularly with those individuals consuming large quantities of shellfish from the Columbia River estuary.

In this study, consumption rates of four seafoods (clam, crab, shrimp and oysters) were obtained for 250 residents of Rockaway, Oregon, and Ilwaco, Washington, by personal interviews. These consumption rates were used in conjunction with \( ^{65} \text{Zn} \) concentrations in the seafoods of interest.

* Deceased
to estimate the $^{65}Zn$ body burden for each individual. Rather sophisticated equations were used to adjust the seafood intake as a function of age for the three years preceding this study. This adjustment was necessary because of the relatively long half-life of $^{65}Zn$. The following equation (adapted from reference 5) was used to calculate the $^{65}Zn$ body burden ($q$):

$$q = \frac{f_w I e}{0.693} \left[ 1 - \exp (-0.693t/T) \right]$$

where

$f_w$ = fractional uptake of $^{65}Zn$ for each type of seafood,
$I$ = intake rate of $^{65}Zn$, nCi/day,
$T_e$ = effective half-life = 194 days
and
$t$ = period over which $^{65}Zn$ was ingested, days.

After each subject was interviewed, he was counted in a mobile whole-body counter located in the vicinity of each of the seacoast communities. The $^{65}Zn$ body burdens measured by this technique were corrected for background and Compton scattering, and were then compared with the body burdens calculated by Equation 1. The results of this comparison for one group of people (seafood company workers) are shown in the figure.

The agreement between the measured and calculated $^{65}Zn$ body burdens for the seafood company workers is considered to be very good. The average measured-to-calculated ratio of body burdens was 1.47, with a range in individual ratios from 0.11 to 3.7. This range of individual ratios is felt to reflect a combination of natural variability of key metabolic parameters and of the ability of an individual to quantitatively estimate his diet.

REFERENCES CITED


...
Seafood Workers

AVE $^{65}$Zn MEASURED = 3.04 nCi
AVE $^{65}$Zn COMPUTED = 2.85 nCi
AVE $^{137}$Cs MEASURED = 4.19 nCi
RADIOLOGICAL EVALUATION OF POST MORTEM TISSUE SAMPLES
I. C. Nelson, K. R. Heid,* P. A. Fuqua, MD** and T. D. Mahony, MD***

This study is concerned with the collection, radioanalytic and evaluation of tissue samples obtained post-mortem from plant workers and residents in the environs of the Hanford Project. The significance of this effort lies primarily in developing the capability for assessing the overall long-term effectiveness of engineering safeguards which govern the release of radioactive materials to the Hanford environs. The data are similarly important for assessing the overall impact on occupational workers through correlation of exposure conditions and resulting body burdens.

Results of plutonium analyses on tissue samples obtained prior to late 1970 were assembled, analyzed and reported. Collection of lung, liver, bone and tracheobronchial lymph node samples continued throughout the year. In November, 1971 samples of spleen and gonads were added to the list of desired samples. The presence of plutonium in the spleen has been suggested from animal studies following intravenous or intramuscular injection of plutonium compounds. Positive findings of plutonium in the spleens of workers may establish the deposition of plutonium as resulting from contaminated wounds. The presence or absence of plutonium in gonadal tissue may be of interest from a genetic point of view.

An expansion of the sampling program to include individuals remote from the Hanford complex and to include analysis for certain selected stable nuclides was discussed at a meeting with representatives of the Los Alamos Scientific Laboratory. Tentatively, Pacific Northwest Laboratory will consider expansion of sampling programs to selected localities west of the Continental Divide whereas the Los Alamos group will emphasize sampling from localities east of the Divide.

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** Hanford Environmental Health Foundation, Inc.
*** Kadlec Hospital, Richland, WA
RADIATION DOSIMETRY

These studies are primarily concerned with the stochastic nature of absorbed dose. The objective is to determine those features of energy deposition patterns that are relevant to the understanding of observed biological effects. Theoretical effort has been in development of mathematical models and computational techniques to couple the spatial distribution of energy deposition with temporal distributions. Emphasis has been on understanding time dependent effects and dose rate dependence of the oxygen effect. Experimental work has been in the application of cross section measurements to the problem of energy transport by \( \gamma \)-rays and to a more detailed description of proton stopping powers.

Energy Transport by Secondary
Electrons
W. A. Glass

In order to understand the nature of energy deposition in small volumes traversed by protons, it is essential to know the probabilities involved for energy transfer in individual collisions between the proton and the absorber molecules. An analysis of the relative importance of the various processes by which the energy of fast protons is transferred to the absorber shows that about 50 to 60 percent appears as kinetic energy of secondary electrons, 25 to 30 percent is involved in overcoming the binding energy of these electrons, and the remaining 10 to 20 percent appears as primary excitation, recoil energy, etc.\(^{(1)}\)

It has been observed that distributions of energy deposited in small volumes by protons are greatly affected by energy transport associated with energetic secondary electrons. The moments of these distributions do not conform to those expected in the absence of energy transport.\(^{(2)}\) Quotients of the variance by the mean of ionization distributions measured in small tissue-equivalent sites were found to be nearly independent of proton energy.

The general distribution of those electrons ejected in primary collisions can be put in perspective by considering the number of electrons and the kinetic energy associated with them above certain selected secondary electron energies. The table summarizes data for two representative low Z gases bombarded by protons between 0.3 and 1.5 MeV. The mean ejected electron energy lies between 30 and 60 eV and this value is only slightly dependent on whether the target is hydrogen or nitrogen. Nearly a constant 75 percent of the kinetic energy is associated with the 20 to 25 percent of electrons with energy greater than the mean value. Half the ejected electrons have energies below 10 to 15 eV - i.e. over
### Summary of Data for Two Representative Low Z Gases Bombarded by Protons Between 0.3 and 1.5 MeV

<table>
<thead>
<tr>
<th>Target Gas</th>
<th>Proton Energy, 1p, keV</th>
<th>Mean Electron Energy $\langle E \rangle$</th>
<th>$\langle E \rangle$ Mean Energy $\langle E \rangle$ Median</th>
<th>Number Median $\langle E \rangle$ range $&lt;10^6$ g/cm²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrogen</td>
<td>300</td>
<td>0.23, 0.75, 0.044, 0.37, 12.5, 0.91</td>
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<td></td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>0.21, 0.76, 0.030, 0.36, 12.5, 0.92</td>
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<tr>
<td></td>
<td>1000</td>
<td>0.19, 0.76, 0.017, 0.34, 13.0, 0.92</td>
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<tr>
<td></td>
<td>1500</td>
<td>0.17, 0.76, 0.012, 0.31, 13.0, 0.88</td>
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<tr>
<td>Nitrogen</td>
<td>300</td>
<td>0.26, 0.88, 0.062, 0.38, 18.0, 0.92</td>
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<tr>
<td></td>
<td>1000</td>
<td>0.19, 0.77, 0.026, 0.31, 15.0, 0.93</td>
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<td></td>
</tr>
<tr>
<td></td>
<td>1400</td>
<td>0.19, 0.77, 0.016, 0.30, 15.0, 0.94</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Target Gas | Proton Energy, 1p, keV | Energy Median $\langle E \rangle$ Median | Locally deposited $\langle E \rangle$ range $<10^6$ g/cm² |
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrogen</td>
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<td>100, 0.060, 0.032, 0.31</td>
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</tr>
<tr>
<td></td>
<td>500</td>
<td>136, 0.062, 0.040, 0.40</td>
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<td>1000</td>
<td>187, 0.044, 0.043, 0.46</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1500</td>
<td>225, 0.040, 0.047, 0.53</td>
<td></td>
</tr>
<tr>
<td>Nitrogen</td>
<td>300</td>
<td>117, 0.10, 0.046, 0.31</td>
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<tr>
<td></td>
<td>1000</td>
<td>232, 0.054, 0.053, 0.54</td>
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</tr>
<tr>
<td></td>
<td>1400</td>
<td>250, 0.048, 0.050, 0.55</td>
<td></td>
</tr>
</tbody>
</table>
half of the ejected electrons are incapable of further ionization.

On the other hand, 90 to 95 percent of the kinetic energy is associated with that half of the ejected electrons having energy in excess of 10 to 15 eV. Half the kinetic energy transferred to secondary electrons is associated with fewer than 10 percent of the electrons and this percentage rapidly decreases with increasing proton energy. Electrons ejected with sufficient energy to travel distances greater than $10^{-6}$ g/cm$^2$ away from the proton path (i.e. kinetic energy greater than 200 eV) number only 3 to 6 percent of the total, but they transport between 30 and 60 percent of the available kinetic energy. It is these few energetic secondaries that are responsible for the large difference between the observed and calculated relative variance in the energy deposition in small volumes.

**Hit Theory for Low-LET Radiation**

W. C. Roesch

The time-dependent hit theory\textsuperscript{(3)} discussed before\textsuperscript{(4)} proved difficult to evaluate numerically in the cases of low-LET radiation such as x or gamma rays because of the small size of some of the parameters (or, equivalently, because of the large number of hits required to produce death). Two approximate expressions for the survival have now been derived for low-LET radiation. Both can be shown to be quite accurate approximations.

One approximation is made by assuming that the hits are uniformly spaced in time rather than randomly spaced. The other is made by averaging over the random distribution in time by pairs of hits rather than averaging all of them at once. It is possible to show that the first approximation is larger than the true survival and the second, smaller. Numerical results confirm that the first approximation is always larger than the second; however, they also show that the two do not differ significantly in the final results they give for the survival. The figure shows survival curves calculated for several different dose rates ($c/b$) from the two approximations open and closed circles; the two sets of points essentially lie on common curves. Since the two approximations bracket the true value and since they are indistinguishable, either one can be taken as the true survival for practical purposes. The two solid lines in the figure are limiting curves for very low and very high dose rates; both of these can be derived without using the approximations.

The expression for the survival, $S$, for an irradiation of length $t$ given by the second approximation is

$$S = N(1 - e^{-bt}) e^{-D/D_0} \quad (1)$$

where $b$ is a constant and $N$ and $D_0$ are what are commonly called the extrapolation number and the mean lethal dose; $D$ is the absorbed dose. The theory yields expressions for $N$ and $D_0$ in terms of more fundamental parameters.

Haynes\textsuperscript{(5)} and Kellerer\textsuperscript{(6)} have
given heuristic derivations of equations of the same form as Equation (1), and these have already been found useful in radiobiology.

The same approximate treatment yields

\[
S = N \left(1 - e^{-b_1 t_1}\right) \left(1 - e^{-b_2 t_2}\right) \frac{1 - e^{-b u}}{1 - e^{-b u^*}} \quad (2)
\]

for the survival to irradiation in two periods, \(t_1\) and \(t_2\), separated by a time \(u\). \(S^*\) is the survival calculated from Equation (1) for irradiation for a single period \(t_1 + t_2\). This expression predicts correctly the basic properties found in fractionation experiments.\(^7\)
Theory of the Oxygen Effect
W. C. Roesch

Alper and Howard-Flanders (8) summarized the data on the oxygen effect at low dose rates with the observation that the $D_0$ at an oxygen concentration $M$ was related to that, $D_{0N}$, for $M = 0$ (i.e. in nitrogen) by

$$D_{0N}/D_0 = (KM + k)/(M + k)$$ (1)

where $K$ and $k$ are constants.

The hit theory reported above can explain these low-rate data as follows: An individual hit product can interact with the medium through reactions that either involve oxygen or through some that do not involve oxygen. Let the probability of the former be $(C/k)M/(CM/k + C)$ and the latter, $C/(CM/k + C)$, where $C$ is a constant. $(C$ and $C/k$ play the role of independent constants, but they have been chosen to lead to the result of Equation (1). Similarly for $s_N$ and $ks_N$ in the next sentence.) Let the probability of a reaction with oxygen leading to death be $ks_N$, and one without oxygen be $s_N$. Then the probability of death is

$$(C/k)MKs_N/(CM/k + C) + Cs_N/(CM/k + C)$$

$$= s_N (KM + k)/(M + k)$$ (2)

It can be shown that this expression leads to the Alper and Howard-Flanders law.

Dewey and Boag (9) found that at very high dose rates the survival for low concentrations of oxygen was nearly the same as that for no oxygen. The points in the figure, taken from Epp et al. (10) show the phenomenon in more detail. In general, the survival at low doses is smaller than in nitrogen, but the survival curve soon turns and runs parallel to the curve for nitrogen. Dewey and Boag explained the high-rate effect by assuming that the early part of the irradiation pulse consumed the available supply of oxygen; then, during the remainder of the pulse, the population was being irradiated in anoxia. The pulses are so short that the re-supply of oxygen during the pulse is negligible.

The Dewey-Boag hypothesis can be coupled with the theory above to give a quantitative explanation of the high-rate effect. It is assumed that the chance of death from an individual hit is given by Equation (2) and that $M$ decreases from one hit to the next. The outcome then depends on the rate of decrease of $M$. Assuming that the rate of decrease of $M$ is $AM/(M + B)$, where $A$ and $B$ are constants chosen to give the best fit to the data, leads to the solid-line curves in the figure. The chosen parameters give a good fit to the data for the higher oxygen concentrations. Further study of the consumption process is planned.
Comparison of Theory and Experiment for the Oxygen Effect
Stopping Power Partition and Mean Energy Loss for Energetic Protons in Hydrogen

W. E. Wilson

The total mass stopping power, \( S(E_0) \), for a particle of energy \( E_0 \) can be related to the various modes for energy loss by the expression

\[
S(E_0) = \frac{n}{o} \left[ \frac{E_x}{E_x} \sigma_x \right] \left[ \frac{I_{\text{max}}}{I_{\text{max}} + I_{n}} \right] \sigma_n(T) \left( T_{n} + T_{d} \right) . \tag{1}
\]

where \( n \) and \( o \) are respectively the number and mass density of the absorber; \( \sigma_x \) is the cross section for excitation of the absorber molecules to state \( x \), with excitation energy \( E_x \); \( \sigma_n(T) \) is the ionization cross section for ejecting a secondary electron from shell \( n \) with energy between \( T \) and \( T + dT \); \( I_{n} \) is the binding energy of electrons in the \( n \)th shell of the absorber atoms.

For incident electrons \( T_{\text{max}} \) is \( E_{0} - I_{n}/2 \), the kinematic maximum energy that an electron of energy \( E_{0} \) can transfer to an identical particle bound with energy \( I_{n} \). For incident protons (or any heavy particle) \( T_{\text{max}} \) is approximately \( 4E_{0} m_{e}/m_{p} \); \( \sigma_n(T) \) is observed to decrease rapidly for \( T \) greater than this value. This limit is the classical maximum energy transfer that a particle of energy \( E_{0} \) and mass \( m_{p} \) can give to a free particle of mass \( m_{e} \).

The three terms of Equation 1 are "partial stopping powers" and are assigned the symbols

\[
S_{E} = \frac{n}{o} \left[ \frac{E_x}{E_x} \sigma_x \right] ; \tag{2A}
\]

\[
S_{I} = \frac{n}{o} \int_{0}^{T_{\text{max}}} \sigma_{n}(T) dT = \frac{n}{o} \int_{0}^{T_{\text{max}}} \sigma_{n}(T) dT ; \tag{2B}
\]

\[
S_{T} = \frac{n}{o} \int_{0}^{T_{\text{max}}} \sigma_{n}(T) T dT = \frac{n}{o} \int_{0}^{T_{\text{max}}} \sigma_{n}(T) T dT ; \tag{2C}
\]

\( \sigma_n \) is the \( n \)th shell total ionization cross section, and \( \sigma(T) \) the cross section for ejection of electrons with energy between \( T \) and \( T + dT \) while leaving the residual ion in any state.

Recently measured cross sections for total ionization and for electron ejection have been employed to obtain the partial stopping powers defined by Equations 2A, B and C for the case of energetic protons incident upon hydrogen gas. The results indicate that the partial stopping power, \( S_{T} \), which is the transfer of proton kinetic energy into kinetic energy of first generation secondary electrons, constitutes about 60% of the total stopping power from 0.2 to 1.5 MeV and is nearly independent of primary proton energy. \( S_{I} \) makes up about 30% of the total stopping power at 0.2 MeV and decreases to about 20% at 1.5 MeV. Experimental data are not available for the \( \sigma_{x} \), but an estimate of \( S_{E} \) from theoretical calculations indicates that \( S_{E} \) can reasonably account for the remaining part of the total stopping power.

Energy-restricted linear collision stopping power \( (L_{0}) \) is related to the fundamental cross sections by the relation
\[ L(\Delta) = \frac{\Delta}{\rho} \left[ \sum E_x \sigma_x + \sum I_n \sigma_n \right] + \int_0^\Delta \sigma(T) dT \right] \]  

for \( E_x < \Delta \) and \( I_n < \Delta \) and where \( \Delta \) is defined to be the energy cutoff for each energy loss channel rather than the limit on the total energy transfer in the ionizing collision.

The ICRU has defined restricted linear collision stopping power to be the quotient of \( dE \) by \( dl \) where \( dE \) is the mean energy loss in \( dl \) due to collisions with energy transfers less than some specified value \( \Delta \). Placing the restriction on the sum of \( E_x, I_n \) and \( T \) couples the Kinetic Energy of the ionization electron with the final state of the ion and the development which leads to Equation 3 cannot proceed. The definition used here leads to calculable quantities without the need for implicit assumptions or approximations.

To the extent that \( E_x \) and \( I_n \) are "locally absorbed energies" we can write (3) as

\[ L(\Delta) = S_E \ast S_1 + \int_0^\Delta \sigma(T) dT \] or (4)

\[ L(\Delta) = S \ast S_T(\Delta) \] , where

\[ S_T(\Delta) = \int_0^{T_{max}} \ast \sigma(T) dT. \] (6)

Equation 6 is a generalization of (2C). Equation 5, along with curves or tables of values for \( S_T(\Delta) \) defined by Equation 6, can be used to obtain restricted stopping powers for arbitrary cutoffs \( \Delta \). Curves of \( S_T(\Delta) \) for energetic protons stopping in hydrogen were evaluated and are to be published in Radiation Research in February.

REFERENCES CITED


This research effort is primarily concerned with basic mechanisms through which radiation interacts with matter. Emphasis is on biologically relevant materials. The studies involve the initial interactions by which ionizing particles transfer energy to matter and the subsequent transport and degradation of that energy. The work includes experimental measurement of the doubly differential cross sections for electron ejection by mono-energetic protons, investigation of the partition and distribution of energy in the radiation field and in the irradiated medium and determination of the rates and mechanism for energy transfer in the early stages of molecular rearrangement.

Electron Emission Cross Sections

L. H. Toburen

The investigation of electron emission cross sections for a number of gases has been carried out during the past year. Particular emphasis was placed on cross sections measured for 0.3 to 1.5 MeV protons on hydrogen and for 1.0 and 2.0 MeV protons on helium. These gases are especially interesting as they are the only atoms for which theoretical calculations exist in which electron emission cross sections differential in both electron energy and emission angle have been calculated. Comparison of measured and calculated cross sections yield information regarding the details of the emission process such as the effects of long range electron-proton interactions and the effects of the distortion of the target atom by the impulsive interaction of the passing proton. In a more general sense, the range of validity of the Born approximation is tested by these results. It was found, for example, that for proton energies as high as 1.5 MeV the Born approximation was not adequate to provide accurate double-differential cross sections for electron emission into small angles (less than 40 degrees). Calculations by Macek based on Faddeev's equations, although only qualitatively correct for low energy proton impact, were found in excellent agreement with our measurements for 1.5 MeV protons on hydrogen. This evidence of agreement was not repeated, however, when Macek's results for helium were compared with our measurements at 1.0 MeV. This result indicates that the interactions and/or wave functions included in his calculations do not properly reflect changes in the target atom. It appears that a good deal of theoretical work will be necessary before one can accurately calculate double-differential electron emission cross sections for even the most simple atoms.

In addition to the detailed study of hydrogen and helium, considerable effort has been directed to more complex atoms and molecules. Electron
emission cross sections have been measured for a series of hydrocarbons including methane, ethane, ethylene, acetylene, and benzene in order to investigate the effects of molecular binding.

We are still in the process of analyzing results obtained from $H_2^+$ impact on molecular hydrogen. These data are quite complicated since we must deal with electrons ejected from the target atom by both incoming simultaneous protons and the attached electron. The incident electrons display both elastic and inelastic scattering from the target molecule.

**Inner Shell Ionization and Fluorescent Yields**

L. H. Toburen

Auger electron peaks are observed superimposed on the continuum electron emission from hydrocarbon targets under proton bombardment. These Auger electrons result from filling vacancies in the K-shell of carbon which are produced by proton impact. Since the fluorescent yield for carbon is small ($\omega_K = 0.002$) the Auger electron yield can be directly correlated with the number of inner shell vacancies produced. When different types of carbon-containing molecules were investigated, the shape and intensity of the Auger electron spectra were found to change. These changes in the shape of the Auger spectra indicate that different initial and final states are being excited for a given proton energy depending on the molecular surroundings of the carbon atom. For 1.0 MeV the total cross section for K-Auger electron emission from carbon was found to vary by 10% as the carbon-carbon bonding progressed from single to triple bonds, e.g., in going from ethane to acetylene. A variation of only 6% was found for similar measurements with 0.3 MeV incident protons. Our present electron energy resolution (3.5%) is not sufficient for detailed analysis; however, if better resolution can be obtained, information regarding final charge states of the residual target particles can be obtained, both as a function of incident particle energy and as a function of the molecular bonding.

The K-shell ionization cross sections obtained in our work are the first measured values which do not rely on an accurate knowledge of the fluorescent yield; this is significant since reported values of the fluorescent yield of carbon vary by an order of magnitude. Our results are in excellent agreement with semiclassical calculations reported by Garcia;\(^4\) calculations based on the Born approximation\(^5\) result in cross sections nearly 30% smaller.

In principle, the fluorescent yield of carbon can be obtained from our Auger electron yields and published X-ray yields.\(^5\) The fluorescent yields determined in this manner are found to be a slowly varying function of the proton energy, e.g., $\omega_K = 0.0016$ for 0.3 MeV protons and $\omega_K = 0.0025$ for 2.0 MeV proton impact. This variation of fluorescent yield with particle energy can be qualitatively explained as due to the production of multiply charged target
particles. There is presently insufficient information, however, to explain this phenomenon quantitatively. These results point out the need to redefine what is meant by the fluorescent yield to include the dependence on the final state of the residual target ion. The fluorescence efficiency appears to depend not only on the final charge state but also on the final molecular state in which the atom containing the initial inner shell ionization resides.

Our preliminary measurements for protons on xenon indicate that M- and N-shell ionization cross sections for heavy atoms may be obtained from electron emission measurements. There is, however, still some question regarding the reliability of our measurements for xenon and any conclusions await experimental work which is in progress at the present time.

Fluorescent Decay of Organic Liquids

M. L. West

Investigation of the fluorescent decay of organic liquids excited by ionizing and non-ionizing radiation has continued throughout the year. Fluorescing molecules produced under different modes of excitation serve as sensitive probes through their extreme sensitivity to the surrounding environment and allow us to explore the temporal stages of radiation action leading up to permanent molecular alterations. The non-exponential fluorescent decay previously observed in solutions of benzene in cyclohexane under proton irradiation has suggested a diffusional quenching model whereby excited molecules are created along a localized proton track interact as they diffuse from their initial reaction site. Diffusion kinetic theory as first formulated by Samuel and Magee (6) to explain the effect of linear energy transfer on radical and molecular yields in water has been developed and programmed for computer manipulation. This is a convenient starting point in our effort to ultimately treat stochastically the effects of ionizing radiation in liquid solution. Since we are interested in the time behavior of molecular fluorescence as influenced by inhomogeneous radical distributions along a track, the following expression was derived:

$$\frac{d}{dt} \ln I(t) = -k_{uv} \cdot C \cdot R(t) \quad (1)$$

where $I(t)$ is fluorescence intensity, $R(t)$ is radical concentration, $k_{uv}$ is a rate constant from u.v. excitation, and $k_r$ is the rate of radical-excited state interaction. Other model parameters include initial radical and excited state densities and radical-radical reaction rates. From the radiation chemist's point of view, $R(t)$ is negligible after $10^{-9}$ in terms of radical-radical interaction or radical scavenging and its effect on final yields. But for fluorescence decay, estimates based on approximate values of model parameters show that the last term of Equation (1) is still comparable to $k_{uv}$ in the time interval $10^{-9} - 10^{-8}$ sec. Attempts are being made to fit this equation to experimental decay curves. Using approximate values of model parameters fixed by independent experiments in radiation chemistry we show that the
observed decay is consistent with this quenching model. Model calculations are presently being extended to include a/8 ratios as an additional check on its success to explain radiation effects. Additional tests will include experiments on selected aqueous solutions where product yields and rate parameters are accurately known.

Electron-Ejection Cross Sections for Several Representative Hydrocarbon Molecules

W. E. Wilson

Cross sections, differential in ejection energy and angle, for electron emission from methane, ethane, ethylene, acetylene and benzene molecules following proton impact have been measured for protons of 0.3 and 1.0 MeV. The purpose in making the measurements is to look for an effect on the electron yield attributable to the chemical form or molecular binding of the target atoms. If such an effect is significant, it has implications for the additivity rules for ionization and energy loss in gaseous mixtures. For example, the ionization one observed in a "tissue equivalent" gas might then depend on whether methane or acetylene were used for the carbon component.

In general, the results indicate that for many practical situations involving energy transport by energetic secondaries there is no significant binding effect. The distributions differential in energy only, obtained by integrating over emission angle, have proportional zero, first, and second moments, so that ratios of the moments are constants for the molecules studied. For all five target molecules and for both proton energies, first generation secondary electrons (FSE) with energies greater than the mean energy, account for 74% of the total energy loss associated with kinetic energy of all secondaries. For both proton energies the distributions, differential in energy only, are scalable in terms of the number of outer or weakly bound electrons in the molecule. If the yield or cross section in units of \( cm^2 ev^{-1} molecule^{-1} \) is divided by the total number of outer electrons in the molecule (total electrons minus the carbon K-shell electrons) the result is a "universal curve" in units of \( cm^2 ev^{-1} electron^{-1} \). The yield 'per electron' is nearly identical for all 5 molecules for secondaries with energies greater than 10 eV. Below 10 eV the data are too sparse to provide a good comparison.

This scalability is equivalent to saying the effective yield per carbon atom is the same for all 5 molecules studied. This does not say that chemical environment or molecular binding in general makes little difference in the yield of secondary electrons. We can demonstrate a definite molecular binding effect and at the same time shed some light on the validity of the Bragg-additivity rule for stopping power by using previously measured molecular hydrogen electron-emission cross sections, properly scaled, to subtract the contribution of the hydrogen from the hydrocarbon yields. The result is an equivalent carbon atom yield and the process is
the reverse of the additivity rule. When one constructs a secondary-electron energy distribution for carbon by taking 1/2 the difference between the acetylene yield and the molecular hydrogen yield 
\[ \frac{1}{2} (C_2H_2 - H_2) \], one obtains a result different from that obtained by subtracting from the methane distribution 2 times the hydrogen distribution, \( [CH_4 - 2H_2] \). The difference shows up as a larger yield of secondary electrons for energies up to about 60 eV for the distribution synthesized from the methane-hydrogen data; the yield of 10 eV secondaries is 30% larger for the carbon atom from methane than from acetylene by this method of analysis. Distributions synthesized in a similar manner from ethane-hydrogen and ethylene-hydrogen agree more closely with the distribution obtained from methane-hydrogen whereas the distribution obtained from benzene-hydrogen falls with the distribution for the acetylene-hydrogen, i.e., the saturated hydrocarbons produce similar secondary electron-energy distributions for carbon atoms and the unsaturated hydrocarbons indicate a different distribution.

The important conclusion we reach is that use of the Bragg-additivity rule can lead to error in predicting the number of ion-electron pairs with secondary electron Kinetic Energy as large as 60 eV.

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SOME EXPERIMENTAL MEASUREMENTS OF AIRBORNE URANIUM (REPRESENTING PLUTONIUM) IN TRANSPORTATION ACCIDENTS

J. Mishima
L.C. Schwendiman

Atmospheric Analysis Section
ATMOSPHERIC SCIENCES DEPARTMENT

AUGUST, 1973

BATTelle
PACIFIC NORTHWEST LABORATORIES
Richland, Washington
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SOME EXPERIMENTAL MEASUREMENTS OF AIRBORNE URANIUM 
(REPRESENTING PLUTONIUM) IN TRANSPORTATION ACCIDENTS

ABSTRACT

Experiments were performed to measure the fractional release of uranium compounds when these materials are involved in gasoline fires. Uranium was used as a reasonable stand-in for plutonium. In the experiments reported uranium dioxide particles or uranium nitrate in solution was deposited on various materials ranging from a smooth metal surface to soil. Gasoline was added to these materials, then ignited. Airborne particles were collected at points downstream in the duct in which the combustion tray was located. Various air flow and burning conditions were used. In the burning experiments in which a uranyl nitrate solution was deposited on a stainless steel plate, 11% was made airborne with an air flow of 23 mph. This release was the largest for the burning experiments; however, as much as 26% of uranium dioxide powder was aerodynamically entrained from dry, sandy soil by air at a velocity of 20 mph. No single variable of the conditions imposed in 40 experiments could be identified as having an over-riding influence on the releases of uranium within the range of variables included in these experiments. A gasoline fire in which 500 gallons was dumped and burned, affirmed that the smaller scaled experiments were reasonably representative of actual fires.

INTRODUCTION

Because of the low maximum permissible concentration of plutonium in air, great care is exercised in every operation involving this material. Extreme precautions are taken to protect workers and people in the vicinity of plutonium facilities, yet experience has shown there can be low probability incidents which may give rise to airborne release of plutonium and its compounds. In order to assess the seriousness of inadvertent incidents in which control may be momentarily lost during some planned operation, those responsible for safety of the operation must have reasonable estimates of the amount and nature of the plutonium which could be made airborne under these circumstances. Only meager information has been available to assess the fractional release of plutonium in various accident situations. Battelle-Northwest has performed studies in the past to furnish data for estimating the potential airborne hazard.
from plutonium in specific situations which are of concern in fuel reprocessing, plutonium recovery, and related operations. Situations with significant potential for the airborne release of plutonium have been identified. In these earlier experiments relatively small specimens of plutonium and its compounds were subjected to heat stress and airflow regimes and the fraction of plutonium airborne measured. The results were regarded as indicative rather than definitive because of the many variables which could operate to change the release fraction. These experiments did reveal the potential for airborne release under several circumstances. The desirability of carrying out similar experiments on a more realistic scale was early acknowledged; however, the difficulties, expense and risks posed in using plutonium in large-scale experiments required that a simulant be used. A study of several possible materials for a suitable stand-in for plutonium showed that uranium could be used for this role. In the work to be described, uranium was used in experiments on a scale such that reasonable simulation of gasoline fires engulfing uranium oxides and nitrate spilled on various surfaces could be achieved. A tacit assumption was made that plutonium compounds would behave similarly.

Hypothetical, serious situations occurring during the transport of plutonium within the Hanford site boundaries were used as reference cases for the experimental program. Following is a brief description of the most serious potential transporting accident considered and some observations on the availability of data to permit consequences to be assessed.

A vehicle carrying up to 20 kg of plutonium as a nitrate solution, or 2 kg of plutonium as a dry dioxide powder collides with a gasoline truck. The contents of both vehicles spill onto the ground and intermix. The gasoline ignites and burns.

In the situation described various factors may influence the fraction of plutonium that might be made airborne.

- The physical characteristics of the surface involved — porous or impervious, plastic or rigid, rough or smooth — and interaction between the surface and the materials spilled.
- Windspeed and direction.
- Physical and chemical nature of the plutonium compound.
- Sequence of events during accident — local conditions and mechanisms.
operative at time and site of release.

- Presence of other materials -- i.e., if vegetation is present, the active material could be distributed in the vegetation already in the air stream.
- Rate of burning may produce thermal plumes and cause near-ground turbulence.

Some data were available which could be used to evaluate the airborne release of plutonium in such an accident. Data available were the following:

**Nitrate Solutions**

Some data existed on fractional airborne release from nitrate solutions during slow drying from a hard impervious surface\(^3\); from the solid residues after evaporation at high temperatures\(^3\); from the dried residues on sand\(^4\); and during the burning of flammables impregnated with a nitrate solution.\(^4\)

The fraction airborne during the evaporation (less than 130°C) of nitrate solution from a hard impervious surface (stainless steel) in air with a velocity of up to 100 cm/sec was low -- maximum value detected under experimental conditions imposed was 0.003 wt. percent. Even less of the dried residue was made airborne under these conditions -- maximum quantity measured was \(3 \times 10^{-6}\) wt. percent. The quantity of plutonium airborne during the heating of a nitrate solution to near dryness indicated dependency on heat input with a maximum value under experimental conditions imposed of 0.18 wt. percent. An additional quantity, as much as 1.7 wt. percent was deposited as "fallout" close to the vessel, presumably from droplets too large to be carried away. As much as 0.018 wt. percent of partially air dried plutonium nitrate held in sand was made airborne when heated to high temperatures (probably in excess of 900°C) in an upsweep of air at a nominal velocity of 100 cm/sec. When incorporated with flammable materials producing a fine ash, as much as 8 wt. percent of a uranium nitrate solution could be made airborne in air at a velocity of 100 cm/sec.

**Dry Dioxide Powder**

Experimental data were available on the fraction airborne during the heating of dry dioxide powder on an impervious surface and during the burning of flammables in which dry dioxide had been incorporated. Limited data on entrainment from various surfaces by air in the absence of heat were available. Only
a moderate amount (maximum value of 0.025 wt. percent) of a dry dioxide powder ranging from 15 to 150 microns aerodynamic equivalent (AED) was made airborne from a hard, impervious surface (stainless steel) during heating. A considerably greater quantity was airborne (up to 50 wt. percent) during the burning of flammable materials incorporating dry uranium dioxide powder in air with a velocity of 100 cm/sec. A considerable amount of active material can be entrained by impinging high velocity air from the proper angle upon the surface.

Transporting radioactive materials intuitively appears to be an operation possessing a great potential for release. The carrier in motion possesses great energy and is capable of inflicting great damage upon impact, and shipping containers, though having high integrity, can be caused to fail under extreme stress.

With the advent of wide-scale nuclear power reactors and the attendant need for transportation, reprocessing, and other handling of very large quantities of plutonium, it is important that the consequences of failure in these operations be known.

**OBJECTIVE**

The objective of this study was to provide data on fractional airborne release and aerosol properties to permit more realistic evaluation of the potential airborne hazard from plutonium in accidents during the transport of plutonium.

**SUMMARY**

The amount of aerodynamic equivalent size distribution of particles that may become airborne under conditions found in some shipping accidents were studied in a special wind tunnel at the 242-B facility. Airborne release for uranium dioxide powder or nitrate liquid, representing plutonium, dispersed on four types of surfaces were evaluated in forty combinations of conditions believed to be representative of circumstances which could prevail in a transportation accident. A wind speed of 2.5 mph and a higher wind speed of 20-30 mph were used, both with and without gasoline fire (see Appendix A). Procedures and equipment used are described. A large scale gasoline fire test in which 500 gallons of gasoline was rapidly released to the soil and ignited
affirmed that the conditions observed in the engineering scale experiments were a reasonable simulation of the postulated transportation-type accidents. The results of the fractional release indicate:

- No single parameter of those evaluated — physical form of the active material, type of surface, wind speed nor the presence of a gasoline fire — exerts a controlling influence on the fractional airborne release under all circumstances.

- For the combination of variables used in these experiments, the greatest fractional release occurred from aerodynamic entrainment of uranium dioxide powder deposited on sandy soil from which 24 percent was airborne in air at 20 mph. For gasoline fires the largest fraction airborne, 11 percent, was from uranyl nitrate solution on a smooth metal plate, when air was passed through the duct at 23 mph.

**EXPERIMENTAL**

**ENGINEERING SCALE EXPERIMENTS**

Accidents are unique occurrences, and the actual sequence of events can have many variations. Performing a parametric study of all factors would encompass a very large number of experiments. In order to embrace the broadest coverage of situations within a tenable number of experiments, we performed experiments representing forty combinations of parameters (uranium form, wind speed, surfaces and the presence or absence of a gasoline fire) that could attend a serious shipping accident with container breaching. The combinations of parameters involved two physically different forms of the active material deposited on four commonly available surfaces. The amount and size distribution of the particles airborne at two wind speeds with and without a petroleum fire were measured. The individual experiments are tabulated in Table I.

**Description of Experimental Apparatus and Procedures**

All experiments were performed in the furnace section of the inlet duct to the Radioactive Aerosol Release Tank (R.A.R.T.) in the 242-B Building facility[5] (see Figure 1). Uranium was used as a simulant for plutonium in these engineering scale experiments. [5]

The furnace section is located on a concrete pad on the west side of the
TABLE 1

EXPERIMENTAL PROGRAM - AIRBORNE RELEASE UNDER SELECTED CONDITION SIMULATED
FOR SHIPPI NG ACCIDENTS

SA-1 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 23 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-2 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-3 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-10 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 23 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-19 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 23 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-19a Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 23 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-20 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-21 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-22 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-23 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-24 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-25 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-26 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-27 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-28 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-29 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-30 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-31 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-32 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-33 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-34 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-35 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-36 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-37 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-38 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-39 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-40 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-41 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-42 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-43 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-44 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-45 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-46 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-47 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-48 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-49 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-50 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-51 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-52 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-53 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-54 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-55 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.

SA-56 Fractional release of uranium immediately following
a petroleum fire into air passing over the surface
at a speed of 3 mph, uranium nitrate solution
deposited on a stainless steel surface.
Figure 1. Radioactive Aerosol Release Facility, 242-B Building
242-B Building and is a part of the inlet duct to the R.A.R.T. (see Figure 2). Fresh air is drawn through a bank of four 24-inch by 24-inch by 12-inch deep filters by a centrifugal blower at the downstream end, with a rated flow of 400 cfm at 6-inch W.G. static pressure. A flash arrestor separates the filters from the 3-foot long furnace section which is lined with a refractory material. Three half-couplings (3/4, 2 and 1/4 inch) welded over holes drilled in the north wall of the furnace section provided access to the interior. A 24-inch diameter by 6-inch flanged duct enters the furnace section from the bottom and supports a 23-inch diameter by 6-inch deep burner tray (see Figure 3). Air passing through the inlet duct can be sampled via 4 sets of 4-inch blind flanges located on the top and south side of each duct section. A 2-inch half-coupling welded to the bottom of the last duct section can also be used. Air flow in the wind tunnel was controlled by adjusting the damper upstream of the exhaust filters. Nominal velocities at various damper settings determined before and during the experimental program are shown in Appendix A.

All four types of surfaces used in these experiments were held in the 23-inch diameter by 6-inch deep flanged burner tray shown in Figure 3. For experiments involving a smooth surface of sandy soil bare of vegetation, the flanged burner tray was filled with soil collected from the immediate locale. The surface was scraped level with the rim of the rim of the tray using a straight edge. For vegetation-covered soil, small tumbleweeds growing near the building were uprooted and pressed into the soil until the entire surface was covered with vegetation. The stainless steel surface was a 15-inch diameter by 2-inch deep pan embedded in the upstream side of the flanged burner tray. The rim of the pan was made flush with the rim of the tray by a bed of sandy soil and the space around the pan was filled with additional soil. The asphalt was held in a 15-inch diameter by 4-inch deep steel pan with a 4-inch wide lip which rested on the rim of the flanged burner tray. Heated asphalt was tamped into the pan to provide a 2-inch deep bed.

A measured amount of depleted uranium dioxide powder or uranyl nitrate solution was dispersed over the surface. The uranium dioxide powder is a milled material whose size distribution using various modes of dispersion is shown in Figure 4. The powder was sprinkled onto the various surfaces by hand and the uniformity of dispersion estimated visually. The amount of uranium dioxide dispersed was determined by weight difference of the dispenser before
FIGURE 2. INLET DUCT TO RADIOACTIVE AEROSOL RELEASE TANK - WIND TUNNEL
(PNL NEG. 710964-6)
URANIUM DIOXIDE OR NITRATE ON SURFACE

1/4" STL, 23" DIAM
6" DEEP

5/16" HOLE, 12 EQ SP ON 26" BC

1/4" SCH 40 HALF COUPLING
4 EQ SP ON 12" BC

1/4" STL, 28" DIAM

DIAL THERMOMETER

3/4" DIAM STL ROD
4 EQ SP ON 22" BC

APPROX SCALE: 1/4" = 1"

FIGURE 3. FLANGED BURNER TRAY, ELEVATION
FIGURE 4. URANIUM DIOXIDE POWDER SIZE DISTRIBUTION BY WHITBY CENTRIFUGE
and after distributing the powder.

A concentrated (0.51 g uranium per milliliter) uranyl nitrate solution in
nitric acid was dispensed by a graduated cylinder. Three dilute nitric acid
rinses of the cylinder were added to the liquid poured onto the various surfaces.
The tray with contents was inserted into the furnace section and bolted in place.
Air sampling equipment was inserted and placed into operation.

Particles in the air stream were sampled and collected by two filters and
an 8-stage Andersen cascade impactor (see Figure 1). Sample filter holders
were inserted into the air stream through the two 4-inch openings in the last
duct section upstream of the R.A.R.T. and the impactor sample via a 1/2-inch
stainless steel tube inserted through the 2-inch half-coupling in the bottom of
the duct. One of the filter samplers was fixed in the approximate center of
the duct while the other was moved down through the duct during sampling. In
later runs where separate samples were taken during and immediately following
a petroleum fire, the movable filter was held at the top of the duct during
the fire (the duration of the fire in many experiments was too short for any
manipulation) and moved down the duct after the fire. Flow through all
samplers was measured and appropriate corrections were applied to the volumes
of samples collected as required. Nominal sampling rate was 1 cfm on all
three samplers in all runs.

In experiments involving a gasoline fire, a gallon of gasoline was poured
onto the surface via a 1/2-inch stainless steel tube with a 90° bend entering
the top of the furnace section. A spray nozzle was placed over the end of the
tube to lessen the force of the liquid striking the surface and provide greater
dispersal of the gasoline. A gasoline-soaked rag was inserted into the furn-
ace section via the 2-inch half-coupling and ignited with a match. If the
gasoline was uniformly distributed throughout the soil, the gasoline con-
centration would be 0.70 gallons per cu. ft. of soil. At this level the soil was
saturated with shallow standing pools on the surface and exhibited burning char-
acteristics similar to those found in large scale fires (see later section,
"Large Scale Outdoor Gasoline Fires"). A gallon of gasoline produced a pool
approximately 1.3 inches deep over the stainless steel and asphalt (road-like
surfaces).

As soon as combustion was established, air flow was initiated. In most
cases, the gasoline ignited violently and, in a few cases, extinguished the
flames. For soil surfaces, bare or with a cover of vegetation, burning was vigorous for 5 to 10 minutes. The vigorous burning was followed by a period of stable, low flames covering the entire burner surface. Various areas then stopped burning and flames became widely separated. Eventually, all flames were extinguished. All the gasoline in the soil was not consumed; a strong odor of gasoline was detected even after the burner tray had cooled and was removed (approximately 16 hours). The soil surface was dry in most cases, but the soil greater than an inch under the surface was moist. The burning of gasoline on the stainless steel and asphalt surface exhibited the characteristics of a standing pool fire. Ignition was vigorous and the subsequent rate somewhat dependent on air velocity over the surface. The fire on asphalt (road-like) surfaces appeared to burn somewhat longer (see Table II, "Shipping Accident Experiments -- Operating Conditions") and with more smoke indicating some involvement of the asphalt. Air temperatures were monitored via thermocouples inserted through the duct wall and soil, the soil temperature 1/2- and 1-inch under the surface was monitored by dial thermometers inserted through the bottom of the burner tray and the duct wall temperature monitored by surface thermometers attached to the side of the middle of the three downstream sections.

Samples were collected immediately upon completion of a run and the remainder of the equipment allowed to stand overnight before clean-up. The filter holders and cascade impactor were removed and taken to the analytical hood for processing. Sample dissolution was initiated if time allowed. The flanged burner tray was removed and covered with a large plastic bag until the soil could be packaged for disposal. In the experiments involving a stainless steel surface (SA-17, SA-18, and SA-19), attempts were made to recover uranium residues in the 15-inch diameter tray.

The pertinent operational parameters for the 40 shipping accident experiments are tabulated in Table II, and are detailed in Appendix B, "Experimental Observations During Individual Shipping Accident Experiments."

Sample Preparation and Analysis

Particles of uranium compounds collected on the glass fiber filters and glass impactor collection plates were dissolved by mixed acid leach. The filters or plates were placed in pre-marked beakers and covered with a 9N HNO_3-0.1 N HF solution. The solution was warmed for approximately an hour. Impactor plates were then removed but the 2-inch diameter glass fiber filters
<table>
<thead>
<tr>
<th>Sample Designation</th>
<th>Uranium Form Used</th>
<th>Grams Used (g)</th>
<th>Wind Speed Used (km/hr)</th>
<th>Temp. (°C)</th>
<th>Burning Time (min.)</th>
<th>Total Sampling Time (min.)</th>
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<tr>
<td><strong>Smoke Test - No Generation</strong></td>
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<td>UO₂ Powder</td>
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<td><strong>Inhaled Smoke</strong></td>
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<td>UO₂ Powder</td>
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<td>23</td>
<td>550</td>
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<tr>
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<td>23</td>
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<td>SA-14</td>
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<td>550</td>
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<td>6</td>
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<td>78.1</td>
<td>23</td>
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<td><strong>Solidified Metal</strong></td>
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<td>SA-19a</td>
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<td>SA-19b</td>
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<tr>
<td><strong>Rock-like Surface</strong></td>
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<td>0.28</td>
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<tr>
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<td>--</td>
<td>26</td>
<td>0.28</td>
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<td>58.1</td>
<td>4</td>
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<td>0.37</td>
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<td></td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>0.37</td>
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</tbody>
</table>

* Not Measured

UNH - Uranyl Nitrate Hexahydrate
were in almost all cases dissolved. The heat was increased and the solutions concentrated to near dryness. After cooling, the solution was filtered and the volume made up to a known quantity. The solutions were placed in 15 ml, screw cap vials and submitted to the Technical Analysis Section for measurement.

The quantity of uranium in the liquid was measured by fluorimetry. A 100 microliter aliquot of solution or a dilution was dried in a platinum dish and fluxed with NaF. The uranium fluorescence under ultraviolet light was measured. Replicate aliquots were run on each sample as well as "spikes" to measure the amount of fluorescence quenched by extraneous materials in the liquid. Appropriate corrections for quench were applied as required. The minimum detectable concentration under these conditions is $10^{-5}$ gram uranium per milliliter. Due to the quenching problem encountered with these samples, the precision at a 95 percent confidence level is ± 35 percent.

**LARGE SCALE OUTDOOR GASOLINE FIRE**

To investigate the realism of the conditions obtained during the engineering scale experiments, we conducted a large-scale gasoline fire test. Very little recorded data has been published on the temperatures, surface area encompassed and duration of such fires. Information is available on the general characteristics of petroleum fires, [6,7,8] extinguishment[9], and the consequences of fires.[10,13] Combustible liquids burn by the formation of a combustible vaporous mixture near the surface.[14] The rate of burning is largely determined by the rates of mass and heat transfer between phases.[15] Plant fire protection personnel estimated the area encompassed under various conditions as:

- On an impervious surface, in the range of 3 to 5 sq. ft. per gallon of gasoline if the gasoline is immediately ignited. Maximum temperatures are in the range of 1700 to 2500°F.

- On porous surfaces, the gasoline will not spread as widely and encompass an area in the range of 2 to 4 sq. ft. per gallon, depending on the porosity of the soil. Temperatures will be lower in the range of 400 to 700°F.

Large gasoline carriers can hold up to 12,000 gallons of gasoline and would burn in an estimated 2 to 3 hours if unrestrained. Blacktop will burn if sat-
urated with gasoline. [16]

A vehicular accident involving a large quantity of gasoline occurred locally on September 8, 1970. [17] A gasoline truck and trailer carrying some 8000 gallons was struck by another vehicle. The trailer containing approximately 2000 gallons of gasoline was hurled over the truck. Although the side of the trailer was ruptured, the vehicle came to rest on the undamaged side and retained most of its contents. The truck slid on its side, rupturing that side. The contents of the truck spilled onto the road and were ignited by sparks. The fire burned some 2 to 2-1/2 hours intermittently over a 5 hour period -- fire control measures would extinguish the flames which would be rekindled by "hot spots". The fire covered an estimated 10,000 sq. ft. of surface.

To ascertain if the conditions observed during the engineering scale experiments provide a reasonable simulation of a large scale outdoor fire, 500 gallons of gasoline were quickly released, dispersed and ignited on an instrumented test site. The quantity of gasoline chosen is one-third the maximum volume (1500 gallons) trucked within the Hanford site boundaries. The area involved was determined by actual measurement of the area after combustion. Both air and surface temperatures were measured. Wind speed and direction were monitored. An attempt was made to evaluate the fractional airborne release from uranium dioxide powder and nitrate solutions by determining the amount lost from exposed prepared surfaces held in canisters buried in the ground.

**Description of Site and Apparatus**

The study was conducted in a natural depression some 15 to 20 feet deep located on arc 12 of the "unstable grid" system east of the 200-W Exclusion area (see Figure 5). The site is some 1/2 to 1 mile northeast of the 622-R Building. The depression extends some 200 feet in a southwest to northeast direction. A 30 ft. by 30 ft. test plot was located in the southwestern end of the depression with the test equipment for the quick release and dispersal of gasoline (described below) located in the center of the southwest edge of the test section. A 30 ft. wide fire break was bladed around the test site and filled with foam prior to the test. The Fire Protection unit provided coverage during the initial portions of the test. Relationship of the 500-gallon tank, spillway and location of temperature sensors and canisters is shown in Figure 5.
FIGURE 6. LARGE SCALE GASOLINE FIRE-ARRANGEMENT OF CANISTER SPS, TEMPERATURE SENSORS AND IGNITERS
Air and surface temperatures were measured by a cluster of thermocouples and thermistors held by a support rod in 6 locations on the test site (see Figures 6 and 7). Each cluster contained 3 stainless steel clad, iron-constantan thermocouples and 2 thermistors fixed to a 1/2-inch diameter iron support rod. The support rod was buried in the ground so that the thermocouples extended 6, 4, and 2 inches above the surface and the thermistors were positioned 1/2 and 1 inch under the surface. The arrangement is pictured in Figure 8, "Temperature Sensor Cluster". A third thermistor was located at the thermocouple-lead wire junction approximately 12 inches beneath the surface for temperature compensation. The lead wire was buried some 12 inches under the surface of the test site and connected to a magnetic tape data collection unit located on the rim of the depression some 100 feet southwest of the test site. Wind speed and direction by a cup anemometer and vane were monitored and recorded at this location.

Six pairs of 3-inch, open, soil-filled canisters -- one with uranium dioxide powder and nitrate solution on the surface -- were positioned on the test site at each temperature sensor cluster location (see Figures 6 and 7). The canister arrangement is diagrammed in Figure 9, "Large Scale, Outdoor Gasoline Fire -- Uranium Canister". Prior to the test, each canister was filled with soil from the test site within an inch of the top. The desired quantity of uranium dioxide powder or nitrate solution was dispersed over the surface, and a plastic bag taped over the opening. The canisters were inserted 11 inches into the ground so the level of the soil in the canister was level with the test site surface. The rim was allowed to project above the surface to provide some protection from the deluge that would sweep the surface on release of the gasoline. Immediately before the test, the soil in each canister was saturated with gasoline. The plastic bags were left loosely in place to afford a little additional protection to the surfaces in the canisters.

The equipment used for the fast release and dispersal of 500 gallons of gasoline during this study is diagrammed in Figures 10 and 11, and consisted of:

- A 4 ft. by 4 ft. by 5 ft. box of unpainted 1/4 inch thick steel with a hinged lid. The volume of the box was approximately 80 cu. ft. (600 gallons).
FIGURE 7. LARGE SCALE OUTDOOR GASOLINE FIRE, TEMPERATURE SENSORS AND URANIUM CANISTERS (PNL NEG. 713037-5)
FIGURE 9. LARGE SCALE OUTDOOR GASOLINE FIRE, URANIUM CANISTER
FIGURE 10. LARGE SCALE OUTDOOR GASOLINE FIRE, GASOLINE CONTAINER
- A frame of 4 in. by 8 in. "I" beam which supported the box some 4-1/2 feet above the ground. The base of the support was a 6 ft. by 12 ft. rectangle.
- A spillway channeling the gasoline to the edge of the test site. The spillway was 5 ft. wide and about 16" deep.

The box was held in the "I" beam support structure by a 1-3/4 inch diameter axle welded into each end. The axles were held in a bearing welded onto the "I" beam support. Spacers on the axles centered the container. An inherently unstable configuration was produced by welding the axles off-center (approximately 5 inches below and 3 inches behind center). The box was held in an upright position by a 1-3/4 inch rod welded to the back of the box and held at each end by "dogs" of 1/8 inch steel. Positive locks were also provided to prevent inadvertent release of the contents. Gasoline was pumped into the container via a 3-inch nipple welded into the hinged lid used to provide some containment of the vapor yet allowing a rapid release of the contents. The gasoline was released by removing the positive locks and disengaging the "dogs" from a distance by ropes attached to the 4 ft. arms on the "dogs". A mechanical stop, a strip of 2-inch angle iron welded to the "I" beam support -- limited the rotation of the box and assured the release of all the gasoline. The hinged lid was prevented from entering the spillway by a 2-inch angle iron welded on its edge. The gasoline was remotely ignited by smoke bombs held off the ground and fired by a solenoid-activated pin (see Figure 12).

An attempt to disperse the gasoline over the test site by placing large rocks directly in its path were only partially successful. A considerable number of loose rocks and a fair amount of soil were displaced during the initial wave of gasoline. A 12 ft. length of 4-inch by 8-inch "I" beam was positioned at the opposite edge of the test site to limit the flow of gasoline.
FIGURE 12. LARGE SCALE OUTDOOR GASOLINE FIRE, IGNITER WITHOUT SMOKE BOMB ATTACHED (PNL NEG. 713037-2)
RESULTS

ENGINEERING-SCALE EXPERIMENTS

The percentage of uranium airborne based on the quantity of uranium found in each sampler, the fraction of the total airstream sampled and the amount of uranium used as a source is tabulated in Table III, "Uranium Results -- Engineering Scale Experiments". In experiments conducted in the first phase of the study (experiments SA-1 to SA-21), airborne concentrations were calculated from the quantity of uranium collected on the "fixed" and "movable" filter as well as the summation of all uranium collected in stages and filter of the Andersen cascade impactor. The individual samples show considerable variation (in a few cases two orders of magnitude) and may indicate a serious channeling of airflow (see Appendix C). No sampling position exhibited a consistently higher value than any other. Since there is no overriding reason for the choice of one value over another, the percent uranium airborne for a given experiment is simply an arithmetic average of all three samples taken. Only the "fixed" filter values were used in the second phase (experiments SA-22 to SA-26).

The aerodynamic size fractions with which the airborne uranium was associated was determined by measuring the uranium collected on 3.25 inch diameter glass plates and final 2-inch diameter glass fiber filters in an 8-stage Andersen cascade impactor (see Appendix D). The fraction of the total uranium collected in the impactor was determined for each stage and final filter. The cumulative fraction in successive stages was plotted against the nominal stage cut off diameters on log-probability paper to produce the size distributions of the uranium airborne during each experiment. Figure 13 shows some examples of the distributions found, "The Activity Median Diameter" *(AMD) was picked off these curves as was the percent of uranium associated with particles less than 10 microns Aerodynamic Equivalent Diameter (AED). Particles enter and are deposited in the lungs with varying efficiencies dependent upon particle size. Above a certain size, particles are trapped in the nose and throat and are inhaled. Below a certain size, particles behave as a gas and flow with the tidal volume. Considering all particles less than 10 microns AED as respirable would give a "conservative estimate" of the inhalation hazard.

* Activity Median Diameter -- 50 percent of the uranium airborne is associated with particles that have aerodynamic equivalent diameters less than the stated diameter.
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Smooth Sandy Soil</td>
<td>low</td>
<td>2</td>
<td>0.43 6 hrs.</td>
<td>56</td>
<td>3</td>
<td>0.63 6 hrs.</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>22a</td>
<td>0.042 24 hrs.</td>
<td>54</td>
<td>22b</td>
<td>0.048 1 hr.</td>
<td>72</td>
</tr>
<tr>
<td></td>
<td></td>
<td>22c</td>
<td>0.0092 24 hrs.</td>
<td>54</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>moderate</td>
<td>1</td>
<td>2.3 24 hrs.</td>
<td>75</td>
<td>23b</td>
<td>0.34 0.3 hrs.</td>
<td>62</td>
</tr>
<tr>
<td></td>
<td></td>
<td>23a</td>
<td>24.4 24 hrs.</td>
<td>40</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>23c</td>
<td>1.0 24 hrs.</td>
<td>60</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Air Dried</td>
<td>low</td>
<td>4b 0.0069 24 hrs.</td>
<td>74</td>
<td>5</td>
<td>0.0074 5.5 hrs.</td>
<td>64</td>
</tr>
<tr>
<td></td>
<td></td>
<td>8b</td>
<td>0.0054 24 hrs.</td>
<td>77</td>
<td>9</td>
<td>0.004 5 hrs.</td>
<td>18</td>
</tr>
<tr>
<td></td>
<td></td>
<td>6b</td>
<td>0.93 24 hrs.</td>
<td>22</td>
<td>7</td>
<td>0.12 7 hrs.</td>
<td>32</td>
</tr>
<tr>
<td></td>
<td></td>
<td>26b</td>
<td>1.1 24 hrs.</td>
<td>64</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>UNH</td>
<td>low</td>
<td>4a 0.049 6 hrs.</td>
<td>76</td>
<td>10</td>
<td>0.0017 5.5 hrs.</td>
<td>75</td>
</tr>
<tr>
<td></td>
<td></td>
<td>8a</td>
<td>0.012 24 hrs.</td>
<td>84</td>
<td>26a</td>
<td>0.54 0.13 hrs.</td>
<td>40</td>
</tr>
<tr>
<td></td>
<td>moderate</td>
<td>6a</td>
<td>0.039 28 hrs.</td>
<td>68</td>
<td></td>
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<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Vegetation</td>
<td>low</td>
<td>11</td>
<td>0.046 24 hrs.</td>
<td>82</td>
<td>12</td>
<td>0.014 6 hrs.</td>
<td>91</td>
</tr>
<tr>
<td></td>
<td></td>
<td>13</td>
<td>1.0 24 hrs.</td>
<td>40</td>
<td>14</td>
<td>1.1 6 hrs.</td>
<td>19</td>
</tr>
<tr>
<td></td>
<td>UNH</td>
<td>low</td>
<td>-- --</td>
<td>--</td>
<td>16</td>
<td>0.009 6 hrs.</td>
<td>78</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Stainless Steel</td>
<td>low</td>
<td>17b*</td>
<td>0.09 4.8 hrs.</td>
<td>13</td>
<td>17a</td>
<td>0.12 0.12 hrs.</td>
<td>66</td>
</tr>
<tr>
<td></td>
<td></td>
<td>25a</td>
<td>7.6 24 hrs.</td>
<td>14</td>
<td>25b</td>
<td>1.1 0.13 hrs.</td>
<td>7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>25c</td>
<td>0.7 24 hrs.</td>
<td>42</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>UNH</td>
<td>low</td>
<td>19b 0.023 6 hrs.</td>
<td>76</td>
<td>19a</td>
<td>1.1 0.22 hrs.</td>
<td>40</td>
</tr>
<tr>
<td></td>
<td></td>
<td>18b*</td>
<td>2.6 5 hrs.</td>
<td>30</td>
<td>18a</td>
<td>1.1 0.17 hrs.</td>
<td>34</td>
</tr>
<tr>
<td></td>
<td>moderate</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Road-like Surface</td>
<td>low</td>
<td>21b*</td>
<td>0.14 6 hrs.</td>
<td>62</td>
<td>21a</td>
<td>0.041 0.37 hrs.</td>
<td>70</td>
</tr>
<tr>
<td></td>
<td></td>
<td>20b*</td>
<td>0.051 5 hrs.</td>
<td>59</td>
<td>20a</td>
<td>0.4 0.28 hrs.</td>
<td>68</td>
</tr>
</tbody>
</table>

1. Percent airborne less than 10 microns aerodynamic equivalent diameter.
2. Approximately 2.5 mph.
3. In range of 20 to 23.5 mph.
4. Dried in flowing air at stated velocity for a minimum of 24 hours.
5. Uranium Nitrate Hexahydrate.
6. Dried in flowing air at stated velocity for a minimum of 24 hours after gasoline fire.
7. Drying and immediately following gasoline fire.
8. Dried in flowing air at stated velocity for a minimum of 24 hours after gasoline fire.
FIGURE 13. ACTIVITY DISTRIBUTION INTO VARIOUS AERODYNAMIC SIZE FRACTIONS DURING SELECTED EXPERIMENTS
Aerodynamic Entrainment of Uranium from Various Surfaces

The quantities of uranium particles in the respirable range that were aerodynamically entrained in the airstream under the stated conditions are tabulated in Table IV. The data indicate:

a. Entrainment of uranium under all combinations used in this study are low in low wind speeds (maximum of 0.23 percent in 5 hours from uranium dioxide on sandy soil).

b. Entrainment of uranium dioxide powder from all surfaces by wind speeds around 20 mph are considerably higher (0.4 to 24.4 percent in 24 hours).

c. Entrainment of uranium from the nitrate solution on soil is low.

d. The entrainment from the dried solid residue of uranium nitrate solutions is higher than from the solution itself. The residues from solutions involved in a gasoline fire at higher wind speeds appear to be higher.

Since the collection period for these samples range from 5 to 24 hours, some information on the rate of entrainment was obtained in the second phase of the experimental program. The data from these experiments is shown in Figures 14 through 17, where the percent of total uranium collected under the stated conditions are plotted against time. At a wind speed of 2.5 mph, the entrainment of uranium dioxide powder, fresh or involved in a gasoline fire, from sandy soil appears to be essentially complete in approximately 16 hours. The initial rise is fairly rapid -- around 50 percent of the total entrained is entrained in the first two hours. At a wind speed of 20 mph, entrainment from freshly deposited uranium dioxide powder on sandy soil is approximately 99 percent complete within the first hour. If the powder is involved in a gasoline fire, the uranium is entrained more slowly and may not be complete in 24 hours. The entrainment of uranium dioxide powder, fresh or involved in a gasoline fire from a stainless steel surface by air at 20 mph also appears to be incomplete in 24 hours. Again the entrainment of the powder involved in a fire is slower. The entrainment of uranium from the solid residue of a nitrate solution on smooth sandy soil devoid of vegetation appears to be far from complete and could continue for an appreciable period of time.

If the data presented in Table IV is viewed in the light of this information, the aerodynamic entrainment of materials involved in gasoline fires increases in importance. At a wind speed of 20 to 23 mph, the entrainment from residues of uranium dioxide from stainless steel could rise to the levels found...
### TABLE IV

AERODYNAMIC ENTRAINMENT OF URANIUM PARTICLES IN THE RESPIRABLE SIZE RANGE FROM VARIOUS SURFACES

<table>
<thead>
<tr>
<th>Surface</th>
<th>Uranium Dioxide Powder</th>
<th></th>
<th>Uranium Nitrate Solution</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2.5 mph</td>
<td>20-23 mph</td>
<td>2.5 mph</td>
<td>20-23 mph</td>
</tr>
<tr>
<td>Smooth, Sandy Soil</td>
<td>0.34 (6)</td>
<td>1.7 (24)</td>
<td>0.0051 (24)</td>
<td>0.207 (24)</td>
</tr>
<tr>
<td></td>
<td>0.032 (24)</td>
<td>9.6 (24)</td>
<td>0.0042 (24)</td>
<td>0.707 (24)</td>
</tr>
<tr>
<td></td>
<td>0.005 (24)*</td>
<td>0.68 (24)</td>
<td>0.037 (6)</td>
<td>0.027 (28)</td>
</tr>
<tr>
<td>Vegetation Cover</td>
<td>0.0038 (24)</td>
<td>0.4 (24)</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Stainless Steel</td>
<td>0.075 (4.8)*</td>
<td>1.1 (24)</td>
<td>0.017* (6)</td>
<td>0.78 (5)</td>
</tr>
<tr>
<td>Asphalt Road-like Surface</td>
<td>0.087* (6)</td>
<td>--</td>
<td>0.333* (6)</td>
<td>--</td>
</tr>
</tbody>
</table>

+ Solid Residues from air dried UNH solutions.
* Solid residues remaining after a gasoline fire.
Figure 14. Aerodynamic entrainment of uranium dioxide powder from smooth, sandy soil at an air velocity of 2.5 MPH.
Figure 14. Aerodynamic entrainment of uranium dioxide powder from smooth, sandy soil at an air velocity of 2.5 MPH.

Figure 15. Aerodynamic entrainment of uranium dioxide powder from smooth, sandy soil at an air velocity of 20 MPH.
FIGURE 16. AERODYNAMIC ENTRAINMENT OF URANIUM DIOXIDE POWDER FROM A STAINLESS STEEL SURFACE AT AN AIR VELOCITY OF 20 MPH
Figure 17. Aerodynamic entrainment of solid residues from uranium nitrate after a gasoline fire on smooth, sandy soil at an air velocity of 20 MPH.
for freshly deposited powder. Entrainment from uranium nitrate residue could also rise to or exceed this level.

**Fractional Release of Uranium During a Petroleum Fire**

The fraction of uranium which was present in the source that becomes airborne during a gasoline fire, or attached to particles in the respirable size range, are listed in Table V. No single surface or uranium form appears to have an overriding effect on the fractional values. It appears that a combination of circumstances determines the fractional airborne release under these circumstances. Some of these values are for fractional airborne releases during and immediately following a gasoline fire (marked with an asterisk). The data from experiments where separate values were measured indicated that the quantity airborne during the fire in these experiments is significantly greater than the quantity entrained after the fire.

A few general observations can be made:

- Under comparable conditions, increasing the velocity of air passing over the surface increases the amount of material airborne under the stated conditions. As indicated by the aerodynamic entrainment experiments discussed in the previous section, the rate at which the material is airborne increases (see Figure 9 through 12).

- Evaporation of liquids which results in the formation of a film on surfaces appears to provide a potential for greater release. The highest fractional release measured was from uranium nitrate solution on a stainless steel surface at an air speed of 23 mph -- 4.3 percent. The same combination of conditions gave the third highest fractional release at an air speed of approximately 2.5 mph. Uranium nitrate solutions on vegetation-covered soil and asphalt gravel mixtures also had fractional releases in the upper half of the table. The fractional release value for uranium nitrate solutions on vegetation -- 0.17 and 0.075 percent -- represent some 0.39 and 0.19 milliliters of the 0.51 grams uranium per milliliter solution used as the source and could easily be visualized as liquid clinging to the vegetation.

- Fractional releases of both liquid and powder are higher from hard, impervious surfaces.
### Table V

**Fractional Release of Uranium Involved in a Gasoline Fire**
(Weight percent of source airborne as particles in respirable range.)

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Source</th>
<th>Air Velocity (mph)</th>
<th>Surface Area (sq. ft.)</th>
<th>Grams of U in Source</th>
<th>Grams Uranium/ sq. ft.</th>
<th>Percent Release</th>
</tr>
</thead>
<tbody>
<tr>
<td>SA-18a</td>
<td>UNH Solution, Stainless Steel</td>
<td>23</td>
<td>1.23</td>
<td>25.5</td>
<td>20.7</td>
<td>4.3</td>
</tr>
<tr>
<td>SA-14</td>
<td>UO₂ Powder, Vegetation Cover</td>
<td>23</td>
<td>2.89</td>
<td>69.5</td>
<td>24</td>
<td>0.59*</td>
</tr>
<tr>
<td>SA-19a</td>
<td>UNH Solution, Stainless Steel</td>
<td>&lt;4</td>
<td>1.23</td>
<td>25.5</td>
<td>20.7</td>
<td>0.44</td>
</tr>
<tr>
<td>SA-23b</td>
<td>UO₂ Powder, Sandy Soil</td>
<td>20</td>
<td>2.89</td>
<td>50</td>
<td>17.3</td>
<td>0.3</td>
</tr>
<tr>
<td>SA-15</td>
<td>UNH Solution, Vegetation Cover</td>
<td>23</td>
<td>2.89</td>
<td>51.1</td>
<td>17.7</td>
<td>0.17*</td>
</tr>
<tr>
<td>SA-20a</td>
<td>UNH Solution, Road-like Surface</td>
<td>&lt;4</td>
<td>1.23</td>
<td>25.5</td>
<td>20.7</td>
<td>0.16</td>
</tr>
<tr>
<td>SA-25a</td>
<td>UO₂ Powder, Stainless Steel</td>
<td>30</td>
<td>1.23</td>
<td>50</td>
<td>40.6</td>
<td>0.12</td>
</tr>
<tr>
<td>SA-17a</td>
<td>UO₂ Powder, Stainless Steel</td>
<td>&lt;4</td>
<td>1.23</td>
<td>51.1</td>
<td>15.8</td>
<td>0.079</td>
</tr>
<tr>
<td>SA-16</td>
<td>UNH Solution, Vegetation</td>
<td>&lt;4</td>
<td>2.89</td>
<td>51.1</td>
<td>17.7</td>
<td>0.075*</td>
</tr>
<tr>
<td>SA-7</td>
<td>Residue Air Dried UNH, Sandy Soil</td>
<td>23</td>
<td>2.89</td>
<td>51</td>
<td>17.6</td>
<td>0.038*</td>
</tr>
<tr>
<td>SA-3</td>
<td>UO₂ Powder, Sandy Soil</td>
<td>&lt;4</td>
<td>2.89</td>
<td>74.9</td>
<td>25.9</td>
<td>0.030*</td>
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<tr>
<td>SA-21a</td>
<td>UO₂ Powder, Road-like Surface</td>
<td>&lt;4</td>
<td>1.23</td>
<td>58.1</td>
<td>47.2</td>
<td>0.029</td>
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<td>SA-26a</td>
<td>UNH Solution, Sandy Soil</td>
<td>20</td>
<td>2.89</td>
<td>51</td>
<td>17.6</td>
<td>0.029</td>
</tr>
<tr>
<td>SA-22b</td>
<td>UO₂ Powder, Sandy Soil</td>
<td>2.5</td>
<td>2.89</td>
<td>50</td>
<td>17.3</td>
<td>0.017</td>
</tr>
<tr>
<td>SA-12</td>
<td>UO₂ Powder, Vegetation</td>
<td>&lt;4</td>
<td>2.89</td>
<td>78.3</td>
<td>27.1</td>
<td>0.013*</td>
</tr>
<tr>
<td>SA-9</td>
<td>Residue Air Dried UNH, Sandy Soil</td>
<td>&lt;4</td>
<td>2.89</td>
<td>51.5</td>
<td>17.8</td>
<td>0.0072*</td>
</tr>
<tr>
<td>SA-5</td>
<td>Residue Air Dried UNH, Sandy Soil</td>
<td>&lt;4</td>
<td>2.89</td>
<td>51</td>
<td>17.6</td>
<td>0.0045*</td>
</tr>
<tr>
<td>SA-10</td>
<td>UNH Solution, Sandy Soil</td>
<td>&lt;4</td>
<td>2.89</td>
<td>47.8</td>
<td>16.5</td>
<td>0.0013*</td>
</tr>
</tbody>
</table>

* During and immediately following a gasoline fire.
The smallest fractional releases found in these experiments are under those conditions which allow the uranium to penetrate into and becomes a part of the surface.

**LARGE-SCALE OUTDOOR GASOLINE FIRE**

The large scale, outdoor gasoline fire demonstrated that the conditions produced in the engineering scale experiments approach those produced in a large gasoline fire and are thus a reasonable simulation of such occurrences.

- The burning characteristics of the large scale gasoline fire progressed through three stages:
  a. An intense violent stage lasting approximately 15 minutes. Temperatures exceeded 1000°C with flames 15 to 20 feet high (see Figure 18).
  b. A period of about 30 minutes of uniform stable burning with temperatures in the range of 500 to 700°C. Flame heights varied from approximately 6 to 12 inches near the end of the period.
  c. A period of increasing flame breakup and separation lasting some 45 minutes. Temperatures decreased to approximately 300°C.

The sequence, duration and characteristics of the fire are within the range found in the wind tunnel experiments in the 242-2 Building facility. Flame height and ground turbulence may be less accurately scaled.

- The fire covered an area of about 1000 sq. ft. (see Figure 19). The gasoline was thus at a concentration of about 0.5 gallon of gasoline per sq. ft. of surface similar to that used in the engineering scale experiments. Since the gasoline could only penetrate to a maximum depth of 6 inches in the engineering scale experiments while the soil was moist to a depth of 12 inches following the large scale gasoline fire, as much gasoline was probably available in the wind tunnel experiments per unit area as occurred in the large scale test.

- The uranium recovered from the soil-filled canisters following the fire did not vary significantly from unexposed, soil-filled canisters of the same material. All six pairs of canisters were collected and examined following the fire. Three of the twelve canisters distributed over the test site were still intact:
  a. Both canisters (#3 containing uranium dioxide powder and #4 contain-
FIGURE 19. AREA COVERED BY LARGE SCALE OUTDOOR GASOLINE FIRE
FIGURE 18. LARGE SCALE OUTDOOR GASOLINE FIRE, INITIAL STAGES OF BURNING (PNL NEG. 713037-27)
ing uranium nitrate solution) at position #2 were buried under several inches of loose rocks and soil. The plastic bag over both canisters was retrieved intact.

b. Canister #12 at grid position #6 containing uranium nitrate solution was not in the burn area (See Figure 19) and, although the tip of the plastic bag was melted, most of the bag was found on the canister. At no point in the test was this canister observed burning.

All the material in the upper portion of each canister was placed in a pre-marked beaker and repeatedly leached using warm 9 M HNO₃-0.1 M HF. Quantity of material processed varied from 250 to 650 grams. Due to the large amounts of salts generated, reliable answers could not be obtained using controlled potential coulometry. Uranium measurements were obtained via fluorimetry by making large dilutions to reduce the effects of extraneous cations. The average recoveries for the exposed uranium dioxide powder and nitrate solution canisters did not vary significantly from the control and unexposed canisters of the same material. Because of the wide statistical variation in analyses, it is estimated that the minimum fraction which could have been measured and shown to be significantly different from the uninvolved canisters would be about 50%. In light of the anticipated release of a fraction of one percent, from the R.A.R.T. experiments, no conclusions regarding release fraction in the large scale fire experiment can be made.

CONCLUSIONS

The engineering scale experiments conducted in the wind tunnel in the 242-B Building facility reasonably simulated the conditions imposed by a large scale gasoline fire on a porous surface. The sequence, burning characteristics, temperatures and duration of a fire involving 500 gallons of gasoline were within the range of those observed during the engineering scale experiments.

No single parameter of those evaluated in this study — physical form of the active material, surface, wind speed or the presence or absence of a fire — exerts a controlling influence on the fractional airborne release under all circumstances. The combination of all these parameters produces a situation
favorable or unfavorable to release. Thus, a liquid which penetrates, incorporates and binds together a porous surface produces a situation unfavorable to airborne release. The same liquid on a hard, impervious surface may produce a thin film which produces a fine particle when heated. The presence of vegetation cover reduces the airborne release of powder, but may increase the quantity of material airborne from a liquid — by drying of the thin film of liquid clinging to the vegetation and the subsequent release of the particle formed in the air stream. Higher air speeds and fires generally increase fractional airborne release.

The greatest release combination of those evaluated was uranium nitrate liquid on a hard impervious surface in a gasoline fire. During the fire, 4.3 percent of the uranium in the source was airborne in the aerodynamic equivalent size fraction less than 10 microns (nominal respirable range) under a wind at a velocity of 23 mph. At a wind speed of less than 4 mph, 0.44 percent was airborne. Even after the fire, the fractional airborne release remains high.

ACKNOWLEDGEMENTS

Acknowledgement is made to D.W. Glover, J.W. Sloat, and M.T. Dana for their invaluable assistance in preparation and execution of the Large Scale Cuticor Gasoline Fire Test. Also to C.O. Harvey, Technical Analysis, for the uranium analysis.
REFERENCES


APPENDIX A

AIR VELOCITIES VS. DAMPER SETTINGS -- INLET DUCT R.A.R.T. 242-B BUILDING
AIR VELOCITIES VS. DAMPER SETTINGS -- INLET DUCT R.A.R.T. 242-B BUILDING

All engineering scale experiments in this study were performed in the furnace section of the inlet duct of the Radioactive Aerosol Release Tank located in the 242-B Building facility. Since the velocity of the air passing over the contaminated surfaces is one of the variable parameters of the study, air velocities in the furnace section vs. damper settings were measured on three occasions -- before the initial experiments and before and after replacement of the exhaust HEPA filters.

The inlet duct is a 24-inch by 24-inch duct, 21 feet long, made of 1/8 inch steel and is shown in Figure 1. The inlet is sealed by four 24-inch by 24-inch by 12-inch deep HEPA filters (see Figure A1). An 18-inch long flash arrestor separates the filters from the furnace section. Air is drawn into the system by a large blower located downstream of the RART.

The furnace section is a 3-foot long section of the duct lined on the top and sides with castable refractory material and is shown in Figure A2. Three half couplings (3/4", 2", and 1/4", Sch. 40 pipe) provide access into the section. A 24-inch diameter by 6-inch deep flange at the bottom holds the 23-inch diameter Flanged Burner Tray shown in Figure A-2. The internal dimensions of the furnace section are 19-1/2 inch by 18 inch.

Prior to the beginning of the study, velocities were measured by a vane anemometer placed in the duct at its entry into the R.A.R.T., and a pitot tube inserted through the side of the duct. The results are tabulated in Table A-1. The damper position indicator was mounted such that the indicated settings are 90° out of phase with the damper position. Useful accurate readings could not be obtained using the vane anemometer or pitot tube at the damper setting corresponding to minimum flow. Velocities greater than 250 ft. per minute should be measurable with the vane anemometer used. Under the atmospheric conditions prevailing, significant differential pressure readings should be obtained at air velocities in excess of 450 ft. per minute.

An air velocity of 215 ft. per minute in the inlet duct corresponds to a nominal velocity of 4 mph in the furnace section and was arbitrarily assigned to the lowest airflow used. The velocity profiles in the duct and furnace section at higher air flow was also measured, and although a considerable variation in velocity was found across the duct, the centerline velocity was found to be a reasonable average value through the duct.
FIGURE A-1. FILTER BOX, FLAME ARRESTER AND FURNACE SECTION OF INLET DUCT TO THE RADIOACTIVE AEROSOL RELEASE TANK (SECTION)
<table>
<thead>
<tr>
<th>Indicated Damper Setting</th>
<th>Vane Anemometer</th>
<th>Differential Pressure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Open</td>
<td>*</td>
<td>&lt;250</td>
</tr>
<tr>
<td>1/4</td>
<td>92.5</td>
<td>648</td>
</tr>
<tr>
<td></td>
<td>92.3</td>
<td></td>
</tr>
<tr>
<td>1/2</td>
<td>59</td>
<td>1017</td>
</tr>
<tr>
<td></td>
<td>60</td>
<td>1000</td>
</tr>
<tr>
<td>3/4</td>
<td>55.2</td>
<td>1087</td>
</tr>
<tr>
<td></td>
<td>55.2</td>
<td></td>
</tr>
<tr>
<td>Closed</td>
<td>56</td>
<td>1071</td>
</tr>
<tr>
<td></td>
<td>56.6</td>
<td></td>
</tr>
</tbody>
</table>

Relative Humidity: 37%
Barometric Pressure: 30.002 in.
Temperature: 61°F

*Would not turn vane.*
In order to measure air velocities at minimum flow in the system, a vane anemometer with a pressure drop in hundredths of an inch water gauge was obtained. The anemometer was calibrated with a Roots meter. The configuration used in calibration is shown in Figure A3, and the results are shown in Table A-II. A value of 0.097 cu. ft. per count was determined as compared with the factory calibration of 0.093 std. cu. ft. per count. The response of the anemometer checked at various flow rates between 20 and 50 cfm did not indicate any change in calibration related to flow rate.

The pressure drop through the exhaust HEPA increased towards the end of the initial portion of the experimental study. The decision was made to replace the exhaust HEPA filters after the experimental work was completed and the duct could be thoroughly cleaned. The filters were replaced between experiments SA-21 and SA-22.

![Diagram](image)

**FIGURE A3**

**CALIBRATION SETUP FOR PR-3 FLOWMETER**

† SCI-MED model PR-3, SCI-MED, 13010 County Road 6, Minneapolis, Mn. 55441

* Roots Connerville positive displacement flowmeter 7M 125, Connerville, Indiana.
### TABLE A-II

CALIBRATION OF SCI-MED MODEL FR-3 FLOWMETER

<table>
<thead>
<tr>
<th>Flow Rate cfm</th>
<th>Time Min.</th>
<th>Roots Meter Reading-Ft³</th>
<th>Count Flow Sensor</th>
<th>Count Ft³</th>
<th>Ft³ Count</th>
</tr>
</thead>
<tbody>
<tr>
<td>4/13/72 -- Flow Sensor No. 109</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>42</td>
<td>1</td>
<td>42</td>
<td>513</td>
<td>12.2</td>
<td>0.0313</td>
</tr>
<tr>
<td>42</td>
<td>10</td>
<td>420</td>
<td>5356</td>
<td>12.7</td>
<td>0.0787</td>
</tr>
<tr>
<td>43.2</td>
<td>20</td>
<td>863</td>
<td>8534</td>
<td>9.9</td>
<td>0.101</td>
</tr>
<tr>
<td>42</td>
<td>30</td>
<td>1280</td>
<td>13080</td>
<td>10.4</td>
<td>0.0961</td>
</tr>
<tr>
<td>42</td>
<td>60</td>
<td>2522</td>
<td>26166</td>
<td>10.4</td>
<td>0.0961</td>
</tr>
<tr>
<td>50.8</td>
<td>60</td>
<td>3046</td>
<td>31724</td>
<td>10.4</td>
<td>0.0961</td>
</tr>
</tbody>
</table>

| 4/14/72 -- Using Cyclonaire 1 pump |
| 34.7          | 30        | 1040                     | 10859             | 10.4      | 0.0961    |
| 50.3          | 30        | 1510                     | 15848             | 10.5      | 0.0961    |
| 21.8          | 60        | 1312                     | 13945             | 10.3      | 0.0971    |
| 33.5          | 60        | 2010                     | 20784             | 10.3      | 0.0971    |
| 50.5          | 10        | 505                      | 5314              | 10.5      | 0.0952    |
| 22            | 10        | 220                      | 2262              | 10.3      | 0.0971    |
| 21.8          | 60        | 1310                     | 13456             | 10.3      | 0.0971    |
| 21.2          | 60        | 1309                     | 13563             | 10.3      | 0.0971    |

| 4/19/72 -- Flow Sensor No. 109 |
| 23            | 3         | 69                       | 698               | 10.1      | 0.0990    |
| 23            | 10        | 230                      | 2363              | 10.3      | 0.0971    |
| 51.2          | 10        | 512                      | 5372              | 10.5      | 0.0952    |
| 22.9          | 10        | 229                      | 2362              | 10.3      | 0.0971    |
| 22.8          | 30        | 685                      | 6950              | 10.3      | 0.0971    |

| 4/24/72 -- Flow Sensor No. 109 |
| 51.2          | 10        | 512                      | 5168              | 10.1      | 0.0990    |
| 50.8          | 10        | 508                      | 4991              | 9.8       | 0.102     |
| 21.9          | 10        | 219                      | 2162              | 9.9       | 0.101     |

Average: 0.097 cu.ft./count
The air velocities in the furnace section at various damper settings were measured before and after the filter change (see Figure A-5).

A schematic drawing of the sensor placement is shown in Figure A-4, and the results are given in Table A-III. Very little change in air velocity was detected before and after filter replacement at minimum flow. The measured values were 2.46 and 2.65 mph respectively. A value of 2.5 mph was assigned for the air velocity at minimum flow for all subsequent experiments. The air velocity increased at maximum flow from 13.4 to 19.6 mph after replacement. A nominal velocity of 25 mph was assigned for maximum flow experiments.

---

**Dimension Duct:** 24" x 24" (4 ft²)

**Dimension Furnace Section:** 19.5" x 18", 351 in.² (2.44 ft²)

**Cross Section Flow Sensor:** Diameter 4.81 in.
Area 19.13 in.², 0.126 ft²

**FIGURE A-4**

SENSOR PLACEMENT
TABLE A-III

CALIBRATION OF AIR FLOW IN 242-B WIND TUNNEL

<table>
<thead>
<tr>
<th>Damper Setting</th>
<th>Counts</th>
<th>Time Min.</th>
<th>Counts/Min</th>
<th>cfm Sensor</th>
<th>cfm Duct</th>
<th>Air Velocity fpm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Closed</td>
<td>2817</td>
<td>10</td>
<td>281.7</td>
<td>27.32</td>
<td>529</td>
<td>316.9</td>
</tr>
<tr>
<td>1/8 Open</td>
<td>3429</td>
<td>10</td>
<td>343.9</td>
<td>32.44</td>
<td>1596</td>
<td>354.3</td>
</tr>
<tr>
<td>1/4 Open</td>
<td>11963</td>
<td>10</td>
<td>1196.3</td>
<td>116.04</td>
<td>2247</td>
<td>221</td>
</tr>
<tr>
<td>3/8 Open</td>
<td>15296</td>
<td>10</td>
<td>1529.6</td>
<td>148.36</td>
<td>2874</td>
<td>1178</td>
</tr>
<tr>
<td>1/2 Open</td>
<td>16969</td>
<td>10</td>
<td>1696.9</td>
<td>164.60</td>
<td>3186</td>
<td>1306</td>
</tr>
<tr>
<td>3/4 Open</td>
<td>17844</td>
<td>10</td>
<td>1784.4</td>
<td>173.09</td>
<td>3352</td>
<td>1374</td>
</tr>
<tr>
<td>Open</td>
<td>18049</td>
<td>10</td>
<td>1804.9</td>
<td>175.08</td>
<td>3990</td>
<td>1389</td>
</tr>
<tr>
<td>Open</td>
<td>18039</td>
<td>10</td>
<td>1803.9</td>
<td>174.98</td>
<td>3388</td>
<td>1388</td>
</tr>
</tbody>
</table>

\[
\text{fpm} \times \frac{\text{mi}}{5280 \text{ ft.}} \times \frac{60 \text{ min.}}{\text{hr.}} = 0.01136 \text{ mi per hr.}
\]

After HEPA filters at blower changed. \( \Delta p \) before change > 6 in H.G. \( \Delta p \) after change 1.6 in H.G.

<table>
<thead>
<tr>
<th>Setting</th>
<th>Counts</th>
<th>Time Min.</th>
<th>Counts/Min</th>
<th>cfm</th>
<th>1/min</th>
<th>1/min</th>
<th>1/min</th>
</tr>
</thead>
<tbody>
<tr>
<td>Open</td>
<td>22049</td>
<td>10</td>
<td>2204.9</td>
<td>211.8</td>
<td>4140</td>
<td>1697</td>
<td></td>
</tr>
<tr>
<td>1/2 Open</td>
<td>19526</td>
<td>10</td>
<td>1952.6</td>
<td>189.4</td>
<td>3668</td>
<td>1503</td>
<td></td>
</tr>
<tr>
<td>1/4 Open</td>
<td>12626</td>
<td>10</td>
<td>1262.6</td>
<td>122.5</td>
<td>2372</td>
<td>972</td>
<td></td>
</tr>
<tr>
<td>1/8 Open</td>
<td>8510</td>
<td>10</td>
<td>851.0</td>
<td>82.5</td>
<td>1598</td>
<td>655</td>
<td></td>
</tr>
<tr>
<td>Closed</td>
<td>3035</td>
<td>10</td>
<td>303.5</td>
<td>29.4</td>
<td>569</td>
<td>233</td>
<td></td>
</tr>
<tr>
<td>Open</td>
<td>22470</td>
<td>10</td>
<td>2247.0</td>
<td>218.0</td>
<td>4222</td>
<td>1730</td>
<td></td>
</tr>
<tr>
<td>Open</td>
<td>22501</td>
<td>10</td>
<td>2250.1</td>
<td>218.2</td>
<td>4225</td>
<td>1732</td>
<td></td>
</tr>
</tbody>
</table>

\[
\frac{2.44 \text{ ft}^2}{0.126 \text{ ft}^2} = 19.365
\]

Flow increase, low flow \( \frac{3035}{2817} = 107.7\% \)

Flow increase, high flow \( \frac{22340}{18044} = 123.8\% \)
FIGURE A-5. AIR VELOCITY IN 242-B BUILDING WIND TUNNEL VS DAMPER SETTINGS
APPENDIX B

EXPERIMENTAL OBSERVATIONS DURING INDIVIDUAL SHIPPING ACCIDENT EXPERIMENTS
EXPERIMENTAL OBSERVATIONS DURING INDIVIDUAL SHIPPING ACCIDENT EXPERIMENTS

SA-1 -- 58 grams of UO₂ powder were sprinkled over the 2.9 sq. ft. of soil in the flanged burner tray. Nominal air velocity in the furnace section was 23 mph. The air stream was sampled during a period of five hours with the filter collector and the cascade impactor.

SA-2 -- 96 grams of UO₂ powder were dispersed on the soil and air at a nominal velocity of less than 4 mph (see Appendix A) drawn over the surface for 6 hrs.

SA-3 -- The flanged burner tray was left in place upon completion of experiments SA-2, and one gallon of gasoline poured onto the surface. After the gasoline was ignited, air at a nominal velocity of less than 4 mph was drawn over the surface. The maximum recorded temperature in the duct was approximately 750°C with little increase of temperature of the soil or R.A.R.T. surface. Flames were visible in the furnace for around 40 minutes. Air flow and sampling were continued for a total time of 6 hours.

SA-4 -- 71 cc of a 0.51 g U/cc UNH solution and 1-5 Mi HNO₃ rinses were poured onto the soil held in the flanged burner tray. Air at a speed of less than 4 mph was drawn over the surface and sampled for 6 hours (part "a" of experiment). Airflow was stopped and all samplers replaced. After an additional 16 hours drying, airflow and sampling were resumed for 23-1/4 hours (part "b" of experiment).

SA-5 -- The flanged burner tray was left in place upon completion of experiment SA-4 and one gallon of gasoline was poured onto the surface. The gasoline extinguished itself with the violence of its ignition. After reignition, the exterior paint on the flash arrestor and inlet filter holder was smoking. Maximum temperatures of 950°C and 900°C were noted for the downstream and upstream thermocouple projecting 2 inches above the soil. Flames were visible for less than 10 minutes. A maximum air temperature exceeding 1000°C was observed. All HEPA filters over the inlet were checked and replaced although none appeared damaged sufficiently to impair performance. Air flow and sampling were continued for a total of 5-1/2 hours.

SA-6 -- 100 cc of a 0.51 g U/cc UNH solution and acid rinses of the graduated cylinder were poured over the soil held in the flanged burner tray. Air at a nominal speed of 23 mph was drawn across the surface and sampled for a total
of 27-3/4 hours (part "a" of the experiment). Airflow was stopped and all samplers replaced. Airflow and sampling were resumed for an additional 23-1/2 hours (part "b" of experiment).

SA-7 -- The flanged burner tray was left in place upon completion of experiment SA-6 and one gallon of gasoline was poured onto the surface. A considerable quantity of gasoline leaked out of the flanged opening (1/4 to 1/2 gallon). Gasoline ignited and air at a nominal speed of 23 mph was drawn across the surface. Flames were visible for less than 10 minutes and a maximum temperature was not recorded. Total airflow and sampling period was 6 hours.

SA-8 -- Rerun of conditions used in experiment SA-4. Sampling period part "a" was 23-3/4 hours and 24 hours for part "b".

SA-9 -- The flanged burner tray was left in place upon completion of experiment SA-8 and one gallon of gasoline poured onto the surface. After the gasoline was ignited, airflow at a nominal speed of less than 4 mph was drawn over the surface and sampling was initiated. Flames were visible in the furnace for some 58 minutes with a maximum recorded temperature of 650°C. Total airflow and sampling time was 5 hours.

SA-10 -- 93 cc of a 0.51 g U/cc UNH solution was poured onto the soil held in the flanged burner tray. One gallon of gasoline was poured onto the surface and ignited. Air at a nominal speed of less than 4 mph was drawn over the burning surface and sampling was initiated. Gasoline did not appear to burn as vigorously as in previous experiments. Flames were visible in the furnace for approximately 25 minutes with a maximum recorded temperature of 200°C. Total airflow and sampling period was 5-1/2 hours.

The next group of experiments involved the fractional airborne release from ground having a vegetation cover. The flanged burner tray was filled with soil collected from around the 242-B Building. Young, green tumbleweed plants common in the area were up-rooted and transplanted in the soil held in the flanged burner tray.

SA-11 -- 89 grams of UO₂ powder were sprinkled over the vegetation-covered surface of the flanged burner tray which was then inserted in the furnace. Air at a nominal speed of less than 4 mph was drawn across the surface for 23-1/2 hours and sampled.
SA-12 -- The flanged burner tray was left in place at the completion of experiment SA-11 and a gallon of gasoline poured onto the surface. The gasoline was ignited and airflow plus sampling resumed. Flames were visible in the furnace for 50 minutes with a maximum recorded temperature of 550°C. Total airflow and sampling period was 6 hours.

SA-11 -- 79 grams of UO₂ powder were dispersed over the 2.9 sq. ft. of vegetation-covered surface of the flanged burner tray. The tray was inserted into the furnace and air at a nominal velocity of 23 mph was drawn over the surface for 24 hours. Total sampling period was also 24 hours.

SA-14 -- After the completion of experiment SA-13, the samplers were replaced but the flanged burner tray was left in place. A gallon of gasoline was poured onto the surface and ignited. Airflow and sampling were resumed. Flames were visible in the furnace for only 7 minutes with a maximum recorded temperature of 600°C. Total airflow and sampling period was 6 hours.

SA-13 -- 100 cc of a 0.51 g U/cc UNH solution was poured onto the vegetation-covered soil surface held in the flanged burner tray and the tray inserted into the furnace. A gallon of gasoline was added. A force sufficient to extinguish the flames was generated during the ignition. The material was reignited but the vegetation did not burn well -- only the downstream 1/3 of the vegetation burned. Attempts to spread the flames were unsuccessful. Air at a nominal velocity of 23 mph was drawn across the surface. Visible burning continued for 40 minutes. The total period of airflow and sampling was 6 hours.

SA-16 -- 100 cc of a 0.51 g U/cc UNH solution was poured onto the 2.9 sq. ft. vegetation-covered surface held in the flanged burner tray and the tray inserted into the furnace section. A gallon of gasoline was poured onto the surface and ignited. Flames were visible over the entire surface. Air at a nominal velocity of less than 4 mph was drawn across the surface and sampled. Observable burning persisted for 125 minutes. Airflow and sampling were continued for a period of 6 hours.

The next group of engineering scale experiments involved the fractional release of uranium from a stainless steel surface during a petroleum fire. The flanged burner tray was partially filled with soil collected from around the 242-B Building. A stainless steel pan 15 inches ID by 2 inches deep was embedded in the upstream end of the flanged burner tray and additional soil added to make the rim of the pan flush with upper edge of the rim of the flanged burner tray.
SA-17 — 22 grams of uranium dioxide powder were placed in the stainless steel pan held in the flanged burner tray and inserted in the furnace section. A gallon of gasoline was poured into the dish and ignited. Air at a nominal velocity of less than 4 mph was drawn over the surface and sampled. Flow rates on both filter samples could not be maintained and decreased to 0.25 and 0.75 cfm respectively for the fixed and movable filters. The RART surface temperature increased during the experiment reaching approximately 70°C after 7 minutes of burning. The water spray was turned on to cool the air. After 9 minutes, no flames were visible and the airflow and sampling halted (part "a" of the experiment). The samplers were replaced. Airflow and sampling were resumed for an additional 4.77 hours to measure the aerodynamic entrainment of uranium from the fire residue (part "b" of the experiment).

SA-18 — 50 cc of a 0.51 g U/cc UNH solution were poured onto the stainless steel pan held in the flanged burner tray and inserted in the furnace. A gallon of gasoline was poured into the dish and ignited. Air at a nominal speed of 23 mph was drawn over the surface and sampled. Flow through the filter samples decreased during the course of the experiment to 0.67 and 0.9 cfm for the fixed and movable samplers respectively. Airflow and sampling were halted in 9 minutes when burning ceased (part "a" of experiment). After the samplers were replaced, airflow and sampling were resumed for an additional 5 hours (part "b" of experiment).

SA-19 — 50 cc of a 0.51 g U/cc UNH solution was poured into the stainless steel dish in the flanged burner tray and inserted in the furnace. A gallon of gasoline was poured into the dish and ignited. Air at a nominal speed of less than 4 mph was drawn across the surface. Flowrates through the filter samples decreased to 0.67 and 0.3 cfm through the fixed and movable samplers respectively. The water spray within the R.A.R.T. was initiated after 6 minutes of burning to cool the air. The fire went out after 13 minutes, and airflow and sampling were halted. After the samplers were replaced, airflow and sampling were continued for an additional 6 hours.

The last two experiments in the series involved the fractional release of uranium from a "road-like" (gravel/asphalt mixture) surface. The gravel/asphalt mixture was poured into a 15-inch diameter by 4-inch deep steel pan and heated. The material was tamped to produce a smooth bed approximately 1 inch deep in the bottom of the pan. The 4 inch lip of the pan rested upon the rim of the
flanged burner tray creating a continuous smooth surface with the bottom of the furnace section.

SA-20 -- 50 cc of a 0.51 g U/cc UNH solution was poured onto the road-like surface and inserted into the furnace. A gallon of gasoline was poured onto the surface and ignited. Air at a nominal speed of less than 4 mph was drawn over the surface and sampled. A large amount of soot was generated during the burning and filter sample flowrates dropped to 0.4 and less than 0.1 cfm for the fixed and movable samplers respectively. Sampling was terminated in approximately 17 minutes due to the lack of flow through the samplers although the gasoline-surface mixture burned for 36 minutes. Cooling water spray was initiated after 5 minutes of burning. Airflow was halted after the fire was extinguished and all samplers were replaced (part "a" of the experiment). Airflow and sampling were then continued for 5 hours to measure aerodynamic entrainment of the uranium from the fire residue (part "b" of the experiment).

SA-21 -- 66 grams of uranium dioxide powder were sprinkled over a road-like surface and placed into the furnace. A gallon of gasoline was poured onto the surface and ignited. Air at a nominal speed of less than 4 mph was drawn over the surface and sampled. Cooling water spray was instituted after 5 minutes of burning. Flow through the fixed and movable filters decreased to 0.42 and 0.45 cfm during the course of the burning. Flames were no longer visible after 22 minutes and airflow and sampling were discontinued (part "a" of the experiment). After the samplers were replaced, airflow and sampling were resumed for an additional 6 hours.

Upon evaluation of the data from the initial series of experiments (SA-1 to SA-21) additional data in certain areas appeared desirable. Eleven additional experiments were performed to measure the aerodynamic resuspension and fractional release during a fire of uranium dioxide powder from bare, sandy soil and stainless steel and uranium nitrate solution from bare, sandy soil at air speeds not covered in previous experiments.

SA-22 -- 56.8 grams of uranium dioxide powder were sprinkled on a smooth surface of sandy soil held in the burner tray. The tray was inserted into the wind tunnel and air at a nominal speed of 2.5 mph was passed over the surface for 24 hours. The airborne concentration was continuously sampled with filters in the airstream and size distribution of the uranium airborne evaluated
the air drawn over the surface at a speed of 20 mph during this period (part "a" of experiment). Aerodynamic resuspension of uranium from the solid residue at 20 mph evaluated by sampling over the next 24 hours (part "b" of the experiment).
from periodic cascade impactor samples (part "a" of experiment). The burner
tray bearing the material was left in place and a gallon of gasoline was poured
onto the surface. The gasoline ignited vigorously and burned for 60 minutes.
Both filter and cascade impactor sample were taken during the entire period
although flow through the filter was reduced to 0.85 cfm during the final 30
minutes. Upon completion of the burning (part "b" of experiment), airflow was
stopped and the samplers replaced. Airflow was resumed and resuspension of
uranium from the residue from the fire was measured during the subsequent 24 hr
period (part "c" of experiment).

SA-21 — 56.8 grams of uranium dioxide powder were sprinkled on a smooth sur-
face of sandy soil held in the burner tray and experiment SA-22 repeated using
an air speed of 20 mph. Under these conditions, the gasoline burned with a
visible flame for 18 minutes. Some sandy soil was found on the initial sieve
of the cascade impactor after this portion of the experiment.

SA-25 — 56.3 grams of uranium dioxide powder were sprinkled into a 15-inch
diameter stainless steel pan embedded in sandy soil held in the flanged burner
tray. Aerodynamic resuspension by air at a nominal velocity of 20 mph was
measured by continuously sampling of the air over a 24-hour period. Periodic
cascade impactor samples were taken to evaluate the size distribution of the
uranium airborne (part "a" of experiment). A gallon of gasoline was then poured
onto the surface and ignited. The gasoline burned with a visible flame for
18 minutes and the fractional release and size distribution of the uranium air-
borne evaluated from filter and cascade impactor samples taken during this
period (part "b" of experiment). After the samplers were replaced, aerodynamic
resuspension of uranium from the solid residue remaining after the fire was de-
terminated by collection of airborne particles during a 24 hour period. Examin-
ation of the stainless steel pan after the experiment indicated the uranium
dioxide powder was washed towards the edges of the pan during the introduction
of the gasoline.

SA-26 — 100 ml of the 0.51 g uranium per ml solution of uranium dioxide in di-
lute nitric acid plus 3 dilute nitric acid rinses of the graduated cylinder
were poured on a smooth surface of sandy soil held in the flanged burner tray.
The tray was inserted into the wind tunnel and a gallon of gasoline was poured
onto the surface. The gasoline ignited vigorously and, under these conditions,
burned for 8 minutes. Both filter and cascade impactor sample were taken of
APPENDIX C

EVALUATION OF SAMPLING SYSTEM -- INLET DUCT R.A.R.T. 242-B BUILDING
EVALUATION OF SAMPLING SYSTEM -- INLET DUCT R.A.R.T. 242-B BUILDING

Samples to evaluate the concentration and size distribution of particles airborne during the shipping accident experiments were collected by two filters and a cascade impactor sampler located at the downstream end of the inlet duct of the R.A.R.T. Figure C-1 is a representation of a view of this arrangement looking downstream towards the R.A.R.T. A 1/2-inch Swagelok male connector enlarged to allow passage of a 1/4 ODT was attached to the 3-inch flanged opening located on the top of the duct. An open face 2-inch filter holder was attached to a 1/2-inch diameter stainless steel tube with a 90° bend which passed through the Swagelock fitting. A vertical traverse of the duct could be made with the filter holder by loosening the Swagelock fitting and manipulating the tube. The depth of penetration of the centerline of the filter could be determined by marking on the tube. A second open face 2-inch filter holder was inserted via the 3-inch flanged opening located on the side of the duct. The position of this holder was fixed at the centerline of the duct. Samples for the cascade impactor were drawn through a stainless steel 1/2-inch tube with a 90° bend and inserted into the bottom of the duct through a 3-inch diameter pipe half-coupling. The opening of the tube was approximately 2 inches from the floor of the duct.

A series of tests was performed to ascertain if a representative sample of the concentration of airborne particles could be obtained by such an arrangement. Rhodamine particles were generated into the airstream by nebulizing a dilute aqueous solution (0.01 g rhodamine per ml) from various positions in the furnace section of the inlet duct to the R.A.R.T. while drawing air through the duct at velocities of 2.5 and 20 mph. The locations used for generation are designated in Figure C-2. In all cases the rhodamine particles were generated with the nozzle of the nebulizer pointing downstream on approximately the centerline of the duct.

Samples were taken to measure both the quantity of rhodamine airborne at the end of the duct and deposited on the ducts. Airborne concentrations were evaluated by samples collected on both the fixed and vertical traversing filters. Cascade impactor samples were taken to measure the size distribution of the particles airborne. Particles falling to the floor of the duct were collected on an 18-inch wide sheet of aluminum foil which extended from the
FIGURE C-1. WIND TUNNEL SAMPLING SYSTEM
furnace section to the downstream end of the duct. 12-inch by 12-inch sheets of aluminum foil were taped in 5 places at mid-height on each wall to measure the quantity deposited on the wall.

The quantity of rhodamine in various samples was determined by measuring the fluorescence from the aqueous sample in a calibrated fluorimeter. The simplest and most efficient means of removing the rhodamine from the various sample matrices — plastic nebulizer parts, glass fiber filters, glass impactor collection plates, aluminum foil and impactor parts — was to soak the item in distilled water with periodic agitation. Quantitative recovery of standard quantities of rhodamine dried on the material were obtained using this procedure for all matrices tested.

A total of seven tests was performed. The operating conditions used and results are tabulated in Table C-I. Rhodamine particles were generated for periods ranging from 30 to 80 minutes and were generally limited by the capacity of the nebulizer. The quantity generated was assumed to be the difference between the amount placed in the nebulizer (concentration times volume of liquid) and the amount found in the nebulizer at the end of the test. Some difficulties were encountered in the initial experiments (1 and 2) with recovering the rhodamine from the nebulizer and the quantity listed as generated is probably in error.

The quantity airborne as measured by the "fixed" and "movable" (vertical traverse) filter samples are generally in good agreement. The exceptions are run #6 when the "movable" filter was not traversed and run #5. In all but one instance, the quantity airborne calculated by summation of the rhodamine collected on the impactor stages is higher than measured on the filters indicating some channeling of the particles. The effect is not as pronounced when particles are generated in the center of the furnace section (point B) or elevated from the floor of the duct (point C). In the shipping accident experiments, the particles are released from the furnace section (not in the duct) and must be slightly elevated to become airborne.

The amounts of rhodamine found on the floor and walls was insignificant. Considerably greater amounts were found on the floors as anticipated. But a significant portion of the material found on the floor was found in the first few feet and may have resulted from direct impingement of liquid droplets. Thus, the sampling arrangement was found adequate to obtain fairly represen-
TABLE C-1
EVALUATION OF INLET DUCT SAMPLING SYSTEM - OPERATING PARAMETERS AND RESULTS

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Generation Point</strong></td>
<td>A</td>
<td>A</td>
<td>B</td>
<td>B</td>
<td>A</td>
<td>A</td>
<td>C</td>
</tr>
<tr>
<td><strong>Air Speed, mph</strong></td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
</tr>
<tr>
<td><strong>Length of Time Generated, Min.</strong></td>
<td>30</td>
<td>80</td>
<td>60</td>
<td>70</td>
<td>66</td>
<td>4</td>
<td>69</td>
</tr>
<tr>
<td><strong>g * rhd Used</strong></td>
<td>1.0</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td><strong>g * rhd in Neb. at End of Test</strong></td>
<td>0.69</td>
<td>0.41</td>
<td>0.104</td>
<td>0.166</td>
<td>0.22</td>
<td>0.15</td>
<td>0.18</td>
</tr>
<tr>
<td><strong>Percent rhd Generated</strong></td>
<td>0.31</td>
<td>0.09</td>
<td>0.392</td>
<td>0.334</td>
<td>0.28</td>
<td>0.35</td>
<td>0.32</td>
</tr>
<tr>
<td><strong>Percent rhd Recovered</strong></td>
<td>38</td>
<td>259</td>
<td>129</td>
<td>68.4</td>
<td>76.1</td>
<td>72.0</td>
<td>123</td>
</tr>
<tr>
<td><strong>Median Mass Diameter of Particle Airborne, microns</strong></td>
<td>0.72</td>
<td>0.19</td>
<td>1.4</td>
<td>2.4</td>
<td>2.4</td>
<td>2.4</td>
<td>2.2</td>
</tr>
<tr>
<td><strong>g * rhd Airborne, Fixed Filter</strong></td>
<td>0.091</td>
<td>0.168</td>
<td>0.364</td>
<td>0.205</td>
<td>0.154</td>
<td>0.20</td>
<td>0.34</td>
</tr>
<tr>
<td><strong>g * rhd Airborne, Movable Filter</strong></td>
<td>0.91</td>
<td>0.171</td>
<td>0.342</td>
<td>0.205</td>
<td>0.094</td>
<td>0.039**</td>
<td>0.43</td>
</tr>
<tr>
<td><strong>g * rhd Airborne, Impactor</strong></td>
<td>0.15</td>
<td>0.34</td>
<td>0.0019</td>
<td>0.266</td>
<td>0.377</td>
<td>0.47</td>
<td>0.41</td>
</tr>
<tr>
<td><strong>r * rhd Airborne, South Wall</strong></td>
<td>$1.5 \times 10^{-5}$</td>
<td>$2.4 \times 10^{-7}$</td>
<td>$9.6 \times 10^{-7}$</td>
<td>$9.6 \times 10^{-6}$</td>
<td>$1.3 \times 10^{-5}$</td>
<td>$9.2 \times 10^{-5}$</td>
<td>$5.2 \times 10^{-5}$</td>
</tr>
<tr>
<td><strong>r * rhd Airborne, North Wall</strong></td>
<td>$3.8 \times 10^{-5}$</td>
<td>$2.3 \times 10^{-7}$</td>
<td>$1.9 \times 10^{-6}$</td>
<td>$2.3 \times 10^{-5}$</td>
<td>$2.4 \times 10^{-5}$</td>
<td>$2.0 \times 10^{-5}$</td>
<td>$2.2 \times 10^{-5}$</td>
</tr>
<tr>
<td><strong>r * rhd Airborne, Floor</strong></td>
<td>$6.6 \times 10^{-5}$</td>
<td>$6.4 \times 10^{-7}$</td>
<td>$2.1 \times 10^{-7}$</td>
<td>$3.4 \times 10^{-3}$</td>
<td>$5.2 \times 10^{-3}$</td>
<td>$1.4 \times 10^{-3}$</td>
<td>$1.1 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

* Difference between amount rhodamine used and found in inlet air following experiments.
** No traverse, filter held at topmost position during entire experiment.
+ Based on arithmetic average of quantity measured on filter and cascade impactor samples.
* Amount collected on eight stages and back-up filter.
tative samples of the airborne concentration. In most cases, the "fixed" filter result can be used as an overall average since it agrees with the "movable" filter in most cases. The size distributions determined by the cascade impactor samples are probably accurate. After the initial runs, the Median Mass Diameter of the particles airborne remains relatively stable even in experiments where the airborne concentrations calculated from cascade impactor results is significantly higher. Thus the high concentrations are attributed to a "channeling" effect, not settling of particles.