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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1972
TO THE USAEC DIVISION OF BIOMEDICAL
AND ENVIRONMENTAL RESEARCH
VOLUME II: PHYSICAL SCIENCES
PART 1. ATMOSPHERIC SCIENCES

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

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By

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

April 1973

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

Volume I of this report to the USAEC Division of Biomedical and Environmental Research, covering work in the Biological and Ecological Sciences, is issued as BNWL-1750, 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-1751.

FOREWORD

The environmental considerations associated with energy production is one of the major problem areas of concern to the AEC and, indeed, to the country. Consequently, it is important to understand the processes which transport, dilute and deposit energy associated pollutants in the atmospheric boundary layer as well as on regional, continental, and global scales. It is also essential to understand the cloud physics and microphysical processes which contribute to scavenging of pollutants by cloud and rain and which, in response to interaction with pollutants, can contribute to inadvertent modification of the weather. The research conducted within the Atmospheric Sciences Program at Battelle is directed toward a balanced investigation of these processes and the contributions included in this report address the various aspects of this research.

During the past year diffusion, deposition, and turbulence work was broadened to consider wider applications of models to various terrain and surface types and a wider range of conditions, including near calm winds, which must be considered as nuclear power reactor siting extends throughout the country. Also, the deposition of particulates is being studied in experimental and modeling efforts to account for this very significant but, as yet, ill-defined effect on air concentrations and surface distributions of contaminants.

The resuspension of surface contamination, particularly in connection with plutonium in the environment, is a subject receiving increased attention in our research. This problem is of importance not only in connection with existing AEC facilities utilizing plutonium, but future breeder power reactors and fuel fabricating and reprocessing facilities must also be considered. The escape of plutonium to the environment presents a continuing potential inhalation hazard resulting from resuspension of particulate material from the surface into the air by wind forces. An initial evaluation and identification of significant factors were possible using Rocky Flats, Colorado, data from plutonium sampling in air and soil and existing meteorological records from surrounding areas. Controlled tracer experiments have also been designed and set up at Hanford to study resuspension. Detailed definition of the distributions of resuspended soil particles which act as hosts for deposited contaminants has also begun and is discussed in this report.

On regional scales, the removal of pollutants by wet scavenging processes is of particular importance. The intrusion of pollutants into cloud and storm systems and their eventual rainout, as well as below-cloud scavenging, is very significant in determining regional levels of pollutants in the air and their distribution over the surface. Scavenging experiments have been conducted in both frontal storm and convective cloud systems and are providing insights and information necessary for modeling these processes.

Initial development of research aircraft capabilities with unique tracer generating and aerosol measuring equipment was carried out for extending research on energy related environmental problems to greater heights and distance scales of importance. The aircraft was used in METROMEX for inserting tracers into convective storms in precipitation scavenging studies and for defining aerosol distributions upwind and downwind of the urban area.

Another problem area, that of defining and explaining the large scale continental and global distribution of contaminants, was initially aimed at distinct nuclear testing events. While concern for studying the spectrum of radionuclides from this type of source remains, the identification of effluents from the increasing number of nuclear power plants throughout the world, their transport, and variations in their atmospheric background levels should be studied. Similarly, effluents from other power production sources need to be concurrently studied from the same point of view. Extension of continental scale studies in these latter directions has recently begun and initial results are reported here.

The research carried out this past year, and the reports included in this volume, are aimed at more clearly focusing on AEC needs and priorities. The growing significance of environmental aspects associated with nuclear power requires that our research give continued and increased attention to identifying and solving the most pertinent problems.

C. E. Elderkin, Associate Manager
Atmospheric Sciences Department

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INFLUENCE OF SOIL RESUSPENSION ON THE AIRBORNE
PARTICLE SIZE DISTRIBUTION

G. A. Sehmel and F. D. Lloyd

The airborne particle size distribution function of resuspended soil has been determined as a function of particle diameters from 1 to 230 μ m and sampling heights from 0.3 to 30 m. An apparent lower limit is determined for the size distribution function. Soil resuspension can cause relatively high soil injection rates into the atmosphere which increase the magnitude of the size distribution function above this lower limit. The soil resuspension rates are a direct function of the hours of high wind speed occurring during dusty time periods. However, the resuspension rates must also be a function of the persistence of these high winds since less resuspension occurred for the same total hours of high winds when these hours were accumulated over a longer time period.

Particle resuspension of potentially hazardous trace level materials can produce an immediate downwind inhalation hazard if the hazardous material such as plutonium is in the respirable size range. We do not know the fraction of particles resuspended which are unattached tracer particles and the fraction of this tracer which is attached to host soil particles. We do however believe that a large portion of the hazardous tracer can be attached to resuspended soil particles. Consequently, we have initially tested our resuspension sampling equipment by experimentally measuring the airborne soil size distribution function during visibly dusty and nondusty time periods.

The airborne soil particle size distribution functions were determined during four selected dry time

periods of from 32 to 570 hours duration during the spring and summer in a sparsely vegetated region on the Hanford reservation. The measurements were made in a reasonably flat area where a range fire had burned the grass and small vegetation the previous year. The prevailing westerly winds blew across the sampling site toward the nearest highway which was over a mile distant. Upwind, the terrain was undisturbed and unpopulated for several miles and included a region without sagebrush extending about 200 feet upwind of the sampling tower.

Both wind instrumentation and hi-volume air samplers* were mounted at six levels of the sampling tower shown in Figure 1. The wind instrumentation included integrating anemometers at the lower heights and an aerovane at the 32 m height to continuously record wind speed, v , and wind direction, θ . The details of

* See "Evaluation of Hi-Volume Cascade Impactors," this report.

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FIGURE 1. Particle and Meteorological Sampling Tower

analyzing the airborne soil samples and wind data have been reported.⁽¹⁾

The airborne size distribution function n_d is the number of particles per cm^3 per unit μm particle diameter. The measured airborne soil size distribution functions are shown in Figure 2 for succeeding air sampling periods of 32, 38, 430, and 570 hours. The first two short sampling periods were periodically dusty while the last two longer periods were not visibly dusty.

The lower limits of the size distribution functions are represented by line A,

$$n_d = 6.6(D)^{-3.88}, \quad (1)$$

and line B,

$$n_d = 2200(D)^{-5.48}. \quad (2)$$

These lines are considered⁽¹⁾ to approximate the limiting background aerosol size distribution function when large quantities of resuspended soil are not injected into the atmosphere.

When significant local soil resuspension occurs, the size distribution function is increased above the limiting background aerosol lines. This increase is shown for both the periodically dusty periods of March and April. As expected, the increase in n_d is greatest at the 0.3 m sampling height.

The size distribution functions increased with an increase in the number of hours of high winds during the sampling period. This increase is shown by analysis and comparison of the April and March sampling periods. There were only 5 hours of average wind speed greater than 8 m/sec in the March sampling period. However, the winds increased to a total of 10 hours of wind speed greater than 8 m/sec in the April sampling period. As shown in Figure 2, the increased winds in April do produce relatively greater size distribution functions.

This apparent simple increase in n_d with an increase in hours of high winds is more complex when we compare the n_d 's obtained during the spring to those n_d 's obtained during the summer. The last three time periods

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FIGURE 2. Airborne Particle Size Distributions

all had 10 hours of accumulated wind speed greater than 8 m/sec, but the summer time periods did not show any large injection of particles above lines A and B. Consequently, we must presently conclude that the differences in the spring and summer n_d 's must be explained either by (1) accumulated hours of high wind speed are more efficient in causing soil resuspension when the accumulated hours

are nearly consecutive (accompanied by high gustiness and turbulence) rather than accumulated over a long time interval, or by (2) vegetation which had grown into the test site hindered the high winds from causing soil resuspension during the summer time periods.

The April data are the more consistent in showing the decrease in the size distribution function, n_d .

Consequently in Figure 3 these data have been converted to show the changes with height in the volume distribution $\Delta V/\Delta \ln D$. Line A shows that the volume distribution increases with an increase in particle diameter. This increase means that, even though the size distribution function, n_d , is decreasing rapidly, the relatively greater increase in particle volume more than compensates for the decrease in n_d . Line A intersects line B at a particle diameter of 9 μm . For this limiting size distribution of larger particles represented by line B, a decreasing volume distribution is indicated with an increase in particle diameter. The volume distributions indicated by the curves through the data are greater than either the background lines of A or B since local resuspension was high during this dusty sampling period. The maxima in these curves are probably the second mode in the bi-modal distributions that have been summarized by Whitby, et al. (2)

FUTURE RESEARCH

We have demonstrated that resuspended airborne soil particles can be adequately sampled as a function of particle diameter and height. We now need to determine airborne soil distribution functions as a function of wind speed. Our research has developed a model* to predict plutonium resuspension at one site as a function of hours of each wind speed. We

* See "Resuspension by Wind at Rocky Flats," this report.

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FIGURE 3. Volume Distribution of Airborne Soil

now need to determine a better model for predicting particle resuspension. By mass budgets using both tracer and soil particles, we will experimentally determine the data base from which to develop a general resuspension model for predicting particle resuspension as a function of surface type, tracer particle diameter, and meteorological parameters such as hours of wind speed.

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2. K. T. Whitby, R. B. Husar, and B. Y. H. Liu, "The Aerosol Size Distributions of Los Angeles Smog," *J. Colloid. Interface Sci.*, vol. 39, no. 1, pp. 177-204, April, 1972.

INITIAL RESUSPENSION MODELS

W. G. N. Slinn

A progress report is presented of efforts made to understand the resuspension of aerosol particles from the earth's surface. It is seen that for $u_* \gtrsim 20$ cm sec⁻¹, horizontal wind fluctuations can dislodge most sub-millimeter particles from dry soil. This suggests that the soil particle size distribution is the controlling factor of the airborne particle size distribution in the near-surface layer. Above the surface layer, it is shown that the convective diffusion equation is incapable of explaining experimental data. A statistical approach based on the probability distribution of vertical wind speeds is explored and it is shown that observed wind speed data does correctly describe the size distribution of large, resuspended particles. Directions are indicated for future theoretical and experimental studies.

INTRODUCTION

Resuspension is familiar to anyone who has blown dust from his desk. Interest in resuspension is topical for the evaluation of potential hazards from noxious material which might be spilled. In this report, resuspension is considered in the following subject order:

- I. MOTION OF SINGLE PARTICLES
 - A. Individual "Large" Particles
 1. Dry Soil

2. Wet Soil
 - B. "Large" versus "Small" Particles
 - C. Individual "Small" Particles
- II. POLYDISPERSED PARTICLES
 - A. Ballistic Effect
 - B. Statistical Considerations
 - C. The Soil Particle Size Distribution
- III. RESUSPENSION
 - A. Diffusion Approach
 - B. Statistical Approach
- IV. CONCLUSIONS

I. MOTION OF SINGLE PARTICLES

A. INDIVIDUAL "LARGE" PARTICLES

1. Dry Soil

For an individual large particle protruding above other neighboring particles, the onset of motion should depend on the dynamic pressure on the windward side of the particle, on the torque resulting from the drag force on the top of the particle, and on the lift force resulting from the reduced pressure at the top of the particle. Dimensional analysis tells us that, for particle Reynolds number smaller than unity, all these forces are proportional to the dynamic pressure $1/2 \rho_a u^2$ multiplied by a characteristic area of the particle, where ρ_a is the density of the air and u is the mean velocity within the roughness region of the boundary layer. For particles larger than the viscous sub-layer, we take the velocity at height r to be given by⁽¹⁾

$$u = u_* \left[2.40 \ln\left(\frac{ru_*}{\nu}\right) + 5.84 \right] \quad , \quad (1)$$

where $u_* = (\tau/\rho_a)^{1/2}$ is the friction velocity in which τ is the shear stress transmitted to the surface by the air and $\nu \approx 0.15 \text{ cm}^2 \text{ sec}^{-1}$ is the kinematic viscosity.

Motion of particles larger than the viscous sub-layer should then occur when the applied forces overcome the forces and resulting torques caused by gravity. Thus for resuspension to occur, it seems that u_* must be great enough to satisfy

$$1/2 \rho_a u_*^2 4\pi r^2 = \epsilon 4/3 \pi r^3 \rho_p g \quad , \quad (2)$$

where ρ_p is the density of the particle and ϵ is an "ignorance" parameter which contains all the factors that we have neglected. Equation (2) fits the experimental data⁽²⁾ well for particles whose radius is larger than about 50μ . This data was obtained by measuring the resuspension of monodispersed quartz particles. In the range indicated, the data can be fit with the expression

$$u_* = 30 \frac{\text{cm}}{\text{sec}} \left(\frac{r}{200\mu} \right)^{1/2} \quad , \quad (3)$$

which corresponds to Equation (2), using $\rho_p = 2.6 \text{ gm cm}^{-3}$ for quartz, provided $\epsilon = 1.14$.

2. Wet Soil

For an individual "large" and wettable particle attached to neighboring particles by a water film, it is easy to see that the water surface tension force is vastly larger than the gravitational force for all particles of interest. The forces are equal when

$$4/3 \pi r^3 \rho_p g \approx \sigma 2\pi r \quad ,$$

where $\sigma = 73.5 \text{ dynes cm}^{-1}$ at 20°C ; i.e., the forces are equal when $r \approx 1 \text{ mm}$. For a 100μ particle completely surrounded by a water film, the surface tension force is about 10^2 times larger than the gravitational force. The consequences of the above to the magnitude of u_* required to resuspend a wetted particle are that the critical u_* would be proportional to $r^{-1/2}$ and, more importantly, that resuspension will not occur until all particles are essentially dry.

B. "LARGE" VERSUS "SMALL" PARTICLES

The above considerations for large, dry particles would not be appropriate if the particles were buried within the viscous sub-layer. The characteristic height of this layer, in which viscous forces dominate the velocity profile, is well known to be $0(u/u_*)$, where u is the kinematic viscosity of the air. Thus the distinction between large and small particles occurs for a critical particle size of $2r_c \approx u/u_*$, which for $u_* = 15 \text{ cm sec}^{-1}$ yields $2r_c \approx 100 \mu$. This is the size from which we make the distinction of "large" versus "small" particles.

C. INDIVIDUAL "SMALL" PARTICLES

For a particle buried in the viscous sub-layer, we assume that the dynamic pressure is developed by a velocity which is only the fraction (r/r_c) of the velocity at the top of the sub-layer. Then Equation (2) becomes

$$\begin{aligned} \frac{1}{2} \rho_a \left(\frac{r}{r_c} 5.84 u_* \right)^2 4\pi r^2 \\ = \delta \frac{4}{3} \pi r^3 \rho_p g \quad , \quad (4) \end{aligned}$$

where δ is another "ignorance" parameter. Bagnold's data⁽²⁾ can be fit well by the expression

$$u_* = 28 \frac{\text{cm}}{\text{sec}} \left(\frac{5\mu}{r} \right)^{1/4} \quad , \quad (5)$$

which corresponds to Equation (4) with $\delta = 4/3$.

II. POLYDISPERSED PARTICLES

A. BALLISTIC EFFECT

The above considerations were restricted to the case of a monodispersed aerosol particle size distribution. We now consider the case of polydispersed particles. It was seen above that, even when interparticle forces are ignored, as the particle size decreases, motion becomes increasingly more difficult. However, this difficulty can be somewhat overcome by the ballistic effect discussed by Bagnold:⁽²⁾

"If the dust deposit contains, as it usually does, an appreciable proportion of coarse grains of ballistic size, i.e., larger than 100 to 200 μ , it will be very easily disturbed by direct wind action. And if at the same time it contains a considerable proportion of very fine grains, an initial disturbance of the easier-moved coarse grades by a wind will result in the fine grades being dispersed upwards in resuspension, in spite of a reasonable degree of cohesion. For not only will the fine grain surface be pitted by the bombardment of coarse grains, but the bombardment is likely to break the cohesive bonds."

B. STATISTICAL CONSIDERATIONS

In the above, turbulent fluctuations have been essentially ignored, except insofar as they dictated the mean wind profile. What we seek now is to relate the probability density function (pdf) for the fluctuations of the longitudinal component of the wind to the pdf which describes the size distribution of the particles

which are dislodged from the surface. Since gravitational settling of the particles will be ignored in this section, the results cannot be expected to be valid at significant heights above the surface.

Unfortunately, the statistical properties of turbulent flow are not known well. Lumley and Panofsky⁽³⁾ suggest that for neutral condition, the variance of u , σ_u^2 , is approximately $(u_*/k)^2$, where k is Von Karman's constant. If we assume that the probability density function is Gaussian, then we have in the forced convection layer near the surface

$$P(u) = \frac{k}{(2\pi)^{1/2} u_*} \exp - \left[\frac{(u - \bar{u})^2}{2u_*^2/k^2} \right], \quad (6)$$

where $\bar{u}$ is given by Equation (1). From (2) we have that a "large" particle will be dislodged if $|u| \geq G^{1/2} r^{1/2}$, where

$$G = 2/3 \epsilon \frac{\rho_p}{\rho_a} g. \quad (7)$$

If the contribution to the motion of large particles that occurs from negative values of u can be ignored, then we have that the distribution of large particles at low elevations will be the integral of $P(u)$ from $G^{1/2} r^{1/2}$ to all larger speeds. Thus

$$f_o(r) = 1/2 \operatorname{erfc} \left[\frac{G^{1/2} r^{1/2} - \bar{u}(r)}{2^{1/2} u_*/k} \right]. \quad (8)$$

There are, of course, other approaches that could be taken. For example, evidence is accumulating⁽⁴⁾ that there is a "long-tail" to $P(u)$, which can be fit fairly well with an $\exp(-cu)$ rather than a Gaussian.

Higher probabilities for large values of u would be thought to be especially significant to resuspension. Also, we could justifiably treat Equation (8) as a conditional probability, depending on a specific u_* , and then discuss the pdf of u_* . However, we can easily see that (8) contains sufficient information for resuspension estimates.

Equation (8) is plotted in Figure 1 for $u_* = 20 \text{ cm sec}^{-1}$, $\rho_p = 2.66$, and $\epsilon = 1.3$. In addition are plotted some data taken by Sehmel* at a height of 0.3 m, during a dust storm and by Slinn** at a height of 2 m, in the updraft of a cumulonimbus cloud. In addition, for interest sake, is shown the size distribution of aged Sahara dust, taken at a large distance from the desert.⁽⁵⁾ The point to be gained from Figure 1 is that for "large" particles, and probably also for "small" particles if the ballistic effect is evaluated, the particle size distribution of resuspended particles is not significantly influenced by the capability of the wind to dislodge the particles from the surface. Thus from Figure 1, for u_* only equal to 20 cm sec^{-1} , essentially all particles smaller than 100μ will be mobilized. Of course, that a particle is dislodged from the surface does not mean it will be transported to a significant extent vertically, but the point here

* See "Influence of Soil Resuspension on the Airborne Particle Size Distribution" in this report.

** See "Some Measurements of Atmospheric Aerosol Particles" in this report.

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FIGURE 1. Particle Number Distributions. Rightmost curve shows the size distribution which turbulent winds with $u_* = 20 \text{ cm sec}^{-1}$ could dislodge from the surface. For comparison, some measured, resuspended particle number distributions are shown.

is that the fluctuations of the horizontal wind do not seem to significantly influence the total resuspension process; if the horizontal wind is sufficient to dislodge some particles, it will essentially dislodge "all."

C. THE SOIL PARTICLE SIZE DISTRIBUTION

If the horizontal fluctuations of the wind do not dictate the surface layer aerosol particle size distribution, then it seems reasonable that it is dictated by the size distribution of the available soil particles. From the data of Sehmel, Slinn, and Whitby et al.⁽⁶⁾ it appears that the soil particle number distribution function, f , in the range $1 \lesssim r \lesssim 100 \mu$, may vary as $r^{-\alpha}$ with α between 1 and 2. It would be a valuable test of the hypothesis that the soil particle size distribution dictates the size distribution ($\lesssim 1 \text{ mm}$) of the surface layer aerosol, if simultaneous with resuspension observations, the size distribution of the soil particles was measured.

III. RESUSPENSION

In the above, we have used the term dislodgement to describe the initial motion of the soil particles and have neglected the settling velocity of the particles. We now include the settling velocity and use the word resuspension to describe the process of the vertical transfer of the material. The third phase of the total process, horizontal motion and dry deposition, will not be considered here.

A. DIFFUSION APPROACH

One method to attempt to describe the vertical distribution of the re-

suspended aerosol is to use the convective diffusion equation. This approach is not without limitations because we are proposing to use the diffusion concept in a region where the scale of the diffusion process (the eddy sizes) is comparable to the scale of the fluid flow (the height above the surface). Nevertheless, we shall investigate what this approach predicts.

In the lowest levels of the atmosphere (typically less than 100 m), regardless of the stability, the rate of production of mechanical energy is large compared with the production of convective energy; i.e., the turbulence is governed by mechanical as opposed to thermally generated forces. For this forced convection layer, we assume that the eddy diffusivity for the particles is given by $\epsilon(z) = cu_* z$, where c is an unknown parameter (equal to Von Kármán's constant for the diffusivity of momentum). If $f(r,z)drdz$ is the number of particles per unit volume whose radius is from r to $r + dr$ at height from z to $z + dz$, then for the implied conditions, f satisfies the convective diffusion equation

$$\frac{\partial}{\partial z}(cu_* z \frac{\partial f}{\partial z}) = -v_s(r) \frac{\partial f}{\partial z}, \quad (9)$$

where v_s is the magnitude of the particle settling velocity. The solution to (9) subject to the boundary conditions $f(r, z_0) = f_0$, the surface layer density function, and $f(r, z \rightarrow \infty) = 0$ is

$$f(r, z) = f_0 \left(\frac{z}{z_0}\right)^{v_s(r)/cu_*}, \quad (10)$$

or

$$f(r, z) = f_0 \exp\left[-\ln\left(\frac{z}{z_0}\right) \frac{v_s(r)}{cu_*}\right] \quad (11)$$

Results obtained from Equation (11) are compared with Sehmel's smoothed data in Figure 2. Also plotted in Figure 2 are data obtained by Slinn, Whitby et al.,⁽⁶⁾ Noll and Pilat,⁽⁷⁾ and Jaenicke and Junge.⁽⁸⁾ Notice that the ordinate is the volume distribution, related to the number distribution *via*

$$\frac{\Delta V}{\Delta \ln D} = D \frac{\Delta V}{\Delta D} = \frac{\pi}{6} D^4 \frac{\Delta N}{\Delta D} = \frac{\pi}{6} D^4 f(D)$$

In the comparison with the data, f_0 and z_0 of (11) were taken to be Sehmel's 0.3 m data. Notice that if f_0 is representative of the soil particle number distribution, then $f_s \sim r^{-4} \cdot r^{2.5} \sim r^{-1.5}$. For (11) we have used v_s as given empirically for water drops, for the case $z = 3$ m, and forced the maximum of (11) to occur at the same D as the data by choosing the arbitrary parameter $cu_* = 40 \text{ cm sec}^{-1}$. For $z = 1$ m and 10 m, we have increased v_s by $\rho_p = 2.66$ and chosen $cu_* = 12 \text{ cm sec}^{-1}$ to fit the maximum of the $z = 1$ m data. It is seen in Figure 2 that the convective diffusion equation predicts neither the correct z nor the correct r dependence of $f(r, z)$. As stated earlier, the probable source of the inadequacy is that the resuspension process cannot be described as one of diffusion. The length scale of the "diffusion," z , is the same as the length scale of the flow field, which is the standard criterion for failure of a diffusion analogy.⁽³⁾

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FIGURE 2. Resuspended Particle Volume Distributions. The majority of the data was obtained by Sehmel. For comparison are shown predictions based on the convective diffusion equation, Equation (10), and the statistical approach, Equations (12) and (15).

B. STATISTICAL APPROACH

If we attempt to use a statistical approach, we are soon impeded by a lack of information about the pdf and other statistical properties for the vertical component of the wind. Lumley and Panofsky⁽³⁾ suggest that in neutral conditions, the standard deviation σ_w is Au_* with $A \approx 1.05$. Pritchard et al.⁽⁹⁾ suggested that the pdf for w can be approximated by a sum of exponentials of the form

$c_i \exp(-|w|/\gamma_i)$. Some data for $P(w)$ obtained and analyzed respectively, by Horst and Powell⁽¹⁰⁾ is shown in Figure 3.

Even if $P(w)$ is known, the analysis is not without serious difficulties. To see this, consider the conditions for which the size distribution is maintained at a steady state at some elevation by an equality of upward and downward fluxes. The velocity of a particle of radius r buried in an eddy which has speed

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FIGURE 3. Probability Density Function, $P(w)$. Data points, obtained by Horst and Powell(10) show the relative frequency of occurrences of different vertical wind speeds for neutral conditions and $u_* = 50 \text{ cm sec}^{-1}$. There is a suggestion that the low elevation Gill anemometer may not respond well to the large wind speeds (if the spatial scale $\approx 1 \text{ m}$). Gaussian density functions are plotted for comparison.

w is $v_p = w - v_s$, where v_s is the magnitude of the settling velocity. The upward flux of particles of a given radius, carried by an eddy of speed w from a lower elevation $z - \zeta$, is then equal to the number of particles at this lower elevation,

$f(r, z - \zeta)d\zeta$, multiplied by the probability that the gust will reach z , $P(z, w; \zeta)$, and then multiplied by the velocity and its probability. Then the total upward flux of particles of radius r is

$$\int_{\zeta=-z}^0 d\zeta \int_{-\infty}^0 dv_p f(r, z - \zeta) P(z, w; \zeta) v_p P(v_p + v_s) ,$$

where the pdf for v_p is obtained from the pdf for w simply by using $v_p = w - v_s$.

We have no information immediately available on this "propagation probability" $P(z, w; \zeta)$, which is related to Prandtl's mixing length concept, but is not a mean value over all eddies. It is related to the correlation of w values at different heights phase lagged in time by the height differential divided by w and also phase lagged in x . Presumably $P(z, w; \zeta)$ is strongly dependent on atmospheric stability. If it can be assumed that at some height the balances of fluxes occurs because of propagation all the way from the surface, balanced by the flux from a characteristic height z , then we can proceed, provided $P(w)$ is known. As an example, if $P(w) = (2\gamma)^{-1} \exp(-|w|/\gamma)$, then the equality becomes

$$f_o(r, z_o) \int_0^\infty \frac{v_p}{2\gamma} \exp\left[-\frac{(v_p + v_s)}{\gamma}\right] dv_p \\ = \frac{f(r, z)}{2\gamma} \int_{-\infty}^0 -v_p \exp\left(\frac{-|v_p + v_s|}{\gamma}\right) dv_p .$$

Thus

$$f(r, z) = f_o(r, z_o) \left[1 + \left(\frac{2v_s}{\gamma} \exp \frac{v_s}{\gamma} \right) \right]^{-1} . \quad (12)$$

In Figure 2 we have shown the mass distribution that is predicted by Equation (12) for the case $\gamma = 20 \text{ cm sec}^{-1}$. It is seen that, similar to (10), Equation (12) does not contain

the correct large r behavior. Recognizing the difficulties with this approach and noting, for example, that (12) contains no z -dependence, suggests that this approach is even less valuable than that via the convective diffusion equation. However, from Figure 4 it is seen that the upper end of Horst and Powell's $P(w)$, which is most effective in resuspending large particles, can be fit better by a "power-law" than with an exponential. Specifically we can fit it with

$$P(w) = \left(\frac{w_o}{w}\right)^{5.85} , \\ w > 1 \text{ m sec}^{-1} . \quad (13)$$

When this is substituted into an equation for the balance of fluxes, the large r behavior of the mass distribution becomes

$$f(r, z) v_s \sim f_o(r, z_o) \left(\frac{c}{v_s}\right)^{3.85} \\ \text{for large } r . \quad (14)$$

If $f_o(r) \sim r^{2.0}$ and since for large r , $v_s \sim r$, then (13) yields the large r behavior

$$f(r, z) \sim \frac{1}{r^{2.85}} . \quad (15)$$

This asymptotic behavior is shown in Figure 2 and it is obviously very close to the behavior of Sehmel's data. This encouragement impels us to proceed further with this approach.

IV. CONCLUSIONS

In summary, it appears that the problem of predicting when individual

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FIGURE 4. Limited Range of $P(w)$. The solid line shows that a "power law" fits fairly well the upper end of the 7.5 m, sonic anemometer data of Figure 2.

particles will be dislodged under the action of a turbulent wind is fairly well in hand. The ballistic effect is probably complicated, but it is our contention that the total effect of dislodgment results in a particle size distribution that is dictated by the size distribution of the soil particles. It would be valuable if these two size distributions were reported simultaneously in future resuspension studies.

The resuspension of the material into the forced convection layer depends almost entirely on the statistical properties of the turbulence about which we know too little. It would be most valuable if measurements for the pdf for w as a function of height, and the correlations men-

tioned earlier, were obtained simultaneously with resuspended particle size distributions. Some day it may even be found that measuring particle size distributions is the most straightforward method to obtain information about the exceedance statistics of turbulence.

We have not addressed the problems of deep-layer mixing nor unsteady diffusion of the resuspended material. Certainly, the particle size distribution will evolve with time as dry deposition becomes important. These topics will be addressed at a later time as will the problem of predicting the z -dependence of resuspended particles.

ACKNOWLEDGEMENTS

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RESUSPENSION BY WIND AT ROCKY FLATS

G. A. Sehmel and M. M. Orgill

Airborne plutonium concentrations are being related to meteorological parameters at Rocky Flats in anticipation that these data will increase our understanding of environmental resuspension processes. Airborne concentrations at sampling station S-8 were related to wind speed for one time period from July 1970 to January 1971.

Our research goals are to develop general models for predicting particle resuspension rates and potential downwind hazards. We are experimentally developing the data base for these models by seeding surfaces at Hanford and other proposed sites with inert tracers of known properties and subsequently by mass budgets are determining wind caused particle resuspension rates under various meteorological and surface conditions.

In the case of Rocky Flats, the plutonium ground source characteristics as to particle diameter, concentration, etc., are not sufficiently defined to determine resuspension rates from mass budgets. Neverthe-

less, information applicable to resuspension models has been collected at Rocky Flats. Airborne plutonium concentrations have been measured at 20 air sampling stations operated by Dow Chemical,⁽¹⁾ HASL,⁽²⁾ and the Colorado Department of Health.⁽³⁾ In addition, Dow Chemical has recorded meteorological parameters including hourly wind speed and direction which are applicable to resuspension modeling. Consequently, we are using the reported airborne plutonium concentrations and meteorology to further our development of resuspension models.

Through the cooperation of those investigators developing and reporting

data for monitoring programs, we have obtained all the available airborne plutonium concentrations in the sampling network surrounding Rocky Flats and the corresponding meteorological data. The meteorological data include hourly wind speed and direction and also the maximum daily wind gust at the tower site. The anemometer was located 40 feet above ground and 3/4 mile west of the oil spill area. The air concentrations were measured at filter sampling stations located near the east security fence (east of the oil spill area) and at additional filter sampling stations spaced around the plant area at a 2-3 mile radius. In our modeling, we must ignore the airborne plutonium dependence on particle diameter since only very limited data have been collected for determining the particle size distributions of the airborne plutonium⁽¹⁾ and the respirable size fraction.⁽⁴⁾

Wind surface stresses are resuspending plutonium in the Rocky Flats area and are producing local airborne plutonium concentrations above fallout levels but below maximum permissible airborne plutonium concentrations: the maximum measured airborne plutonium concentrations since 1969 are 51 fCi/m³ for a 6 hour sampling period;⁽⁵⁾ 20 fCi/m³ for 24 hours;⁽³⁾ 410 for 48 hours;⁽³⁾ 6.6 for a week;⁽²⁾ and 37 fCi/m³ for a month.⁽³⁾ Average airborne concentrations are much less than these maximums. Despite low activity levels and smoothing of variability in airborne concentrations performed by sampling over relatively long time periods, the reported data are useful in de-

fining some dependencies of resuspension on meteorological parameters.

In the data analysis, we have looked at the inadequacies of resuspension factors for predicting plutonium resuspension. Our calculations of resuspension factors from airborne plutonium concentrations and surface contamination suggest that maximum resuspension factors, for scoping estimates, are in the range of 10^{-5} to 10^{-9} m⁻¹ for 6 hours, 48 hours, and weekly air samples collected near the east security fence and also at a 2 to 3 mile radius from the plant. These resuspension factors are within the wide range of resuspension factors that have been determined at different sites by several investigators.⁽⁶⁾ However, the simple use of resuspension factors, without detailed qualification, is of doubtful value. One of the principal shortcomings of the resuspension factor approach is that ignoring the physical processes effecting resuspension severely limits the ability to develop a generalized predictive capability. The airborne concentration should be related to an upwind and not a local surface contamination. We show here that a direct examination of airborne plutonium concentrations and the associated meteorology is proving to be amenable to quantitative analysis and is leading to the development of predictive resuspension rate models.

MONTHLY AIRBORNE CONCENTRATIONS

The general oil spill area is still a source of wind resuspended airborne plutonium. This can be

shown by inspection of Figure 1. Time sequences are shown for monthly airborne plutonium concentrations measured at six stations along the eastern security fence and for the associated hours of wind speed greater than 15 mph and the wind gustiness during the month.

The six air sampling station locations, D-1, D-2, APC-56, S-8, D-3 and D-4, are shown in relationship to the oil spill area in the lower left corner of the figure. By comparing the relative locations of the oil spill area to the air sampler locations, it can be seen that westerly winds should cause the most resuspension. Indeed, the contours of highest measured airborne activity are principally in the path of west winds. As seen by the upper two figures, west winds predominate and conse-

quently there is a direct correlation between westerly winds and the maximum airborne plutonium concentrations. It is also seen that station location D-4, located at the south end of the sampling sites, oftentimes shows relatively high airborne concentration. During these times of relatively high concentrations, it would be useful to locate an additional sampling site south or southwest of D-4 to obtain a more complete concentration distribution.

Westerly winds are not the sole criteria for observing high monthly average airborne plutonium concentration. If it were the sole criterion, the airborne concentration data should always show a maximum at about station S-8 and decrease for both the northern and southern sampling stations. We are attempting to quantify

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FIGURE 1. Monthly Airborne Concentration as a Function of Wind

the effect of these other influences, such as precipitation, on resuspension but as yet can only qualitatively discuss their possibly observed effects. It is anticipated that these other influences will be more readily discerned during shorter air sampling periods. Vehicle-caused resuspension can be significant⁽⁷⁾ and is suspected to be important for air samplers located along the east security fence since a road parallels the fence. In fact, higher airborne concentrations at D-1 and D-2 during the summer months of 1972 are attributed⁽⁸⁾ in part to increased local vehicular traffic.

SHORTER SAMPLING PERIODS

Weekly airborne plutonium concentrations measured at the S-8 location by HASL⁽²⁾ have shown the best cor-

relation with wind direction and speed, gustiness, precipitation and snow cover. Figure 2 shows a time series of the weekly airborne concentration from July 1970 to January 1971. Concentrations were consistently above fallout levels. A general, though not definitive correlation can be observed between airborne concentration, number of hours of wind speed ≥ 20 mph and maximum wind gusts. The correlation is better during the fall and winter months.

Precipitation, possibly through wetting the soil, appears to decrease airborne concentrations. Snow cover shows the same tendency but this latter effect cannot be directly confirmed because snow cover was not recorded at Rocky Flats.

Some puzzling aspects of plutonium resuspension have been encountered when examining the 6 hour and 48 hour

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FIGURE 2. Weekly Airborne Concentration as a Function of Meteorology

airborne concentrations data obtained along the eastern security fence. These locations have shown relatively high values of airborne concentration during periods of relatively quiescent wind conditions. We suspect that these high concentrations might possibly be attributed to sampling or radiochemical errors but that other factors, such as vehicular resuspension, buoyant convection, soil surface structure, precipitation, etc., could be complicating the resuspension process.

RESUSPENSION MODEL

The physics of wind-caused plutonium resuspension should as a first approximation be similar to the physics of soil erosion. The rate of soil erosion is proportional to air velocity cubed⁽⁹⁾ (actually the friction velocity, u_*) when the air velocity is above a threshold velocity. Consequently, initial plutonium resuspension modeling has assumed that resuspension is a function of air velocity to an unknown power when the air velocity is above an unknown threshold velocity. However, for plutonium resuspension, the threshold velocity was not significantly different from zero in the simple model that was developed.

The model was developed using Volchok's⁽⁴⁾ weekly average airborne plutonium concentration data for station S-8 shown in Figure 2. Previously, Volchok had presented the data as a figure of weekly average concentrations versus weekly average wind velocity.

The resuspension model was developed for station S-8 on the basis that total plutonium collected on each filter was more important in elucidating wind caused resuspension processes than was the average weekly concentration determined from each air filter. In order to determine the effect of each hour of wind speed on the total filter collection, the wind record was analyzed in terms of its shortest time span of one hour. The model was developed so that the airborne plutonium concentration caused by each hour of wind could be predicted and the effect of all hours summed in order to predict the total filter collection during the week. Since our preliminary analysis indicated that only W and SW winds caused significant resuspension, only the winds from these two directions were used in developing the model.

The model for wind caused plutonium resuspension at sampling station S-8 during July 1970 to January 1971 is

$$(\bar{X}) = 0.45 (\bar{u})^{2.1} \quad (1)$$

in which the average airborne concentration, $\bar{X}$ (in units of fCi/m³) for a one hour time period is a function of the average west and southwest wind speed, $\bar{u}$, in miles per hour during the same time period. The constants were determined by using the technique of least squares between the predicted and the measured weekly average concentrations. The linear correlation coefficient for the model is 0.89.

The exponent value of 2.1 on $\bar{u}$ may be significant in terms of plutonium resuspension rates, Q , described by a diffusion model $\chi/Q = (1/\bar{u})f(\sigma, x, y, z)$. Since the literature suggests that soil movement is proportional to u_*^3 and consequently $\bar{u}^3$, the particle resuspension rate Q as a first approximation is also proportional to $\bar{u}^3$. Thus, $\chi = (\bar{u}^3/\bar{u})f'(\sigma, y, x, z) = \bar{u}^2 f'(\sigma, x, y, z)$, or the airborne concentration is theoretically predicted to be proportional to $\bar{u}^{2.0}$ compared to the experimental $\bar{u}^{2.1}$ for the airborne plutonium concentration model. However, additional modeling for other time periods and sampling stations is required before any model generalization of this exponent is warranted.

At station S-8 near the oil spill area and during the July 1970 to January 1971 time period, the model predicts that the total activity in each hourly time period is

$$\text{fCi/m}^3 = \text{FO} + 0.3 + 0.45 (\text{mph})^{2.1}, \quad (2)$$

in which FO is the plutonium fallout activity in fCi/m^3 during the time period and the constant 0.3 fCi/m^3 is the local activity which cannot be attributed to the W and SW winds. These contributions to total airborne plutonium concentrations are shown graphically in Figure 3. The fallout and local activity contributions are usually insignificant compared to the contributions of high wind speeds. The model predicts that hourly average concentrations can be much higher

than weekly average concentrations. Such large hourly concentrations have not been experimentally observed since the model predicts hourly statistically averaged concentration which would not be sustained over the weekly time period that data were integrated. The model predicts that if only one hour of 60 mph winds were the only winds causing resuspension, the weekly average concentration would be only $(2400) (1/168) = 14.3 \text{ fCi/m}^3$.

FUTURE RESEARCH

The research has developed a model for predicting airborne plutonium concentrations at sampling station S-8 during one time period. Similar analyses of all time periods for all stations should lead to a better prediction of airborne plutonium concentrations and a more definitive inhalation exposure during the hourly time periods of high winds. It would be useful to experimentally determine, on a research basis rather than the monitoring basis currently conducted at Rocky Flats, the potential plutonium resuspension as a function of at least wind speed, the respirable size range of the airborne plutonium, air sampling height, and surface conditions. In such research, the present anemometer site should be changed from on top of a building to an open site or an anemometer site measuring both winds and turbulence should be added.

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FIGURE 3. Airborne Hourly Plutonium Concentrations at S-8 (July 1970 - January 1971)

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EVALUATION OF HI-VOLUME CASCADE PARTICLE IMPACTORS

G. A. Sehmel

Commercially available 20 cfm cascade particle impactors were evaluated for sampling errors caused by wall losses and by impactor orientation. Particle wall loss on the impactor walls increased with both an increase in stage soil collection and an increase in airborne particulate concentration. The particle size distributions obtained with the impactors are biased if the sampler inlet is not directed into the prevailing wind.

INTRODUCTION

The accurate sampling of airborne particulates is important in determining both the airborne particulate concentration and the airborne particulate size distributions. These determinations are important for sampling any airborne particulate pollutant and are especially important in our particle resuspension and particle deposition research. In both research areas we will eventually need to accurately determine mass budgets of airborne materials for field determinations of particle resuspension rates or particle deposition velocities across large surface areas. At present, the sampler evaluation is directed toward adequately sampling resuspended particles.

Resuspended particles can present a more severe downwind health hazard if the particles are initially in the respirable size range. Even if the particles are not immediately respirable, the particles can by deagglomeration become fractionized into a respirable size fraction in succeeding deposition-resuspension sequences. Consequently, we must determine in our mass budgets of airborne material the total transport of material in each and all particle diameter ranges.

We have developed an integrated sampler for measuring airborne particulates as a function of particle diameter. The sampler shown in Figure 1 is an exploded view of our locally designed and fabricated rotating

cowl which is attached to the commercially available Andersen 2000 Inc. 20 cfm hi-volume cascade impactor. The cowl is a rotating device which is attached to the impactor by the spindle extension. The cowl inlet is continually directed into the wind by the wind forces on the wind orientation tail fin.

The two purposes of the rotating cowl are (1) to collect by gravity settling within the cowl body the airborne particles greater than about 10 to 20 μm which enter through the

cylindrical sampler inlet and (2) to eliminate particle sampling bias which would have been caused by failure to direct the sampler inlet continually into the wind. Anisokinetic sampling errors however are not eliminated by this design. Since the air flow rate is held constant at 20 cfm as required by impactor design, the air entering the 6 inch diameter cylindrical sampler inlet corresponds to a wind speed of only 1.2 mph. This low equivalent wind speed means

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FIGURE 1. Exploded View of Rotating Cowl and Impactor

that we are always sampling subisokinetically⁽¹⁾ for wind speeds of interest to resuspension. Consequently particle inertia will consistently cause more particles to enter the sampler inlet than were initially in the air drawn through the sampler inlet. This increased particle collection means that we are presently estimating airborne particle concentrations to be greater than actual. Thus, our particle resuspension rates will be estimated to be too high and consequently our measured resuspension rates will be conservative. We do, however, have plans for experimentally determining the subisokinetic sampling errors correction factors for the cowl inlet as a function of particle diameter and wind speed.

IMPACTOR ORIENTATION

Airborne particle dust collection on each impactor stage was determined at identical sampling heights for a cowl-impactor system and an impactor without cowl. For the impactor without cowl, the impactor inlet was either pointed vertically down towards the ground surface or was pointed up away from the ground surface. In order to determine the effect of sampler orientation on stage collection, the collection on each impactor stage was normalized to the corresponding stage collection in the cowl-impactor system. Collection on each stage was determined gravimetrically.

The effect of impactor orientation was determined for impactors located

at the 0.3, 1, 2, 3 and 10 m heights above ground. The comparisons are shown in Figure 2 as a function of the 50 percent cutoff particle diameter at each impactor stage. Also shown is a comparison for particle less than 1 μm collected on the filter backup. The curves show in general that (1) an impactor pointed upwards will collect more particles than an impactor pointed downwards, (2) the normalized collection on each stage is a function of particle diameter, and (3) the normalized collection is unexpectedly also a minor function of sampling height. The gravitational force can be used to

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FIGURE 2. Effect of Impactor Orientation on Stage Collection

rationalize that an impactor pointed upward should collect more particles than an impactor pointed downward. However, the effect of sampling height is difficult to explain unless impactor collection efficiency is a function of particle airborne concentration which is known to be a function of height.

Although additional data would be required to completely explain these orientation effects, we can conclude that the most accurate sample of airborne particulates is obtained when a rotating cowl is attached to the impactor.

IMPACTOR WALL LOSSES

We have also determined that the wall losses can be significant in this impactor. Wall loss as defined is the ratio (1) of the amount of dust brushed off and weighed from the back side of the impactor stage immediately opposite the impactor stage of interest divided by (2) the amount of dust collected on the impactor stage of interest. These wall losses are shown in Figure 3 as a function of the mg of dust collected at each stage. The wall losses are minimal at a 50 mg collection but rapidly increase for a 100 mg collect. We have other data showing that wall losses are even more significant for samples collected during higher airborne soil concentrations.

FUTURE RESEARCH

Accurately sampling to determine particle size distributions still re-

mains a problem in sampling large quantities of airborne particles. The largest air flow rate impactor commercially available has large wall losses and probably particle bounce off and re-entrainment problems somewhat limiting their satisfactory usage for the present sampling application. Nevertheless, this is the largest commercially available impactor which tends to solve our sampling problem. We have plans for coating the collection surfaces of these impactors in order to minimize particle re-entrainment and

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FIGURE 3. Impactor Wall Losses During a 430-Hour Sampling Period

possibly wall losses and also for designing a new higher air flow rate impactor which will be more directly applicable to determining size distributions of resuspended particles.

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FRACTIONAL AIRBORNE RELEASE OF RADIOACTIVITY DURING THE BURNING OF CONTAMINATED SOLVENTS*

J. Mishima and L. C. Schwendiman

Laboratory scale experiments are being conducted to indicate the airborne release of radioactive materials in the event of a fire involving flammable solvents used in the separation of nuclear materials. The fractional release of materials during the burning of 30 percent normal tributyl phosphate in a kerosine diluent is being measured. The solvent contains realistic amounts of materials representative of those found in contaminated process solvents--uranium, cesium, cerium, zirconium, iodine, and ruthenium. Trace quantities of active isotopes are used to follow the distribution of the materials except in the case of uranium. Release measured thus far under nominal conditions (distillation of the diluent) have been under 1 percent.

INTRODUCTION

Because nuclear systems provide an increasing portion of the nation's growing energy demands, the behavior of radioactive materials under various conditions which that material encounters in the entire cycle is of concern. Fissile and fertile materials are a resource and must be recycled to insure a continuing contribution from this source. Under current technology, liquid-liquid extraction using normal tributyl

phosphate (TBP) in a kerosine diluent (Normal Paraffin Hydrocarbon) is a commonly used means of separating fissile and fertile materials from waste products generated in reactors. Although the n-tributyl phosphate is relatively nonflammable (flash point 295°F, Class I flammability),⁽¹⁾ kerosine is used as a burner fuel. Thus, this extraction process solvent could conceivably be involved in a fire. This study is involved with the release of radioactive materials during the burning of a 30 percent TBP in NPH.

* Work supported by the Atlantic Richfield Hanford Company.

DISCUSSION

A series of experiments is being performed to determine the fractional release of various elements (broadly representative of various types of radioactive materials found in extraction process solvents) during the burning of 30 percent TBP in NPH under nominal conditions. A single 30 percent TBP in NPH solution containing 306 g of uranium per gallon with part per million concentrations of zirconium, cerium, cesium, iodine and ruthenium is used in all experiments. A 25 milliliter aliquot is placed in a 50 milliliter beaker and warmed. The vapors are ignited and placed under the inlet of a 12-inch long chimney made of 75 millilimeter outside diameter pyrex tubing. Air is drawn up the chimney at a rate of 1 or 2 cfm and airborne particles are collected on a glass fiber filter at the upstream end of the chimney. Air passing the filter is subsequently passed through a bed of charcoal or gas bubbler containing 1N NaOH depending upon the element under test.

The fractional airborne release of uranium measured in the initial two experiments was small--0.023 and 0.027 percent. The low fractional releases are not surprising. Under the nominal conditions applied, the solvent temperatures remained low (less than boiling) and the surface of the liquid calm. Heat is not readily transmitted by the glass and vapor generation is primarily from the surface layer heated by radiation.

Under these circumstances, the more volatile component, NPH, is distilled slowly from the mass of liquid and is effectively separated from the other components. Under more rigorous conditions with rapid boiling and surface disturbance, larger quantities of heavy metals in the solvent would be airborne. Greater than 99 percent of the uranium measured was recovered from the liquid residue, TBP, remaining after the fire.

In the experiments involving the fission products, trace quantities (approximately 0.1 μ Ci) of the specific isotopes of interest were added to the solvent. The quantity of these materials present in each sample was determined by comparison of the counting rate on a gamma spectrometer to the rate of a reference containing a known amount of the isotope in the same matrix counted at the same time.

Experiments have been performed under these nominal conditions using ^{137}Cs , $^{144}\text{CePr}$ and $^{95}\text{Zr-Nb}$. Fractional airborne release thus far have remained less than 1 percent. Additional tests involving ^{131}I under these nominal conditions and all materials under more rigorous conditions (charring of the liquid residue) are planned.

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AIRBORNE RELEASE OF PLUTONIUM AND ITS COMPOUNDS
DURING OVERHEATING INCIDENTS*

J. Mishima and L. C. Schwendiman

A study has been in existence for some time to evaluate the amount and distribution in aerodynamic size fractions of plutonium airborne under some conditions found in postulated accidents. Two recent studies are covered here. Data are presented from forty experiments performed in the wind tunnel in the Radioactive Aerosol Release Test Facility to measure releases under various combinations of parameters involving uranium (a plutonium simulant) forms, surfaces, wind speed and the presence or absence of a gasoline fire. The second study involves airborne release during the burning of contaminated waste in the Radioactive Aerosol Release Tank.

INTRODUCTION

Utilization of increasing quantities of plutonium is considered this nation's prime hope for meeting its power requirements.⁽¹⁾ The record of the nuclear industry in protecting workers and the general public from the harmful aspects of plutonium have demonstrated that it can be handled safely. Increased usage means a larger number of people involved and changes in technology can alter conditions. Both increase the opportunity for error. Thus additional attention is required in designing and evaluating each operation. A review of incidents which have occurred in the nuclear industry has shown that the majority of incidents are normal industrial accidents and involve fires.⁽²⁾ About 70 percent have occurred during what is nominally considered chemical processing

and an additional 28 percent during handling and storage.⁽²⁾ This study is involved in measurement of the fractional airborne release from plutonium and its compounds under thermal and aerodynamic stresses which may accompany some accidents.

DISCUSSION

The program has progressed through reviews of incidents involving plutonium and its compounds where control criteria were exceeded⁽²⁾ and of existing data on airborne release from these materials.⁽³⁾ Data generated from small scale laboratory experiments yielding order of magnitude estimates of the airborne release from plutonium and its compound have been covered in last year's annual report.⁽⁴⁾ A larger scale facility--The Radioactive Aerosol Release Test Facility--has provided the capability for measurement of fractional airborne release of radioactive particles from burning materials on a more realistic scale. Uranium was

* Work supported by the Atlantic Richfield Hanford Company's Environmental and Regulatory Technology Program.

shown to be the most suitable stand-in for plutonium in the experiments.

Two studies are reported here. The first concerns the amount and size distribution of particles that may become airborne under conditions found in some shipping accidents. Some forty experiments were performed in the wind tunnel in the test facility. Eleven experiments were performed in the Radioactive Aerosol Release Tank (RART) to measure the amount and distribution in aerodynamic size fractions of the activity airborne during the burning of contaminated flammable wastes.

Fractional Airborne Release and Distribution of Activity in Aerodynamic Size Fractions Under Some Conditions Found in Selected Shipping Accidents

Forty experiments were performed in the wind tunnel in the Radioactive Aerosol Release Test Facility to measure the airborne release of uranium (representing plutonium) using a combination of parameters which could be found in some types of shipping accidents. The wind tunnel is shown in Figure 1. Uranium dioxide powder, uranium nitrate liquid or the solid residues from an air dried nitrate solution--representing plutonium

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FIGURE 1. Wind Tunnel in Radioactive Aerosol Release Test Facility

forms that might be encountered in shipping accidents--were dispersed on four surfaces--smooth sandy soil, sandy soil with a vegetation cover, stainless steel, and an asphalt-gravel aggregate commonly used on the Hanford site for roads. Air at 2.5 or 20 mph was passed over the surface and airborne release measured in the presence or absence of a gasoline fire. The results from the experiments are tabulated in Table 1.

No single variable appears to exert an overriding influence on fractional release. It appears that a number of interacting factors may cause a situation which is favorable or unfavorable to release. The

largest fractional release was by aerodynamic entrainment from smooth, sandy soil by air with a velocity of 20 mph--24.4 percent in a 24 hour period. All the material appeared to be airborne within the first hour. Engulfing the residue in a gasoline fire made an additional 0.34 percent airborne in some 0.3 hour. An additional 1 percent was aerodynamically entrained in 24 hours from the residue after the fire. A plot of uranium airborne versus time for this experiment indicates additional material may have been airborne with further sampling. The next largest release was during a gasoline fire engulfing uranium nitrate liquid on a

TABLE 1. Uranium Results--Engineering Scale Experiments

Uranium Form Used	Wind Speed	Expt. #	Aerodynamic Entrainment		Expt. #	Petroleum Fire	
			Wt. Percent Source Airborne (hours sampled)	Less Than ¹ 10 Microns		Wt. Percent Source Airborne (hours sampled)	Less than ¹ 10 Microns
SMOOTH, SANDY SOIL	UO ₂	Low ²	2 0.43 (6)	56	3+	0.63 (6)	5
		22a	0.042 (24)	54	22b	0.48 (1)	72
		22c*	0.0092 (24)	54			
	Moderate ³	1	2.3 (24)	75			
		23a	24.4 (24)	40	23b	0.34 (0.3)	62
		23c*	1.0 (24)	68			
	Air Dried ⁴	Low ²	4b 0.0069 (24)	74	5+	0.0074 (5.5)	64
		8b	0.0054 (24)	77	9+	0.004 (5)	18
		6b	0.93 (24)	22	7+	0.12 (7)	32
	UNH ⁵	Moderate ³	26b* 1.1 (24)	64			
VEG. COVER	UO ₂	Low ²	4a 0.049 (6)	76	10+	0.0017 (5.5)	75
		8a	0.012 (24)	84			
		Moderate ³	6a 0.039 (28)	68	26a	0.54 (0.13)	40
	UNH ⁵	Low ²	11 0.0046 (24)	82	12+	0.014 (6)	91
		Moderate ³	13 1.0 (24)	40	14+	3.1 (6)	19
		Low ²	- -	-	16+	0.096 (6)	78
	UNH ⁵	Moderate ³	- -	-	15+	0.2 (6)	86
		Low ²	17b* 0.09 (4.8)	83	17a	0.12 (0.15)	66
	UO ₂	Moderate ³	25a 7.6 (24)	14	25b	1.3 (0.13)	7
		25c*	0.7 (24)	42			
STAINLESS STEEL	UO ₂	Low ²	19b* 0.023 (6)	76	19a	1.1 (0.22)	40
		Moderate ³	18b* 2.6 (5)	30	18a	11.4 (0.17)	34
	UNH ⁵	Low ²	21b* 0.14 (6)	62	21a	0.041 (0.37)	70
		Moderate ³	20b* 0.063 (5)	58	20a	0.24 (0.28)	68
ROAD	UO ₂	Low ²	21b* 0.14 (6)	62	21a	0.041 (0.37)	70
	UNH ⁵	Moderate ³	20b* 0.063 (5)	58	20a	0.24 (0.28)	68

¹Percent airborne less than 10 microns Aerodynamic Equivalent Diameter

²Approximately 2.5 mph

³In range of 20 to 23.5 mph

⁴Dried in flowing air at stated velocity for a minimum of 24 hours

⁵Uranyl nitrate hexahydrate

*Residue after gasoline fire

†During and immediately following a gasoline fire

stainless steel surface by air with a velocity of 20 mph--11.4 percent in 0.17 hour. An earlier experiment involving air drying of plutonium nitrate solutions at ambient temperatures and low air velocities indicates fractional releases in the order of 10^{-7} percent in 24 hours from a stainless steel surface.⁽⁵⁾ An additional 2.6 percent was entrained in 5 hours from the solid residues remaining after the fire. Lesser but still significant quantities were airborne under other combinations of parameters.

Fractional Airborne Release and Distribution of Activity During the Burning of Contaminated Waste

Realistic quantities of packaged wastes containing uranium were burned in the RART and the quantity and size distribution of the particles airborne were measured. Eleven experiments were performed in which three forms of uranium--uranium dioxide powder, uranium nitrate liquid, and the solid from an air dried uranium nitrate solution--corresponding to the plutonium forms most frequently involved with flammable waste, were mixed with 4.283 to 4.696 kg of assorted flammable waste held in a cardboard carton. The various flammable materials used were: cardboard, various forms of paper, plastic, rubber and miscellaneous items such as rags, oil, etc. Three packaging configurations were used: 1) half the uranium was placed on loose waste sealed in plastic bags which were randomly dispersed along with the remainder of the uranium on the loose waste held in a sealed carton;

(2) all the uranium was randomly mixed with the loose waste held in a sealed carton; and 3) unsealed cartons with uranium dispersed on loose waste.

The RART is a cylindrical, stainless steel vessel 9.5 feet in diameter by 10 feet tall. The vessel is equipped with ten 4-inch diameter sampling ports, a 24-inch by 48-inch entry port, a 100 cfm closed loop for recirculation of the enclosed gases, and a filtered inlet and exhaust. The vessel is shown in Figure 2. The

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FIGURE 2. Radioactive Aerosol Release Tank

packaged waste was burned on a screened tray (shown in Figure 3) which retained the residue from the fire. Wall deposition and fallout collectors were used. The quantity and distribution of activity into aerodynamic size fractions was monitored by periodic filter and cascade impactor samples of the atmosphere of the vessel. The uranium content of the various samples was measured by fluorimetry.

The results of the eleven experiments are shown in Table 2 and indicate the fractional airborne release is low--a maximum value of 0.053 percent. The results are consistent with small scale laboratory experiments previously reported⁽⁴⁾ and appear reasonable for the conditions imposed. Some degree of correlation is anticipated between the quantity airborne and deposited on various surfaces in a closed system as used in this experiment. A rough correlation is evident and more agreement is anticipated if there were a clear separation between the burning materials and the vessel. The low values are further corroborated by the relatively small size of the particles airborne--Activity Median Diameters ranged from 0.24 to 5.2 microns Aerodynamic Equivalent Diameter (AED). In 8 of 11 cases, greater than 80 percent of the uranium airborne was associated with particles less than 10 microns AED. Packaging configuration nor uranium form appear to correlate significantly with fractional

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FIGURE 3. Screened Burner Tray

release. Some variation is discernible within each uranium form for the three packaging configuration--higher fractional releases are found using dioxide powder with 2 of the 3 packaging configurations--although no clear trend is established. Packaging configuration does not have a significant effect. Thus, although the limiting parameter or combination of parameter was not identified, fractional airborne release under the conditions studied were low.

TABLE 2. Fractional Airborne Releases of Uranium
During the Burning of Contaminated Waste

Experiment No.	Uranium Compound Used	Weight Uranium Used (grams)	Waste* Config.	Fractional Airborne Release (% of Uranium Used as Source)		Percent Uranium Recovered	Percent Uranium Recovered In			
				Max. Airborne Concentration	Wall Deposition		Residue	Floor	Walls	Airborne
FW-1	UO ₂	9.1	1	0.053	0.048	78	98.2	1.7	0.06	0.07
FW-2	a.d.UNH	25.5	1	0.003	0.045	80	99.9	0.06	0.008	0.003
FW-3	UNH	25.5	2	0.0083	0.011	85	99.5	0.46	0.013	0.01
FW-4	UNH	25.5	1	0.0047	0.11	177	99.9	0.01	0.06	0.003
FW-5	UO ₂	64.1	2	0.017	0.23	69	98.6	0.9	0.5	0.02
FW-6	UO ₂ /UNH	51	2	0.0041	0.0033	89	99.9	0.08	0.004	0.005
FW-7	UNH	25.5	2	0.0030	0.0094	85	99.9	0.1	0.01	0.004
FW-8	UO ₂	50.2	3	0.0052	0.010	74	99.3	0.7	0.01	0.007
FW-9	a.d.UNH	25.5	3	0.015	0.016	65	99.9	0.07	0.03	0.06
FW-10	UNH	51	3	0.015	0.0029	66	99.7	0.3	0.004	0.02
FW-11	UO ₂	51.6	2	0.015	0.024	91	99.4	0.5	0.03	0.008

X 97

UNH - Uranyl nitrate hexahydrate solution

a.d.UNH - Air dried following addition to waste as UNH solution

*See text

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PROFILE-DERIVED ESTIMATION OF CROSSWIND INTEGRATED EXPOSURE
FROM ELEVATED SOURCES: MODEL REVISIONS AND APPLICATIONS

C. E. Elderkin and D. C. Powell

The prediction of ground level exposure to a depositing tracer released from an elevated source is shown to be highly sensitive to changes in the roughness parameter, z_0 . At close downstream distances the order of magnitude differences in predicted exposure are due to differences in σ_z associated with the differences in z_0 . At downstream distances on the order of 5 to 15 kilometers, the predicted exposure differences are due primarily to different deposition effects associated with differences in z_0 . The same analysis for a nondepositing tracer shows slight differences in predicted exposure at the greater downstream distances associated with changes in z_0 . However, the predicted exposures at these distances for the nondepositing tracer are of a higher order of magnitude than those for the depositing tracer.

During the last decade problems of concern to the AEC involving diffusion estimates generally arose in connection with facilities in open desert-like areas and were limited in scale usually within 1 kilometer or so. The model developed and applied to such practical continuous point source diffusion problems, the Pasquill-Gifford model, served ideally for such applications since it was uncomplicated (easy to understand, and easy to use) and was empirically based on diffusion data appropriately gathered over flat open areas out to distances of nominally 1 kilometer from the source. In recent years, however, a wider variety of pollution problems has arisen, and the application of the Pasquill-Gifford model has been pressed beyond the types of problems for which it was originally intended and beyond the limitations of the data on which it was based. For example, as concern for worst case dif-

fusion estimates associated with ground source accidental releases is replaced with concern for realistic diffusion estimates of routine releases from elevated sources, it becomes important to account for changes with height which are not considered in a simple extrapolation of the surface experimental results.

Also, since nuclear power generating facilities are being planned and installed throughout the country, it is important to assure that models used for diffusion assessments over widely varying types of surfaces (from very rough mountainous and forested surfaces as well as built-up areas near and within cities to smooth surfaces found over plains and large water bodies) account for any significant effects on concentrations due to the degree of roughness. Models presently in use do not recognize any dependence on surface roughness.

Our interest has broadened to include a wider variety of pollutants in recent years. The diverse interactions of these with a given surface type, to say nothing of the wide variety of surfaces now of interest, result in removal properties and efficiencies that vary widely and influence air concentration distributions in a significant but ill-defined manner.

The model discussed here, based on the Hay-Pasquill technique⁽¹⁾ applied to vertical dispersion and the description of deposition summarized by Van der Hoven,⁽²⁾ attempts to account for some of these effects which are now becoming important. Only the crosswind integrated exposures have been modeled^(3,4) since the effects mentioned above are associated primarily with the vertical dispersion. It is not a completed model, ready for application, since it has not been evaluated and adjusted for a wide variety of surface and meteorological conditions. However, it serves to explore the seriousness of effects not previously included in practical continuous point source models, but which are becoming involved in real problems.

REVIEW OF THE MODEL

In its present form, the model focuses on the vertical problem by estimating the crosswind-integrated exposure (CIE) downstream at ground level only. The exposures are due to the release of a known amount of depositing tracer over a short length of time.

The following input is required for calculation of crosswind-integrated exposures as measured on the Hanford stable arc grid.

Geometrical and diffusion input

1. Mass of depositing tracer released, Q
2. Height of release, H
3. Measured crosswind-integrated exposure, E , at each arc
4. Distance from the source to arc, X
5. Surface roughness parameter, z_0

Meteorological input

1. Mean wind speed at 15 meters, $\bar{U}(15)$
2. Mean wind speed at 2.2 meters, $\bar{U}(2.2)$
3. Richardson number at the geometric mean height, 5.7 m, $Ri(5.7)$.

Analytical input

1. A power spectral model for the vertical turbulence component, $nS_w(n)$
2. A characteristic diffusion height, z_c , for scaling the power spectrum
3. A stability-dependent formulation of the Hay-Pasquill β parameter, $\beta = A_1 + A_2 Ri(5.7)$
4. A deposition velocity of the form $V_d = C \frac{u_*^2}{\bar{U}(2.2)}$
5. A settling velocity, V_s (not used thus far).

From these inputs, values of σ_z^2 and the final crosswind integrated exposures were calculated for each test and for each arc according to formula given.^(3,4) The σ_z^2 values

were calculated from the Pasquill equation; the crosswind integrated exposures from a Gaussian equation using a σ_z value and from the deposition function given in Reference 2, Equation 5.48.

REVISION OF THE MODEL

There are two revisions of the model to be offered. One of these is a genuine change; the other is simply better perception of what was already in the model.

The change consists of using a different function to shift the spectral frequency curve with changes of stability. Kaimel, et al.,⁽⁵⁾ state that for neutral and stable cases the power spectrum for each wind component exhibits a universal shape.

The change consists of using a different function to shift the spectral curve frequency wise with stability changes. From examination of the abscissas of the peaks in the curves for stable spectra in⁽⁵⁾ Figure 4, the following empirical shift function is suggested.

$$f_m\left(\frac{z}{L}\right) = f_m(0)\psi\left(\frac{z}{L}\right)$$

where f_m is the nondimensional frequency, $f = nz/\bar{U}$, at which the maximum spectral value occurs and where

$$\psi\left(\frac{z}{L}\right) = 1 + 3 \tanh\left(0.75 \frac{z}{L}\right),$$

$$0 \leq \frac{z}{L} \leq 2 \quad (1)$$

When this is inserted into the model, the stability dependent formula for

β that best fits the calculated estimates to those measured is

$$\beta = 4 + 250 \text{ Ri}(5.7) \quad (2)$$

which yields values of β for the individual runs that seem more consistent with the original definition of β than those resulting from the formulation offered in the initial model.⁽⁴⁾

Attention is called to the fact that $\psi(z/L)$ is an empirical fit to a graphical display of values of f_m . In previous work, theoretical arguments were used to propose similar use of the dimensionless wind shear, $\phi_M(z/L)$. When this function is given as⁽⁶⁾

$$\phi_M(z/L) = 1 + 4.7 \frac{z}{L} \quad , \quad (3)$$

the resulting frequency shift is considerably greater than that indicated by Figure 4 in Reference 5.

In Reference 3, the discussion following the Pasquill equation from which σ_z was calculated indicated that the resulting values were functions of downstream time rather than downstream distance. But when the downstream time is calculated by dividing the downstream distance by some $\bar{U}$ [in the case $\bar{U}(z_c)$] and when the power spectrum is one of the usual forms of the nondimensional frequency, $f = nz_c/\bar{U}_c$, i.e., when the frozen field hypothesis is assumed, the Hay-Pasquill equation exhibits σ_z^2 as a function of downstream distances, X , of stability, and of surface roughness, with all T (downstream times) and $\bar{U}_c$ cancelling out.

The integral becomes

$$\sigma_z^2 = \left(\frac{kX}{\log \frac{z_c}{z_o} + 4.7 \frac{z_c}{L}} \right)^2 \int_0^\infty \frac{n S_w(n)}{u_*^2} \left[\frac{\sin(\pi f \psi X / z_c \beta)}{\pi f \psi X / z_c \beta} \right]^2 d \log n \quad (4)$$

where k is van Karman's constant, taken at 0.4, u_* is the friction velocity, and L is the Monin-Obukhov turbulence characteristic length, which indicates the stability. The dimensionless spectrum, $n S_w(n)/u_*^2$ is available from any one of several similar formulations. That used in this program is

$$\frac{n S_w(n)}{u_*^2} = \frac{1.15 F}{1 + 1.5 F^{5/3}} \quad (5)$$

$$F = \frac{n z_c}{(0.46 \psi) \bar{U}_c} \quad (6)$$

SENSITIVITY OF THE MODEL TO CHANGES IN SURFACE ROUGHNESS AND DEPOSITION

Equation (4) shows that σ_z is sensitive to changes of both surface roughness and stability. The Pasquill curves, selected on the basis of wind speed and insolation categories show σ_z at a given X to be a function of stability, only. Therefore, using the previously chosen set of other input parameters:

$$H = 26 \text{ m}$$

$$z_c = 15 \text{ m}$$

$$\beta = 4 + 250 \text{ Ri}(5.7)$$

$$V_d = 3.0 u_*^2 / \bar{U}(2.2)$$

value of the CIE, σ_z , and the suspension ratio, E_x/E_o , were calculated

using a variety of z_o values. (E_x and E_o are the calculated CIE values at a particular downstream distance, X , for depositing and nondepositing tracer, respectively.) It will be noted that although the first three parameters above are held constant for all z_o , the deposition velocity, V_d , changes since u_* changes with z_o .

It will be of interest to compare the contribution to changes in CIE due to changes in σ_z , only, with those due to changes in both σ_z and V_d , both of which are induced by z_o changes. The calculations are exhibited for three tests (Table 1).

TABLE 1. Test Information

Test Number	Ri(5.7 m)	$\bar{U}$ (15 m)
U-78	0.005	5.5 mps
U-79	0.08	3.8 mps
U-82	0.15	3.4 mps

The comparison of calculated CIE values resulting from four z_o values with the measured CIE is shown for the three tests in Figures 1a, 1b, and 1c. The solid line is the calculation for $z_o = 3 \text{ cm}$, which is the accepted value for the Hanford diffusion course. In each case, it is seen that the predicted CIE values for different z_o differ from each other by two orders of magnitude for the shortest and for the longest downwind distances considered.

The differences at each X extremity are indicated by the model to be due to different physical causes. Figures 2a, 2b, 2c show the

a.

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b.

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c.

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FIGURES 1a, b and c. Calculated CIE for $4z_0$ Values and Measured CIE

a.

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b.

Neg 731171-7

c.

Neg 731171-6

FIGURES 2a, b and c. Suspension Ratios for $4z_0$ Values

suspension ratios for the same three tests. These figures show that although the suspension ratios differ little in a geometric sense for the shortest distances from the source, there is considerable geometric difference for the longest distances. The figures also indicate that the differences of suspension ratio from one z_0 to another decreased with increasing stability.

When the model is run for a non-depositing tracer and curves similar to those in Figure 1 are drawn, the calculated CIE values for X greater than 2000 m differ only within a factor of 2 for the four z_0 values, rather than over two orders of magnitude, as in Figure 1. At the short downstream distances, the CIE values for the nondepositing tracer are the same as shown in Figure 1 for the depositing tracer. This suggests that for short downstream distances the elevated source model for stable cases is sensitive to changes in roughness through the changes in turbulence alone, and that for the long downstream distances the sensitivity of the model is due to the changes of deposition caused by the change in turbulence.

Peak values and distances at which they occur do not differ much between depositing and nondepositing cases. (This is as one would expect since deposition is not effective before the majority of plume reaches the ground.) But at greater distances, say 7 kilometers, the nondepositing CIE curves for the Hanford z_0 exceed by a factor of from 4 to 8 (depending on stability) the curve which in-

cludes the effect of deposition and which also compares favorably with the measured CIE values shown.

For larger z_0 , the difference between depositing and nondepositing CIE curves are even greater, up to a factor of 50 for $z_0 = 30$ cm in the neutral case. For smaller z_0 , the difference is less.

Figure 3 shows the values calculated for σ_z for Test U-79 for three values of z_0 compared with Pasquill curves. For the meteorological conditions prevailing during this test period, the E curve applies. The calculated σ_z for $z_0 = 3$ cm approximates this curve. The important point is that the calculated σ_z curves for the three z_0 values are separated from each other by approximately the same distance that separates the Pasquill curves for the different stability categories. Therefore, the effect of roughness, though not verified by observations over differing surface roughnesses, would appear to be important to include in calculating σ_z .

SUMMARY

It would appear from the results discussed above that two significant effects on exposures downwind of continuous releases which have not been accounted for in models in common use are those of roughness and dry deposition. It is indicated that turbulence controlled by roughness will influence vertical dispersion about as much as changing from one Pasquill stability category to the next when z_0 is changed an order of magnitude,

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FIGURE 3. Comparison of Calculated σ_z Values with Pasquill Curves

e.g., from a grass to a brush-covered surface. The distance to the point of maximum surface concentration can vary easily by a factor of two and the concentration can change by more than an order of magnitude at distances both inside of and beyond the point of maximum. Other turbulence characteristics, such as the increase in scale with height cause noticeable but not major variations in the vertical dispersion. For unstable conditions, this may be more serious since, over the range of distances investigated, correlation would not be lost in the vertical motions as they are indicated to be by the slope of 0.5 in the stable σ_z curves on Figure 3. Then variations in the size and strength of

the energy bearing eddies may be felt more significantly. Also, for releases above the constant flux layer, variations in the turbulence structure may have more influence on the surface concentrations. Incomplete modeling of the turbulence structure precludes any further definitive statements about this aspect at this point, however.

Deposition will alter the concentration distribution in a number of ways. First, in connection with varying roughness, the change in u_* changes the deposition velocity, which, in turn, is responsible for a majority of the concentration change at the larger distances from the source. Also, the differences between concentrations of strongly

depositing and nondepositing pollutants were estimated to exceed an order of magnitude. Depending on the type of pollutants and surfaces involved and the interaction between them, the concentration curves could fall anywhere in this range. It is somewhat disconcerting that for the strongly depositing case, assuming the surface to be a perfect sink, it was necessary to use the factor of three in the deposition velocity equation, $v_d = 3.0 u_*^2 / \bar{U}$, in order to obtain a good comparison between the model and observed diffusion data. This would imply that the diffusivity for particulates is considerably larger than for momentum. This indeed may be the case in regions of strong wind shear as suggested by wind tunnel experiments.⁽⁶⁾ In addition, however, the model for deposition reported by Van der Hoven, and used here, assumes that the concentration deficit resulting from the deposition is distributed uniformly through the vertical extent of the plume. Actually, the deficit is felt most strongly in the lower part of the plume.⁽⁷⁾ This would cause lower measured concentrations near the surface than forecast by the model when the appropriate deposition velocity is used and would force a larger factor in the expression for the deposition velocity than appropriate in matching the model to observations.

It is clear from the scoping exercise presented that both modeling im-

provements and more dispersion data for depositing materials over a variety of surface types are needed.

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DRY DEPOSITION PROCESSES

G. A. Sehmel, S. L. Sutter and M. T. Dana

A predictive model for deposition velocities was developed to describe isothermal particle deposition to flat surfaces when gravity is assisting, opposing or not influencing deposition. In order to determine the effect of surface roughness on deposition experimental deposition velocities were determined over gravel surfaces. Comparisons in deposition velocities are made for the flat surface, the gravel surface, and an artificial grass surface. For the grass surface, the results of a field experiment showed that deposition velocities obtained in the laboratory can be applied to similar surfaces in the open atmosphere.

INTRODUCTION

Dry deposition is considered to be a primary means by which a significant portion of pollutant particles and gases are removed from the atmosphere directly onto the various earth surfaces. Even in the case of radioactive debris removal from the stratospheric reservoir, it is gener-

ally accepted that about 20 percent of the total removal is by dry deposition. Despite its great importance, dry deposition is not adequately predictable at the present state of knowledge.

The rate of particle deposition onto a surface has been characterized by a deposition velocity, k , defined as

$$K = \frac{\text{amount deposited/cm}^2 \text{ of surface/sec}}{\text{airborne particle concentration above the surface}} \quad (1)$$

which has units of cm/sec. This deposition velocity is the combined effect of all processes contributing to

particle deposition. For several years we have thought that these overall deposition processes should

be described by at least a three-box model. The conceptual three-box model has now been accepted by the 3rd Chemist-Meteorologist Workshop panel members (January 15-19, 1973) as the best means to describe particle deposition. These boxes consist of:

1. the atmospheric turbulent boundary layer in which the transfer processes are best described by micro-meteorological eddy diffusivity;
2. a layer just above and just within the vegetative canopy or surface elements in which the transfer processes are modified by the presence or structure of the canopy or surface; and
3. a layer (occupied by the canopy or surface elements) in which the final transfer process is best expressed by surface mass transfer coefficients, where the interaction between the surface material and the pollutant is important.

Our research has been mainly directed toward determining the mass transfer resistance at the deposition surface in box 3. Indeed, we have reported our measured deposition velocities as K_1 in which the subscript one refers to the particle concentration at 1 cm from the deposition surface. Nevertheless, we have also initially looked at the total three-box model from a continuum standpoint and have shown that for a wide range of variables the controlling mass transfer resistance must be in the last 1 cm adjacent to the surface. That is, box 3 may contain the most limiting process in the

overall deposition process, but particle mass transfer through box 3 is not adequately predictable at the present state of knowledge.

During 1972, we have been developing deposition models, experimentally determining deposition velocities of monodispersed particles in a wind tunnel, and have initially verified these wind tunnel results in a field experiment. The research has been directed toward quantitative prediction of dry deposition in a manner such that the predictions can be interfaced between the three boxes. Segments of these results will be discussed below.

ISOTHERMAL DEPOSITION MODEL

Particle deposition velocities to isothermal smooth surfaces have been shown by Sehmel⁽¹⁾ to be predictable from effective eddy diffusivities for particles. These particle eddy diffusivities are greater than the eddy diffusivities of air momentum adjacent to a flat deposition surface.

Improved correlations for predicting particle eddy diffusivities were developed⁽²⁻³⁾ from particle deposition velocity data which were experimentally measured for particle diameters smaller than previously reported. For deposition to a dry flat isothermal surface on the wind tunnel floor, the deposition velocities have been measured as a function of air friction velocity, u_* , for particle diameters from 0.1 to 30 μm . These floor deposition velocity data combined with the deposition velocity data to the wind tunnel ceiling were

used to separate out the effect of gravity on particle eddy diffusivities. Thus, we now have three correlations for predicting particle eddy diffusivities when gravity is assisting, opposing or is not influencing the final particle mass transfer at the deposition surface. These three models can lend insight into the different deposition processes of foliar deposition.

The purpose of using effective eddy diffusivities for particles was to establish a procedure for predicting deposition velocities to smooth surfaces. In reality, this eddy diffusivity procedure first predicts the overall mass transfer resistance and subsequently predicts the deposition velocities. Although only the overall mass transfer resistance can be predicted, reasonable estimates of particle eddy diffusivities and comparison with air eddy diffusivities can be made.

Particle deposition near a surface was assumed in the model to occur by diffusion and gravitation effects. The basic model assumptions are that there is a constant flux of particles from a uniform concentration source C_1 at 1 cm, that particle eddy diffusivities can be predicted, that the gravitational force can be described by the terminal settling velocity, and that the particles are completely retained by the surface after contact. Based upon these assumptions, the deposition flux to a floor surface is

$$N = -(\epsilon + D) \frac{dC}{dy} - v_t C \quad (2)$$

Similar equations can be set up for deposition on wall and ceiling sur-

faces. Prediction of the deposition velocity

$$K_1 = -N/C_1 \quad (3)$$

is from a dimensionless integral form of Equation (2) in which

$$-\int_{C_1}^0 \frac{u_* dC}{N + v_t C} = \int_{y^+}^{r^+} \frac{dy^+}{\epsilon/\nu + D/\nu} \quad (4)$$

at 1 cm

The right-hand integral is termed the resistance integral Int . Once Int is predicted, the deposition velocity can be predicted from

$$K_1 = \frac{v_t}{1 - (1/\alpha)} \quad (5)$$

where

$$\alpha = \exp(-v_t \text{Int}/u_*) \quad (6)$$

Deposition velocities to floor, wall, and ceiling surfaces were calculated from Equation (5) using the appropriate form of eddy diffusivity given by Sehmel.⁽²⁾ The resulting deposition velocities are shown in Figure 1 for a nominal velocity of 6.7 m/sec. As expected, the ceiling deposition is least, wall deposition intermediate, and floor deposition the greatest. For particles smaller than about 0.8 μm , the predicted deposition velocities are all the same. Even though the particle eddy diffusivities are small for particles below 0.8 μm , the eddy diffusivities were large enough to predict a greater deposition velocity than for particle transport caused by only

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FIGURE 1. Comparison of Deposition Velocities to Smooth Floor, Wall, and Ceiling Surfaces for $A u_* = 34.1$ cm/sec and $y_0 = 0.004$ cm

Brownian diffusion and gravity settling to a floor surface which is shown by the $u_* = 0$ curve.

Much remains to be learned about particle deposition. Nevertheless, models were developed to estimate particle sampling losses in smooth straight lines as well as the lower limit of deposition velocities expected for atmospheric surfaces. The real practical problem now is determining methods for predicting particle deposition to environmental surfaces in which factors such as

surface roughness, particle agglomeration, impaction, interception, electrical, and phoretic effects are more important in the eddy deposition process.

SURFACE ROUGHNESS

The effect of surface roughness on particle deposition velocities is being determined by experimentally measuring deposition in our aerosol research wind tunnel. Deposition velocities were determined for the deposition of monodispersed particles onto clear chip gravel placed in a uniform layer on the wind tunnel floor. The gravel was minus 5/8 inch in diameter as received. Subsequently, we sieved out all gravel less than 0.185 inch. Particle deposition velocities were determined for monodispersed particle diameters from 0.03 to 6 μm for average wind speeds of 5 and 30 mph. In addition, we have also experimentally extended down to the 0.03 μm particle diameter the measured deposition velocities for a grass surface. This grass⁽⁴⁾ is the 0.7 cm high artificial grass that we had previously used in our first experiments in determining the effect of surface roughness on particle deposition.

The experimental deposition velocities are summarized in Figure 2 for the three different isothermal surfaces that we have investigated. The curves were experimentally determined for particle diameters from 0.03 to 30 μm . Curves outside this range were predicted using the particle eddy diffusivity model.

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FIGURE 2. Comparison of Deposition Velocities for Different Isothermal Surfaces

From a comparison of these curves, we can currently conclude the following:

1. The deposition velocity increases with an increase in air velocity for a constant particle diameter. We are anticipating that the eventual general predictive model for particle deposition will include the use of the friction velocity, u_* , and the roughness height, y_o , in correlating these curves.
2. The curves for the three different surfaces all show a minimum deposition velocity which is caused by the transition in importance of eddy diffusivities and Brownian diffusion on the overall mass transfer process.
3. The particle diameter at the minimum deposition velocity is in the

range of $0.1 \mu\text{m}$, but the location of the minimum cannot be adequately predicted. A tentative conclusion is, that for the minimum deposition velocity, that the largest particle diameter occurs at about $0.5 \mu\text{m}$ for a deposition velocity of about $5 \times 10^{-2} \text{ cm/sec}$. Based upon this conclusion, the particle diameter at the minimum is anticipated to be about $10^{-2} \mu\text{m}$ for a minimum deposition velocity of 10 cm/sec . Since deposition velocities show a general increase with surface roughness, a minimum deposition velocity of 10 cm/sec might be anticipated for the greater roughness of a forested area.

4. Deposition velocities are strongly influenced by the type of surface roughness. We must determine deposition velocity curves over several different types of surface roughnesses in controlled laboratory experiments in order to determine the dependency of deposition velocities upon surface roughness as well as the projected surface area of vegetation, etc.

DIFFUSIOPHORESIS

We have also briefly looked at diffusiophoresis as another force effecting particle deposition. We wet the gravel with water to determine if we could measure the decreased deposition velocity caused by water evaporation. This was done for 0.25 and $1 \mu\text{m}$ diameter particles

at the 5 mph wind speed. The experimental results show that for these conditions the observed deposition velocities are decreased by about a factor of two below the deposition velocities for the dry isothermal gravel.

FIELD TEST

Our research is directed toward predicting deposition velocities which can be used in diffusion-deposition transport models in the open atmosphere. We are determining these deposition velocities in a laboratory wind tunnel under controlled conditions. The control is very important for many reasons, but possibly one of the most important reasons is that deposition velocities can be determined as a function of particle diameter. In contrast for field experiments, sufficient quantities of single sized particles cannot be generated to determine the dependency of deposition velocity on particle diameter. Monodispersed particles if generated under field conditions would be too dilute for measurement at the deposition site. Consequently, the deposition flux would not be greater than the background sensitivity of the analytical technique used to measure the deposited particles.

We have however, measured particle deposition for a polydispersed rhodamine-B aerosol under field conditions in order to compare predicted with total observed deposition. The aerosol used had a mass medium diameter of about $0.7 \mu\text{m}$ but the size

distribution showed that 3 percent of the particles were greater than $4.5 \mu\text{m}$ diameter.

For this field test, the polydispersed aerosol was deposited onto artificial grass identical to that used in the wind tunnel. The experiment was conducted near the base of a 200 m arc tower during a diffusion experiment. For the experiment, a $6 \times 6 \text{ ft}^2$ grass section was placed upwind of the tower at a site which was nearly level and which was free of sagebrush in the upwind direction. During the test period, we measured the air velocity profile, the airborne particle concentration, and the airborne particle size distribution.

The velocity profile was determined from integrating anemometers located at 0.5, 1.0, and 1.5 m heights. The profile indicated a friction velocity u_* of 24 cm/sec and a roughness height y_0 of 0.4 cm.

The prediction of the average deposition velocity required some assumption as to how to interpolate between the wind tunnel data shown in Figure 3 since neither u_* nor y_0 were strictly comparable. Consequently as shown in the figure the assumed curve was used for the field test size distribution. This deposition velocity curve was combined appropriately with the polydispersed aerosol size distribution in order to predict the average deposition velocity of 0.17 cm/sec.

The deposition velocity of the polydispersed aerosol on the central 7×11 inch portion of the grass was determined to be 0.21 cm/sec. This

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FIGURE 3. Comparison of Field and Wind Tunnel Deposition Velocities for "Grass"

experimental point is shown in Figure 3 as the open circle plotted at the mass medium particle diameter. The agreement between the experimental and predicted deposition velocities is considered excellent and confirms that deposition velocities determined in the wind tunnel can be applied to predicting deposition in the open atmosphere.

FUTURE RESEARCH

This deposition research will be initially directed toward determining the particle mass transfer coeffi-

cients in box 3 of the three-box conceptual model. Less is known about this box than any of the three boxes. Experiments in the wind tunnel will be mainly used to determine the interrelationships between deposition velocities and surface roughness. Nevertheless, some of the less controlled field deposition experiments will be conducted to validate and direct the wind tunnel research.

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FIELD EXPERIMENTS ON THE HANFORD DIFFUSION GRID DURING 1972

P. W. Nickola

Five successful major field experiments were carried out on the Hanford field diffusion grid during calendar year 1972. Considerable effort was expended in reconfiguring portions of the grid for compatibility with objectives of the experiments. At this writing, assay and analysis of field samples and data from four of the five tests are not complete.

Three puffs of ^{85}Kr were released in a continuance of the investigation of diffusion under thermally stable atmospheres with extremely low wind speeds--stagnation conditions. Instrumentation problems, and an unfortunate direction of puff drift resulted in essentially no useful data following two of the puff releases. Measurements following the third release reinforced conclusions reached following similar puff releases reported in last years annual report.⁽¹⁾ Maximum exposures (the maximum amount of contaminant a stationary receptor would experience) measured during wind speeds of less than 1 mps and with relatively unsteady directions were approximately the same as those measured under light but steadier winds of 1 to 2 mps. The 1972 puff release into a near calm atmosphere also resulted in a short period (5 sec) maximum concentrations very comparable to those observed with the 1 to 2 mps steadier winds. In short, field measurements to date do not reveal the excessively high exposures or short period concentrations that might intuitively be expected under very light but variable winds.

Some reservation remains concerning the evidence just presented. Although 128 sampling locations were employed in the described tests, the necessity of being prepared for movement of a puff in any direction from its release point left a less dense sampling grid than desired. It is possible that significantly higher concentrations and exposures were in existence than those that were measured. Several steps are underway to remedy this deficiency: (1) The field array is being reconfigured to lessen the chance of a puff "slipping between" samplers; (2) An additional 20 samplers are being added to the array, (3) Puff releases will be replaced by releases of 10 or more minutes duration. These "continuous" releases will also simulate a greater number of practical problems than did the instantaneous puffs.

Following the last of the puff releases in the stagnation condition study, an extensive reorganization of the field diffusion grid was undertaken in preparation for the experiments involving simultaneous release of two particulate tracers (FP 2210*

* FP 2210 is a zinc sulfide fluorescent dry particulate tracer with a specific gravity of 4, and a mass median diameter of 5 microns.

and rhodamine*) and one inert gas tracer (^{85}Kr) from an elevation of 26 m. Sampling arcs, with radii ranging from 200 to 3200 m from the source, included 20 towers with heights up to 62 m. FP 2210 and rhodamine were sampled at 586 locations, and ^{85}Kr at 128 locations. Mean vertical profiles of temperature and wind were monitored at two near-source locations, and an L-shaped array of eight 26 m towers supported one sonic and eight Gill anemometers for monitoring atmospheric turbulence at a mid-field-grid location.

Tracer releases and concurrent turbulence measurements were designed with several objectives in mind. Deposition effects on particulate plumes will be investigated by a comparison of downwind depletion of the depositing particulate plumes with the nondepositing ^{85}Kr . Improved turbulence and deposition measurements coupled with the simultaneous measurements of diffusion will aid in development and testing of theoretically based models of dispersion. Measurements of vertical profiles of tracer concentration will supplement --perhaps revise-- currently widely used estimates of vertical dispersion which are based primarily on deduced vertical profiles.

Four successful field dispersion experiments resulted from the nine times the field course was activated. All successful experiments were carried out during daylight hours. At-

mospheric stability, as indicated by the Richardson number for the layer between 2 and 15 m were -0.22, 0.08, -0.03 and 0.28. The positive stability observed during the fourth test resulted primarily from a shallow temperature inversion caused by a cold snow cover. The temperature lapse rate was negative above the 6-meter elevation.

Plans are to continue this series of experiments during calendar year 1973. Inasmuch as the tests to date have taken place in less stable atmospheres, emphasis will be placed on obtaining at least a few releases during stable atmospheres.

Analysis of the meteorological, turbulence, and dispersion data is not yet complete. In fact, assay of ^{85}Kr and rhodamine samples has not yet been completed. Analysis to date of tracer data has been restricted to FP 2210, as is evidenced by other contributions*,** in this annual report.

Following one experiment, advantage was taken of an unplanned and unusual opportunity. The presence of a ground snow cover during tracer dispersal provided an opportunity to collect "natural" deposition samples. More than 150 snow samples of 1.6 ft² each were skimmed from the surface near sampling locations where

* Rhodamine B was dispersed as a dry particulate with a specific gravity of 1, and a mass median diameter of about 0.8 microns.

* P. W. Nickola. "A Comparison of Measured Exposures with Exposures Computed from Measured and from Pasquill's σ_y and σ_z Values."

** C. E. Elderkin and D. C. Powell. "Profile-Derived Estimation of Crosswind Integrated Exposure from Elevated Releases: Model Revisions and Application."

tracer had been monitored. These samples (currently unassayed) should provide a measured deposition pattern to compare to the computed pattern. Hopefully, the snow samples can be assayed for both FP 2210 and rhodamine.

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A COMPARISON OF MEASURED EXPOSURES WITH EXPOSURES COMPUTED FROM MEASURED AND FROM PASQUILL'S σ_y AND σ_z VALUES

P. W. Nickola

Plume dimensions and exposures (time integrated concentrations) observed during an example field test are presented. The observed dimensions and exposures are compared to exposures computed by means of a commonly used Gaussian plume model. The σ_y and σ_z values used in the model are those prescribed by the Pasquill curves, and those actually observed during the field test. The observed and computed exposures are at considerable variance in all cases. The discrepancies are partially due to lack of consideration of deposition in this Gaussian model.

Field-diffusion Test V1 was carried out on October 26, 1972. Primarily because the plume passed down the center of the field grid during this experiment, this test has to date been examined more carefully than other 1972 field diffusion experiments. Although assay of FP 2210 samples has been completed, further data analysis awaits the assay of rhodamine and ^{85}Kr samples taken during this test. However, analysis to date does present an opportunity to point out some of the problem areas addressed by the 1972 field experiments.

The elevation of dispersal (h) of fluorescent particulate FP 2210 was

26 m. Dispersal continued for a period of 30 minutes beginning at 1000 PST. Richardson number of -0.22 for the 2.1 to 15.2 m layer indicated an unstable atmosphere. Other meteorological parameters observed during the test, and listed in Table 1, show minimal wind direction shear with height and a brisk wind speed.

Figure 1 shows a vertical cross section of E/Q (exposure normalized to unit emission) at a distance of 200 m from the source as observed on the five 33-m towers and 51 ground-level samplers at that distance. Each dot on the figure represents a sampler location. From these exposure observations, measurements of

TABLE 1. Meteorological Parameters Observed During Test V1

	z , Height Above Surface (m)				
	2.1	15.2	30.5	61.0	122
$\bar{u}$ (mps)					
Mean Wind Speed	4.0	5.5	5.8	5.8	6.3
$\bar{\theta}$ (degrees)					
Mean Wind Direction	295	288	291	289	292
σ_{θ} (degrees)					
Direction Standard Deviation	13.1	10.3	9.5	7.5	6.2
T ($^{\circ}F$)					
Temperature	55.5	52.5	52.1	51.1	50.3

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FIGURE 1. Vertical Cross Section of Exposure at 200 m from Source for Test V1

$\bar{z}$ (the mean elevation of the vertical exposure distribution) and of σ_y and σ_z (the horizontal and vertical distributions of tracer in the plume) can be computed. These computations result in the "observed" values listed in Table 2.

The Gaussian plume model specifies that any slice through a plume in the y (horizontal) or z (vertical) direction results in the same σ value. Although the five points on all but one abscissa are too few to use confidently (the tabled σ_y was derived from the 1.5-m samplers), the 17 measurements at each tower location provide the means for five estimates of σ_z . The five σ_z estimates resulting from data plotted on Figure 1 are 9.2, 9.4, 8.7, 9.0 and 7.3 m. A σ_z based on the crosswind sums for each elevation is 9.1 m.

Table 2 also lists the $\bar{z}$, σ_y and σ_z values observed on vertical planes at distances beyond 200 m. Figure 2 presents the crosswind sums of tower

TABLE 2. Parameters Specifying Plume Elevation and Plume Dimensions for Field Test V1

	Distance from Source, m			
	200	840	1640	3230
$\bar{Z}$ (b)	21.3 ^(a)	25.9	34.4	35.9
z_m (c)	14	11	22	43
σ_y Values:				
Observed	26.3	102	191	341
Pasquill A	45	175	320	580
Pasquill C	24	91	170	320
Pasquill D	17	62	113	212
σ_z Values:				
Observed	9.1	14.4	19.3	20.5
Pasquill A	32	330	>1000	>>1000
Pasquill C	15	58	100	175
Pasquill D	9	28	45	69

- a. Units of all data in body of table are meters
 b. $\bar{Z}$ is mean elevation of vertical exposure distribution
 c. z_m is elevation of maximum observed exposure

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FIGURE 2. Vertical Profiles of Crosswind Sums of Tower Exposure ($E\Delta y/Q$) Versus Height

exposures ($E\Delta y/Q$ in sec/m^2), and, by ignoring the extrapolated portions of the curves, indicates the tower heights at each distance from the source, x .

There are several techniques available for determining the Pasquill stability category. Use of Test V1 meteorology and USAEC Safety Guide 23⁽¹⁾ thermal stratification specifies Pasquill class A. Use of Pasquill's key to stabilities based on relatively simple weather observations^(2,3,4) specifies class C. Use of σ_θ classification^(1,3) specifies Pasquill class D. The σ_y and σ_z values associated with these classes are listed on Table 2. Note that

with but one exception, the observed σ_z values are smaller than the Pasquill values.

Figures 3 and 4 present observed and computed $E_p \bar{u}/Q$ values (center-line exposure values normalized to source strength and observed source height wind speed) for height of $z = 0$ and $h = 26$ m based on the Table 2 data. (The Figure 4 "observed" curve is drawn to the peak observed $E \bar{u}/Q$ regardless of height. The appropriate height is listed beside each data point. The x 's below each point indicate $E \bar{u}/Q$ values observed at $z = 26$ m). Although the observed center-line exposures do not agree well with any of the Pasquill or observed

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FIGURE 3. Observed and Computed Plume Centerline Exposures at Ground Level

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FIGURE 4. Observed and Computed Plume Axial Exposures

curves, Pasquill A provides the closest estimate for the ground level exposures, while Pasquill C is the best choice for the aloft plume.

It is not within the scope of this report to detail all the problem areas in which experimental data of the nature just presented has application, nor to detail many of the possible strengths or weaknesses of the data collection technique. The approach which follows is to present a series of brief comments on conclusions which may be drawn, and further problems which may have solution through use of these data and similar data remaining to be analyzed.

- If the extrapolations above the tops of towers are reasonable, the Gaussian plume model appears well founded with regard to obtaining reasonable estimates of σ_z irrespective of where a vertical sampling slice through the plume is made. Computations of exposure can therefore be made from field data including only one vertical profile of concentration or exposure.
- The extrapolations above tower sampling heights well may have a significant influence on the σ_z values computed. The error should be less for the 200 m distance. It is at this location that the "observed" and "computed from observed σ_y and σ_z " exposures are in best agreement for the aloft plume (Figure 4). Tracer releases under more stable atmospheres should result in better definition of σ_z , and hence more confident computations of exposure in the checking of the Gaussian model.
- Isopleths of Figure 1, the profiles of Figure 2 and $\bar{z}$ and z_m values of Table 2 all suggest a depression in elevation of the plume axis at 200 m. A faulty dispersal technique (fallout) is one possible answer. The high "observed" point at 200 m (with respect to the "computed from observed" curve) on Figure 3 could also be due to dispersal fallout near the source. Future assay of samples of the simultaneously dispersed fine particulate rhodamine and the inert gas krypton should help resolve the question of fallout. Dispersal techniques with these latter two tracers are less open to question.
- The observed σ_z values may well be decreased by loss of tracer at the surface through deposition. Figure 2 suggests a rapid drop of tracer concentration at elevations less than 5 meters for distances beyond 200 m. If tracer reflection were taking place at the surface, an increase in exposure at near-surface locations could be expected rather than a decrease.
- If tracer were not being depleted from the plume and the wind speed were presumed to be invariant with height, the areas under the curves on Figure 2 would be identical. This is obviously not the case. Plume depletion by deposition may explain the discrepancy. The possibility of grossly improper extrapolations above tower heights is an alternative explanation of

the unequal areas. Very superficial considerations of other factors (such as different sampling efficiencies for different sampler flow rates, and the influence of higher wind speeds aloft on the flux of material transported) result in an estimate of about 60% of the tracer depleted between the 200 and 800 m sampling arcs. Deposition of this magnitude has been suggested by previous Hanford field work.^(5,6)

- The Pasquill stability class selected varies greatly depending on parameter(s) used in the selection.
- Neither Pasquill nor "observed" σ_y and σ_z values when employed with the Gaussian plume model resulted in a reasonable computation of peak exposures. Likely one of the most significant discrepancies in the modelling is the lack of consideration of deposition.

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APPLICATION OF THE ^{85}Kr MONITORING SYSTEM TO METEOROLOGICAL
DIFFUSION-DEPOSITION EXPERIMENTS

J. D. Ludwick and P. W. Nickola

The ^{85}Kr field tracer system has been modified by the incorporation of additional field sensors, by the addition of improved techniques and instrumentation for tracer storage and dispersal, and by the incorporation of recording procedures which permit rapid data processing. To date, the improved tracer system has been employed during four field diffusion-deposition experiments.

Expanded field monitoring and an automated krypton sensing system are now in a highly developed stage for information accumulation and data processing. Recently four field tests were conducted using the continuous release of ^{85}Kr gas as a tracer. The information from each of these tests was accumulated by two 4096 Packard memories and subsequently transferred to the DEC-type tapes. A computer program is now being written to facilitate information transfer from these DEC-tapes to the various functions available on the 1108 master computer. The completion of this final phase is expected before the end of April, 1973. At this time, only a direct digital information printout is planned which identifies the instantaneous krypton concentration at each detector at any particular time interval. As appropriate computer programs are written for this diffusion study, experimental data from krypton experiments can be immediately processed and permanently cataloged in a library of DEC-tapes.

The present series of field tests is designed primarily to investigate

deposition associated with particulate tracers/contaminants. In these tests krypton and two particulate tracers, ZnS and Rhodamine B, are simultaneously released from the same elevation. The intent is to compare exposures of the depositing particulates with the nondepositing inert gas krypton at successive downwind sampling arcs. The anticipated depletion of the particulates can be ascribed to deposition. A differential deposition of the particulate tracers is also expected on the basis of the fact that the Rhodamine particles are less dense and are of smaller size than the ZnS.

In order to facilitate dispersion of the tracer gas ^{85}Kr in a continuous manner, a high pressure apparatus was constructed to aid in the transfer of 20 Ci sources of krypton from Oak Ridge shipping containers to one liter stainless steel pressure bottles. Radioactive krypton is vacuum transferred from the shipping containers into a liquid nitrogen cooled steel U-tube. Argon gas, at high pressure, sweeps the radioisotope from the U-tube section into the stainless cylinder. The cylinder is

then further pressurized with argon gas to 600 psig. During and subsequent to loading, the cylinder is shielded by placing it in an aluminum container which allows 2 cm of lead shot on all sides. The amount of shielding weight is kept to a minimum consistent with safety. The overall weight of such a source is about 80 pounds and the surface dose is 200 mR.

During release, the sources were located at the base of the emission tower and a 1/8 inch copper tube conveyed the gas to the 26-m level, the point of particulate generation. Flow control of the krypton was maintained by a special low flow digital readout regulator. This allowed relatively even dispersal of the 20

Ci sources over a 30 minute test period.

The counting information from each of the 128 detectors was integrated by the computer memory over 10 second intervals. Each memory has 32 such intervals before the entire 4096 memory is full. The total time to fill the memory is, then, 320 seconds or about 5-1/2 minutes. Since each DEC-tape is capable of storing 16 dumps from this memory, each one can accumulate almost 1-1/2 hours of experimental data. This was normally satisfactory for a test. The electronic data handling system can, of course, collect data continuously for any length of field test at integrating time intervals as short as 0.1 second; however, several DEC-tapes would be required.

SPECTRAL TRANSFER FUNCTIONS FOR A THREE-COMPONENT SONIC ANEMOMETER

T. W. Horst

The transfer functions which describe the effect of line averaging and path separation on the response of a three-component sonic anemometer have been computed specifically for the Kaijo-Denki PAT 311 and have been used to facilitate the design of a sensor specifically for use on an aircraft. These computations differ from the previous work of Kaimal, et al. (1968), by accounting for the two parallel paths which compose each measurement axis and for the vertical separation of the two horizontal axes.

A sonic anemometer measures the speed of the wind by transmitting sound waves between a pair of acoustic transducers. The wind component

along the sonic path, defined by the line connecting the transducer pair, is determined from the difference in

transit time for sound waves transmitted in opposite directions along that path. In order to measure all three components of the wind vector, it is necessary to have three non-parallel, noncoplanar sonic paths which do not interfere with each other either in the transmission and reception of the sound waves or in exposure to the wind field. The Kaijo-Denki PAT 311 array, used for tower measurements at Hanford, is shown in Figure 1. The W component axis measures the vertical wind, and the A and B axes measure two independent

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FIGURE 1. Dimensions Relevant to Calculating the Response of a Three-Component Sonic Anemometer

components of the horizontal wind which are 120° apart and from which are calculated the orthogonal components of the horizontal wind. This design works well for moderate wind speeds and for gusts whose direction α , relative to the bisector of the angle between the A and B axes, is $\leq 45^\circ$. A different array is proposed for aircraft use in the accompanying report, "Aircraft Instrumentation for the Measurement of Turbulent Wind Fluctuations." It utilizes two axes at right angles to each other and to the aircraft flight direction in order to measure the lateral and vertical components of the wind and a third, nonorthogonal axis in the horizontal plane for the determination of the longitudinal wind component. It is designed for use in the presence of a high mean wind with very small variations in the wind direction.

The response of these arrays is a function of the line averaging over each individual sound path, the separations and relative orientations of the three paths, and of the eddy size. Kaimal, et al.⁽¹⁾ has calculated a set of transfer functions, the ratio of the measured to the actual one-dimensional power spectra, for the array shown in Figure 1. For the purpose of the computations it was assumed that each measurement axis or component of the anemometer averaged the wind over a single path of length l and that the centers, denoted by the vectors $\vec{x}_A$ and $\vec{x}_B$, of the two horizontal axes were in the same horizontal plane, separated by a distance d . In actuality each measurement axis of the Kaijo-Denki anemometer

has two parallel paths of length l which are separated by a distance p , and the two horizontal axes are additionally separated in the vertical by a distance h . Horst⁽²⁾ has included these additional geometric details in the calculations and found changes in the predictions of high frequency response which could be important for interpreting data in the inertial subrange portion of the spectrum.

These same computational methods were used to optimize the design of the configuration proposed for aircraft use. The response to eddies much larger than the dimensions of the sensor should be perfect and thus the transfer function T should equal one for large wavelengths. Further, the spatial averaging of the sonic anemometer sharply attenuates the response to eddies whose size is much smaller than the sensor, giving a limiting value of $T = 0$ for small wavelengths (Figure 2). For eddy sizes comparable to the sensor dimensions the transfer function is a complicated function of sensor geometry. A design was sought which had a transfer function close to unity for wavelengths as small as possible and which decreased monotonically to the zero limit at very small wavelengths. Resonances in the transfer function, especially those greater than unity, were thought to be undesirable. After trying numerous variations, it was found that this could best be achieved with the original geometry of the Kaijo-Denki anemometer (Figure 1), simply using it with the mean wind or flight direction at an angle of

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FIGURE 2. Spectral Transfer Functions for the Aircraft Sonic Anemometer

30° to the B axis and 90° to the A axis, i.e. $\alpha = 30^\circ$. The resulting transfer functions are shown in Figure 2 for the longitudinal wind component T_1 derived from the measurements along both the A and B axes and for the lateral and vertical wind components, T_2 and T_3 , measured by the A and W axes, respectively. The transfer functions are shown as a function of both wavelength λ and, assuming an aircraft speed of 50 m sec^{-1} , frequency n .

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AIRCRAFT INSTRUMENTATION FOR THE MEASUREMENT
OF TURBULENT WIND FLUCTUATIONS

T. W. Horst and K. M. Busness*

Instrumentation is being developed for the measurement of the three orthogonal components of the wind from a light, twin-engine aircraft. Aircraft orientation is measured by vertical and directional gyroscopes and aircraft velocity relative to the ground is determined from an orthogonal set of three accelerometers. The velocity of the wind relative to the aircraft will be measured with a nose-boom-mounted sonic anemometer. The aircraft motion system has been flight tested and is operational. The sonic anemometer sensor is currently being designed. It will be constructed and the complete system will be flight tested during 1973.

A basic understanding of atmospheric diffusion requires knowledge of the structure of the turbulent eddies which effect the diffusion. Currently there is a great deal more known about turbulence in the lowest tens of meters of the atmosphere than in the remaining 90 percent of the planetary boundary layer. There are two reasons for this: first, this constant flux layer adjacent to the surface is less complex than the entire boundary layer, and second, it is much more accessible to measurements from towers. Since diffusion to the distances of interest to the AEC is effected to a large extent above the surface layer, a capability is being developed at PNL for the study of the structure of the planetary boundary layer from an instrumented aircraft.

The measurement of turbulent wind fluctuations from an aircraft requires the measurement, separately, of the wind velocity relative to the aircraft and of the velocity of the aircraft

relative to the ground. The required velocity of the air relative to the ground is then the sum of these two measurements. Aircraft motions are being determined with a vertical gyroscope measuring pitch and roll, a vertically stabilized, orthogonal set of three accelerometers measuring vertical, fore-and-aft and lateral acceleration, and a directional gyroscope measuring aircraft heading. These six measurements uniquely specify the aircraft orientation and velocity at any time. It is planned to measure the three components of the wind relative to the aircraft with a sonic anemometer mounted on a boom projecting forward of the aircraft nose into undisturbed air.

AIRCRAFT MOTION SYSTEM

The components of the aircraft motion system, manufactured by Humphrey, Inc., San Diego, have been successfully integrated and flight tested on the Cessna 411. This followed the design, construction and laboratory

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testing of control circuitry which supplies the required power to the various components and assures that the proper sequencing of gyroscope control functions is followed. Preparation was made for future data analysis by calibrating the pitch, roll and yaw outputs with known rotations of the system in the laboratory and by developing the relationships by which the direction cosines, which best describe the aircraft orientation, can be calculated from the recorded outputs. Frequency and probability distributions were calculated for test flight data to assure proper sampling and recording of the data and to determine the magnitudes and dominant frequencies of aircraft motions. A sample of these calculations is utilized below to discuss the measurement of the vertical wind component.

Figure 1 displays some results from an aircraft test flight at a height of 300 m in the early afternoon. The logarithmic power spectra are shown for the true vertical wind and for several components of the aircraft motion which must be removed from the measured vertical wind. The level of turbulence, as indicated by a measured dissipation ϵ of $16 \text{ cm}^2 \text{ sec}^{-3}$, could be classified as moderate.⁽¹⁾ Curve A is the inertial subrange portion of the power spectrum of the true vertical wind, as determined by the measured dissipation. Note that this measurement could be made with simple instrumentation and without accounting for aircraft motions, because at wavelengths λ in the vicinity of 100 m the aircraft motions are an order of magnitude smaller than the turbulence.

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FIGURE 1. Logarithmic Power Spectra of the Vertical Wind and of Various Components of the Aircraft Motion Which Complicate Its Measurement

At wavelengths greater than 1000 m, the extension of curve A would have a slope of +1, and a peak might occur in the range 200 to 800 m.

Figure 1 shows that aircraft motions cannot be ignored for wavelengths greater than 500 m. Curves B, C, and D are several components of this motion. The error due to the vertical aircraft velocity, curve B, is determined by integrating the measured vertical acceleration. This is the simplest error to visualize, but

several others must be considered due to the pitch and roll of the aircraft. These angles are small enough (the measured standard deviation were 0.68 and 1.24°, respectively) that the resulting mixing of orthogonal turbulent wind components is negligible.⁽²⁾ The forward speed of the aircraft V is large enough however that its contribution to the measured vertical wind due to the forward pitch of the aircraft must be accounted for (curve C). The similarity between curves B and C at the larger wavelengths is a result of the fact that these two effects cancel to some extent and it is only their difference which produces an error in the vertical wind measured from the aircraft. Finally, the pitching motion of the aircraft swings the anemometer up and down relative to the point at which the vertical aircraft acceleration (or velocity) is measured. The spectrum of this additional motion, shown in curve D, demonstrates that this factor can probably be neglected.

AIRCRAFT ANEMOMETER

The three components of the wind relative to the aircraft will be measured with the three component PAT 311 sonic anemometer manufactured by Kaijo-Denki, Tokyo, Japan. This requires that the configuration of the sensor array be altered for best performance in the presence of a high ($\sim 50 \text{ m sec}^{-1}$) mean wind speed and a small variation in mean wind direc-

tion. Two measurement axes will be placed at right angles to each other and to the aircraft flight path to measure the lateral and vertical components of the wind. Thus two components of the wind are determined with a minimum of contamination from the forward speed of the aircraft. The component of the wind along the aircraft axis will be measured with a nonorthogonal axis which is in the horizontal plane and is inclined to the flight path. The longitudinal wind component is then calculated from the data recorded from both this inclined axis and the lateral measurement axis.

Detailed studies were made of the anemometer response as a function of the geometric arrangement of its axes* in order to determine how best to implement the above design. The resulting configuration is currently being translated into detailed blueprints for construction, aircraft mounting and flight testing during 1973.

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STUDIES OF WASHOUT FROM THE PLUME
OF A COAL-FIRED POWER PLANT*

M. Terry Dana, Jeremy M. Hales and M. A. Wolf

Washout of sulfur compounds from the plume of the Centralia power plant was studied experimentally. In this study precipitation was collected at various locations beneath the plume and analyzed chemically for SO₂, hydrogen ion, and SO₄²⁻. Five field tests were completed during this series of experiments.

INTRODUCTION

Past field studies of this ongoing project have been centered at the Keystone Plant in Western Pennsylvania and at Quillayute, Washington. The controlled SO₂ release studies at Quillayute were conducted to assess individual gas scavenging mechanisms in a low-background, controlled-release situation. The next stage of this project was to apply the findings from the Quillayute experiments to an actual power plant operating in a low-background environment. This objective has been completed during the Centralia study, which was carried out during early 1972. Five separate tests were completed during this series.

The Centralia Steam Plant is located in the Hanaford Valley near Centralia, Washington. Currently in the final stages of construction, only one of its two 700 megawatt units was operating during the time of the study. This plant offers the opportunity to study washout from

the plume of a moderate-size stack in an environment that possesses relatively little background contamination. Figure 1 shows the arrangement of precipitation collection locations and instrumentation sites relative to the position of the then active plant stack. The three outer sampling lines, arranged along state and county roads, are at distances from the stack of about 3, 7, and 11 km.

Field experiments were initiated by the release of a pilot balloon from the control center located at the plant site. The resulting mean wind measurement was used to establish placement of the precipitation collectors on the sampling lines. During the experiments, rawinsonde flights were conducted to record temperature, humidity, and winds in the vicinity of the plume. Also, rain-drop-size spectra samples were recorded periodically, and precipitation rates were recorded continuously at the control center.

A 30.5 m portable tower was located downwind of the plant stack at a distance of about 3 km. A Gill three-component anemometer on this

* This project is funded by the Environmental Protection Agency under a related services agreement with the Atomic Energy Commission.

tower provided continuous mean and turbulent wind velocity data. The location of the tower, on a 134 m elevation ridge, placed the anemometer approximately 40 m below the top of the plant's 143 m stacks.

Each of the 2 or 3 rain collectors at a collection site (approximately 60 collection sites were used for each run) consisted of a 20 cm diameter funnel, supported by a

laboratory ring support and steel rod. Attached to the funnel was a 500 ml polyethylene bottle, of sufficient capacity for 1.5 cm of rainfall. At every site, one collector containing tetrachloromercurate (TCM) sampled for rainborne SO_2 and a separate collector containing hydrogen peroxide sampled for rainborne sulfate. In the latter sample, both SO_2 and $\text{SO}_4^{=}$ were retained as $\text{SO}_4^{=}$.

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FIGURE 1. The Centralia Sampling Network

At every third collection site, an additional collector sampled untreated rain water for pH measurement and for trace metal analysis. SO_2 concentrations in air were measured at these locations also, using portable bubbler samplers.

The SO_2 , $\text{SO}_4^{=}$, and pH analyses were accomplished at the Battelle mobile laboratory, located in Centralia. Two Technicon Autoanalyzers were used for analysis of SO_2 , through the previously described procedure.⁽¹⁾ A third Technicon was set up for automated analysis of sulfate, following the methylthymol blue method of Lazrus, et. al.⁽²⁾ The accuracy limits have been established as ± 2 percent at 10 micromole ℓ^{-1} for SO_2 , and ± 4 percent at 5 mg ℓ^{-1} for $\text{SO}_4^{=}$.

Trace metals in the rain are being measured to determine possible catalytic activity for SO_2 oxidation. The rainwater samples were acidified, frozen and returned to Battelle in Richland for trace metal analysis using a Perkin-Elmer 306 atomic absorption spectrophotometer.

Results of the Centralia study will be analyzed using the revers

ible washout model developed during the previous year. Although quantitative calculations have not been carried out at present, the data collected during this study appear to conform to earlier predictions based on this model. The measurements of SO_2 and $\text{SO}_4^{=}$ in rain showed the presence of the plume vividly under all circumstances, and background appears to have been only a minor problem. In addition to providing a final check on the SO_2 washout theory, it is expected that analysis of these data will reveal substantial information concerning $\text{SO}_4^{=}$ formation and washout in plumes.

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THE REDISTRIBUTION OF A GAS PLUME CAUSED
BY REVERSIBLE WASHOUT*

W. G. N. Slinn**

An analytical study is made of the influence on a gas plume of reversible scavenging by rain. The result predicts that at large time, a plume will be washed downward at a rate $H'J_0$, where J_0 is the rainfall rate and H' is the dimensionless Henry's law constant. Results are shown for the specific case of an SO_2 plume.

In a recent report, Hales⁽¹⁾ has called attention to the possibility that a gas plume could become redistributed in the atmosphere, under the action of reversible scavenging by rain. Thus, as a raindrop traverses a plume, the gas concentration in a drop first approaches a concentration saturated with respect to the plume. If no chemical conversion of the gas occurs within the drop, then it can emerge from the plume with a concentration, supersaturated with respect to ambient conditions, and desorption of the gas may occur. This can result in an effective lowering of the plume and thereby an increased dosage to low-level receptors. The purpose of this note is to attempt to predict the rate at which this sorption-desorption effect might lower a gas plume.

The mathematical description of the complete problem is rather formidable. If we assume that the stack

exit is high enough and the atmosphere is stable enough so that a convective diffusion equation is adequate, and if we neglect longitudinal diffusion compared with transport, and further, if we take the diffusivities to be constant and that there is negligible wind shear, then the convective diffusion equation becomes

$$\frac{\partial \chi}{\partial t} + u \frac{\partial \chi}{\partial x} = D_y \frac{\partial^2 \chi}{\partial y^2} + D_z \frac{\partial^2 \chi}{\partial z^2} + \beta C - \alpha \chi, \quad (1)$$

where χ is the concentration of the gas in the air (gas-phase concentration) and C is the concentration in the rain (liquid-phase concentration). The other symbols have their usual meaning and βC represents the rate at which the gas is desorbed from the rain back to the air, while $-\alpha \chi$ is the usual washout term. Hales has related α and β to physical parameters (drop size, mass transfer coefficient, and Henry's law constant) and these results will be used later in this paper. The companion equation to (1), describing the concentration of the gas in the rain is

$$-v \frac{\partial C}{\partial z} = \alpha \chi - \beta C, \quad (2)$$

* An extended version of this report has been submitted for publication in *Atmospheric Environment*.

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where γ is related to the magnitude of the rain drop's fall velocity.

Although we have made progress toward the solution of the general equations subject to realistic boundary conditions, we shall report here only on the simpler problem for which diffusion and horizontal transport of the plume can be ignored. In this case, (1) and (2) simplify to

$$\frac{\partial \chi}{\partial t} = \beta C - \alpha \chi, \quad (3)$$

and

$$-\gamma \frac{\partial C}{\partial z} = \alpha \chi - \beta C. \quad (4)$$

If C is eliminated between (3) and (4) we obtain an equation describing only the plume:

$$\frac{\partial \chi}{\partial t} = \frac{\alpha \gamma}{\beta} \frac{\partial \chi}{\partial z} + \frac{\gamma}{\beta} \frac{\partial^2 \chi}{\partial z^2 \partial t}. \quad (5)$$

To solve (5), a Laplace transform (parameter s) is taken in time. Solving the resulting ordinary differential equation in z , subject to the single boundary condition $\hat{\chi}(z \rightarrow \infty, s) = 0$, gives

$$\hat{\chi}(z, s) = \frac{\chi_0(z, 0)}{s + \alpha} + \frac{\alpha \beta}{\gamma} \frac{e^{Az}}{(s + \alpha)^2} \int_z^\infty \chi_0 e^{-A\zeta} d\zeta, \quad (6)$$

where $A = \frac{\beta}{\gamma} \frac{s}{\alpha + s}$. Inverting the first term on the rhs of (6) is trivial. It leads to an exponential decay of the initial plume profile: $\chi_0(z, 0) \exp \{-\alpha t\}$.

The second term is a little more difficult. The integral to be evaluated is the integral on the Bromwich contour:

$$I = \frac{1}{2\pi i} \int_{B.C.} \frac{e^{A(z-\zeta)} e^{st}}{(s + \alpha)^2} ds. \quad (7)$$

Point $-\alpha$ in the complex s plane is an essential singularity because of the s dependence of A . If near $s = -\alpha$ we take $s = \epsilon - \alpha$, then the expansion of the integrand for small ϵ leads to

$$\frac{e^{A(z-\zeta)+st}}{(s + \alpha)^2} = \frac{1}{\epsilon^2} \exp \left\{ \frac{-\beta}{\gamma} \eta - \alpha t \right\} \left[1 + \epsilon t + \frac{(\epsilon t)^2}{2!} + \dots \right] \cdot \left[1 - \frac{\eta \alpha \beta}{\gamma \epsilon} + \frac{1}{2!} \left(\frac{\eta \alpha \beta}{\gamma \epsilon} \right)^2 - + \dots \right], \quad (8)$$

where $\eta = \zeta - z$. By collecting all terms of the coefficient of ϵ^{-1} , and identifying the resulting series as that for a first order modified Bessel function, we obtain the residue

$$\frac{2t}{q} \exp \left\{ \frac{-\alpha \beta}{\gamma} \eta - \alpha t \right\} I_1(q), \quad (9)$$

where $q^2 = 4\alpha t \frac{\beta}{\gamma} (\zeta - z)$. The solution is then obtained from (6) to be

$$\chi(z, t) = \chi_0(z) e^{-\alpha t} + \frac{1}{2} e^{-\alpha t} \int_z^\infty d\zeta \chi_0 \frac{e^{-\frac{\alpha}{\beta}(\zeta-z)}}{(\zeta-z)} q I_1(q). \quad (10)$$

Into (10) can be substituted any initial plume concentration, including, for example, a plume which has diffused to any realistic profile downwind of the stack.

A particular case for (10) is quite informative. If initially the plume is a delta function at the height h , then subsequently, $\chi = 0$ for $z > h$, and for $z \leq h$,

$$\chi(z,t) = \delta(z - h)e^{-\alpha t} + 1/2 \exp\left[-\alpha t - \frac{\beta}{\gamma}(h - z)\right] \frac{q I_1(q)}{(h - z)}, \quad (11)$$

with $q^2 = 4\alpha t \frac{\beta}{\gamma}(h - z)$. Plots of this solution will be given later. The large q behavior of (11) can be found using the known asymptotic expansion of $I_1(q)$ and leads to

$$\chi(z,t) \sim \frac{1}{(h - z)} \left(\frac{q}{8\pi}\right)^{1/2} \exp\left\{-\left[(\alpha t)^{1/2} - \sqrt{\frac{\beta}{\gamma}}(h - z)^{1/2}\right]^2\right\} \left(1 - \frac{3}{8q} + \dots\right). \quad (12)$$

To obtain the concentration in the rain, we return to the original equation, (4), and obtain

$$C(z,t) = \frac{\alpha}{\gamma} e^{\frac{\beta}{\gamma} z} \int_z^\infty \chi(\zeta,t) e^{-\frac{\beta}{\gamma} \zeta} d\zeta. \quad (13)$$

In particular, if we substitute the Green's function (11) into (13), and utilize known integrals of Bessel functions, we obtain for $z \leq h$,

$$C(z,t) = \frac{\alpha}{\gamma} I_0(q) \exp\left[-\alpha t - \frac{\beta}{\gamma}(h - z)\right]. \quad (14)$$

It is of interest to obtain an estimate of the rate at which the plume is "washed down" by precipitation. One way to evaluate this is

to determine the centroid of the plume as a function of time. If the centroid is taken about the top of the stack, then for the case of a delta function initial plume (11) yields

$$\begin{aligned} \langle z \rangle &= -\frac{e^{-\alpha t}}{2} \int_0^\infty d\xi e^{-\frac{\beta}{\gamma} \xi} q I_1(q) \\ &= -\frac{\alpha \gamma}{\beta} t. \end{aligned} \quad (15)$$

The "washdown" velocity of the plume is then

$$\frac{d\langle z \rangle}{dt} = -\frac{\alpha \gamma}{\beta}. \quad (16)$$

We now turn to specific applications of these results. From Hales,⁽¹⁾ if we characterize the raindrop size distribution only by a mean drop radius a , mean terminal speed V_t (> 0) and rainfall rate J_o , then

$$\begin{aligned} \alpha &= \frac{3J_o K}{a \bar{C} V_t}, \quad \beta = \frac{\alpha}{H'}, \\ \gamma &= J_o, \end{aligned} \quad (17)$$

where K is the mass transfer coefficient in the gas phase, $\bar{C}$ is the bulk gas-phase concentration in moles cm^{-3} , and H' is the dimensionless Henry's law constant (both liquid-phase concentration C_ℓ , and gas-phase concentration C_g , expressed in moles cm^{-3} , and $C_\ell = H' C_g$). Substituting (17) into (16) gives the simple result for the plume's "washdown" velocity,

$$\frac{d\langle z \rangle}{dt} = -J_o H'. \quad (18)$$

For example, for an SO_2 plume exposed to a rainfall rate of 10^{-4} cm sec ,

and $H' = 3.3 \times 10^3$ (Hales, personal communication) then the "washdown" velocity is only $1/3 \text{ cm sec}^{-1}$. This is consistent with the results shown in Figure 1 which demonstrate that after 10^4 seconds, the plume has fallen about 30 meters. That (18) is independent of the mass transfer coefficient, results from our taking the centroid about h , the initial plume height.

A word of caution is in order about the application of (18) to other gases. The source of uncertainty is in the use of handbook values for H' obtained at high concentrations of the gases. In some cases, e.g., N_2O , CH_4 , tritium compounds, krypton, etc., for which simple solutions of the gases are formed, this is probably acceptable and (18)

predicts that such plumes would fall orders of magnitude more slowly than an SO_2 plume. For other gases which are dissolved and subject to reversible hydration and dissociation, such as SO_2 , CO_2 , I_2 , and NH_3 , H' is strongly dependent on the concentration, and adequate data is not available. The value of H' for SO_2 used above, is for a concentration of about 0.5 ppm. If the trend for the ratio of H' for SO_2 and NH_3 that is revealed at high concentrations does continue to low concentration, then for the same rainfall rate, the washdown velocity of an NH_3 plume is seen from (18) to be an order of magnitude larger than that for an SO_2 plume. This could be significant for the assessment of the environmental impact of an NH_3 plume.

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FIGURE 1. The Redistribution of an SO_2 Plume Caused by Washout. Data used in this numerical example of Equation (11) are: $J_0 = 10^{-4} \text{ cm sec}^{-1}$, $v_t = 300 \text{ cm sec}^{-1}$, $a = 0.03 \text{ cm}$, $K_y = 8.2 \times 10^{-4} \text{ moles cm}^{-2} \text{ sec}^{-1}$, $H' = 3.3 \times 10^3$, and $\bar{C} = 1 \text{ mole}/(22.4 \text{ l})$.

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SNOW SCAVENGING OF LEAD PARTICULATES

R. N. Lee

A short field study has been conducted on the Hanford Reservation to evaluate the effectiveness of snow scavenging for the removal of lead aerosols generated by automotive exhaust. This study employed a grid network of snow samplers in the vicinity of a highway, with subsequent analysis of the collected snow for lead using an atomic absorption technique. The results indicate that under appropriate conditions snow is an effective scavenger of particles of this type.

INTRODUCTION

In 1962, U.S. consumption of the gasoline additives tetraethyl- and tetramethyl-lead was equivalent to 170,000 tons of lead. By 1970, lead consumption in automobile fuel had increased to more than 239,000 tons. Since the bulk of this lead (~57%) is released to the atmosphere following combustion, mounting concern has been expressed as to the relationship between auto exhaust and blood lead levels found in certain segments of the population.⁽¹⁾ While a substantial amount of work has appeared which relates airborne lead concentration to traffic volume and proximity to a highway,⁽²⁾ explores lead uptake by vegetation along well-traveled roadways⁽³⁾ and examines the size distribution of lead aerosol

particles;⁽⁴⁾ very little appears to have been done to study the efficiency of lead aerosol removal by precipitation. Statements generally appear which suggest efficient precipitation scavenging while offering little, if any, experimental evidence.

In order to examine the efficiency of snow in removing lead aerosols introduced by automotive exhaust, a study was initiated during the winter of 1972-73. Since it was desirable to select a site offering a nearly constant source strength, sampling was conducted at the Hanford AEC reservation and initial tests conducted to determine the suitability of the site. This location offers the advantage of isolation from other lead sources as well as nearly constant daily traffic volume. The auto

traffic at the selected site consists solely of vehicles operated by personnel employed on the Hanford project. The traffic pattern therefore conforms to the work schedule of plant personnel. Since peak traffic flow occurs twice daily over periods of approximately 1 hour, sampling time may be kept short, thereby reducing the opportunity for meteorological variations to influence the results.

EXPERIMENTAL

The pattern of sampling sites is described in Figure 1. Traffic flow is in a north-south direction with the four lane roadway turning to the northwest approximately 0.7 miles to

the north. Two points, 30 feet off the pavement, served as reference points in setting out the sampling grid. Additional samples were collected at 30, 60, 125, 250 and 500 feet from these points along lines perpendicular to and at 45° to the highway.

Snow samples were caught in polyethylene bags (21 × 21.5 inches) folded over the edges of standard office wastebasket (13 inch diameter). Bags were generally set out 1 to 1.5 hours before peak traffic flow and collected within 4 hours. During collection, the bags were taped shut and the sealed bags were then stored at subfreezing temperatures. When ready for analysis they were allowed to warm to room temperature before

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FIGURE 1. Sampling Grid Employed to Study Lead Aerosol Removal by Snow

transfer to polyethylene beakers. The inside of each bag was rinsed with 20 ml of 10^{-2} N nitric acid and this solution added to the snow sample. The resulting solution was then evaporated to dryness under a heat lamp, 10^{-2} N nitric acid was added to dissolve any residue, the solution transferred to volumetric flask and diluted to volume with the dilute acid.

Lead concentrations were determined by flameless atomic absorption using a Perkin-Elmer model 306 atomic absorption spectrophotometer equipped with a HGA-70 graphite furnace, deuterium background corrector and model 56 recorder. Nitrogen was used to provide an inert atmosphere for the graphite furnace. Typical recorder tracings are shown in Figure 2 along with pertinent instrumental settings.

The introduction of lead via dilute nitric acid was substantial when using solutions prepared from reagent

grade acid. During the early stages of this work, an acid still similar to that described by Mattinson⁽⁵⁾ was constructed. Analysis of the distilled acid indicates that use of this apparatus has reduced the concentration of lead in the acid from ~100 to 0.08 ppb. Where appropriate, the results have been corrected for the contribution of the untreated acid.

RESULTS

An initial run was made on January 5, 1973. Snowfall began in the early morning hours and continued until mid-morning for a total accumulation of 1.1 inches. Approximately 0.1 inch of snow fell during the hours of interest. Although this snowfall occurred from approximately 6 to 9 a.m. snow samples remained in the field until the early afternoon. The results may therefore be biased

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FIGURE 2. Typical Recorder Tracings Obtained for Lead Analysis by Flameless Atomic Absorption (HGA-70 Graphite Atomizer)

by windblown snow. Winds throughout the day were out of the north-north-west at speeds of 11 to 16 miles/hr.

The results shown in Figure 3 indicate that the concentrations observed are well within the range required for analysis by flameless atomic absorption. Before any conclusions may be drawn regarding snow scavenging of lead aerosols, additional experiments, including dry deposition studies, must be conducted under more clearly defined meteorological conditions. The results to date however, demonstrate the adequacy of the selected site and suggest efficient aerosol removal by snow.

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FIGURE 3. Parts Per Billion Lead in Snow, and Temperatures Recorded During the Sampling Period January 5, 1973

IN-CLOUD SCAVENGING STUDIES

W. G. N. Slinn

A report is given of progress made in the field program designed to determine the influence of pertinent parameters on the in-cloud scavenging of tracer particles by frontal storms.

There are many variables which are potentially important to the in-cloud scavenging of aerosol particles by frontal storms. Aerosol parameters that have been and are being investigated include: particle wettability and solubility, ice freezing and drop nucleating capabilities, and particle size distributions. Meteorological parameters expected to be of importance include natural aerosol concentrations, height of tracer release within the storm, liquid water content, rainfall rate, temperature lapse rate, cloud particle characteristics, vertical velocity distributions, horizontal wind magnitude and shear, and the turbulent diffusion of the tracer. A parameter which unfortunately may be significant but as yet is unknown is the importance of the mechanism by which the tracer is released within the storm. It is planned that this will be examined by comparing the deposition patterns of tracers released external to and within precipitating, orographic clouds.

In last year's annual report, results were shown of the first in-cloud scavenging experiments successfully performed in frontal storms. Since then, four more, larger-scale experiments have been performed and others are underway. It is the pur-

pose of this brief report to describe the goals of these experiments and to outline our progress toward these goals.

Table 1 lists the experiments that have been performed and a brief outline of their goals. The 1971 experiments established the feasibility of the tests and indicated that the natural length scale of the process was larger than the 80 km extent of our sampling grid. During 1972, this grid size was approximately doubled (which required an approximate doubling of the number of field personnel) in the hope of identifying the location of the maximum deposition. This distance is especially important in the theoretical analysis of the data.

In addition to expanding the space scale, we have initiated attempts to define the deposition pattern by obtaining samples spaced sequentially in time. These samples can be obtained at varying distances from the release point simply by varying the tracer release point. If this procedure is successful, the field crew can be reduced considerably, although there will remain the requirements for two men at the radar site, three in the aircraft, two at each of the two rawinsonde release sites, two at

Date	3/11/71	4/16/71	3/23/72	3/23/72	3/29/72	3/31/72	2/22/73	--	--
Release Elevation									
-10°C		In, AgI				In, AgI			Li, AgI
0°C	In, Lithium Stearate			In, AgI			In, AgI (Gen.) Li (Flares)	Li (Gen.) In (Flares)	
5°C			Rhodamine		AgI, In SF ₆				
Met.	Stable	Unstable	Stable	Stable	Unstable Nonprecip.	Neutral	Unstable	Stable	Unstable
Goals	1. Feasibility	1. Feasibility	1. Detect Decrease	1. Detect Decrease	1. Dry Deposition	1. Target Tracer	1. Particle Size Dependence	1. Particle Size Dependence	1. Vary Cosmogenic Radionuclide Concentration & Raindrop Size and Charge
	2. Wettable vs. Nonwetable	2. Ice Nuclei vs. NonIce Nuclei	2. Scavenging of Organic Particles	2. Wettable vs. Soluble	2. Mesoscale Diffusion	2. Propagation of Ice Phase		2. Dependence on w and J _o	
	← 100 Samplers 80 km →		← 250 Samplers 160 km →		← Sequential Samplers →				
	← Space Scale →								
									← Time Scale →

another, larger than 1 μ m. On February 22, 1973, a release was made over the full sampling grid with In and AgI particles from the generators and Li from flares. All systems worked well in the hostile environment of the frontal storm.

In conjunction with this experiment, analysis has been completed which predicts expected differences in the deposition patterns for flare versus generator produced particles. The source of the expected differences is that, on the one hand, the large flare particles can act as cloud condensation nuclei and will rapidly attain a collection efficiency near unity, whereas the generator produced particles take a significantly longer time to attach to the cloud particles and thereby to acquire a large collection efficiency.

for capture by hydrometeors. Consequently at short distances, the rate of increase of the deposition pattern with distance from the release point is expected to be much larger for flare particles than for particles produced by acetone generators. The space-scale grid has been designed so that this effect should be detectable if it is real.

Evidence continues to accumulate about our capabilities to modify scavenging by simultaneously injecting tracers and AgI at appropriate temperatures and liquid water contents. This was discussed in last year's annual report. This year, on March 31, 1972, simultaneous releases

of In and AgI resulted in immediately observable effects. Figure 1 shows a photograph taken by L. F. Radke of a sun pillar that appeared approximately 5 minutes after the release. This photograph indicates that the liquid water was transformed into ice needles. The radar data show that a hole developed in the status deck and propagated across the Peninsula. On the right flank of this hole, there was an especially intense, glaciated cumulus development which from aircraft reports seemed to propagate more slowly than the mean wind. We believe that this propagation of the ice phase was mainly dictated by dynamics rather

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FIGURE 1. A Sun Pillar Observed from the Aircraft Window, a few Minutes After Seeding with AgI During the 3/31/72 Indium Scavenging Experiment. Photo courtesy of Professor L. F. Radke, University of Washington.

than the microphysics. Further experiments of this kind and concurrent theoretical investigations are planned.

During the next favorable storm situation, a number of new experiments are planned. A test will be made to seek the dependence of the rainout rate on significant differences of liquid water contents and rainfall rates. These experiments will be performed over a grid on the Hanford Reservation. An experiment is set up to test if cosmogenic radionuclide concentrations can be varied, as predicted by theory, by varying the natural "lifetimes" of cloud particles via seeding with AgI. Simultaneous with measurements of the cosmogenic radionuclides ^{38}Cl , ^{39}Cl and ^{24}Na , measurements will be made to

determine if raindrop size and charge distributions and the atmosphere's electric field are modified by the seeding. To insure that the seeded water is being sampled, a lithium tracer will be released simultaneously with AgI and subsequently the lithium deposition pattern will be determined.

ACKNOWLEDGEMENTS

These experiments continue to reflect the efforts of a large task force: W. E. Davis, J. M. Thorp, J. C. Draper, D. J. Newland, O. P. Gifford, M. T. Dana, D. W. Glover, J. W. Slood, M. C. Miller, C. W. Thomas, R. N. Lee, V. T. Henderson, F. B. Steele, R. E. Erwin, W. F. Sandusky, F. O. Gladfelder, J. M. Baily, J. A. Young, N. A. Wogman, and students and staff at the University of Washington and Washington State University, especially Professor Lawrence F. Radke, Cloud Physics Group, of the University of Washington.

AIRBORNE MEASUREMENTS OF THE SIZE DISTRIBUTION OF AgI PARTICLES

A. J. Alkezweeny and L. F. Radke*

Airborne measurements were made to determine the size of AgI particles produced by flares and acetone generators. It was found that flare particles are good CCN and many particles are larger than $0.1\ \mu\text{m}$. Particles produced by acetone generator are predominantly below $0.1\ \mu\text{m}$ in diameter.

INTRODUCTION

It appears from various laboratories and field experiments that the size distribution and cloud condensa-

tion nucleus (CCN) activity of AgI particles are the most important factors for their nucleation in clouds. Previous measurements of size distributions were conducted in the laboratory. The size distributions of aerosol particles produced by a generator mounted on an aircraft is

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expected to be quite different from that produced by a ground-based generator. The reason for this is the degree of coagulation, which depends on the amount of ventilation. In the case of generation from an aircraft, both the wind speed and the vortices shed by the wing tips will dictate the degree of coagulation. Dingle⁽¹⁾ has studied a portion of this problem by measuring the size distribution of InCl_3 particles generated from pyrotechnic flares. The size distribution found in a laboratory wind tunnel was found to be significantly different from that measured on the ground, behind the "prop-wash" of an anchored aircraft. Since the majority of cloud seeding projects utilize airborne seeding platforms, the size distribution and CCN activity of the particles should be determined with an airborne instrumentation.

In this report, data is presented on the particle-size distribution and CCN activity of AgI generators. The measurements were conducted utilizing two aircraft.

THE EXPERIMENT

The AgI particles generators were mounted under the wings of the Battelle Cessna 411 aircraft. Two kinds of particle generators were used, AgI flares and AgI acetone generators. The measuring equipment was located onboard the University of Washington B-23 aircraft. With the B-23 aircraft flying behind the Cessna 411, the two aircraft flew at a constant speed and altitude and about 1/2 mile

apart. The altitudes and locations of the experiments were chosen such that the concentration of background aerosol would be very small.

The measuring instruments consisted of a CCN counter⁽²⁾ and a Mee Ice Nuclei Counter, operated at -20°C in addition to a Royco Optical System⁽³⁾ which was capable of simultaneously measuring particle size distributions in the range 0.3 to 5 μm particle diameter. Also employed during this study were a Whitby Aerosol Analyzer⁽⁴⁾ (particle size distribution in the range 0.015 to 0.5 μm), and a Nephelometer⁽⁵⁾ which measures the total light scattering for particles in the size range 0.1 to 2 μm .

RESULTS AND DISCUSSION

The first experiment was conducted on September 28, 1972, at an altitude of about 5,000 feet MSL, just east of Dungeness, Washington. During this flight only AgI flares were used. Before the flares were burned, the concentration of particles greater than 0.3 μm in diameter was very low, less than 10 particles per cubic foot. Figure 1 shows the concentration of CCN active at 5 percent supersaturation, and light-scattering particles during the residence time of the aircraft in the plume. It also shows the contribution of the background aerosol to the measured AgI particles which is less than 10 percent. Furthermore, it shows that AgI particles produced by flares are good cloud condensation nuclei.

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FIGURE 1. CCN Concentration and Light-Scattering Inside and Outside the AgI Flare Plume

Figure 2 shows the complete AgI particle size distribution over the range 0.015 to 5 μm averaged over the entire run. The normalized mass distribution as a function of particle diameter is plotted in Figure 3. This figure indicated that most of the particles are around 0.1 μm in diameter. The second peak around 0.035 μm probably is due to the fact that it is freshly produced aerosol.

The second experiment was made on October 5, 1972, at about 7,000 feet

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FIGURE 2. Size Distribution of AgI Flare Particles

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FIGURE 3. Mass Distribution of AgI Flare Particles

MSL over San Juan Island. AgI acetone solution was burned, followed by the deployment of AgI flares. Figure 4 shows the concentration of CCN active at 0.5 percent supersaturation and light-scattering particles. The fact that the CCN counter detected the particles, but the nephelometer did not is a strong indication that the particles produced by acetone generator are below about 0.1 microns in diameter. The low count shown on the Royco Optical Counter supported this observation. The plume was hardly visible in comparison with the plume formed from burning of the AgI flares. The Ice Nuclei Counter indicated that the particles from this kind of generator are poor ice nuclei.

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FIGURE 4. CCN Concentration and Light Scattering Inside and Outside AgI Acetone Generator Plume

CONCLUSIONS

From these limited measurements, the following conclusions can be drawn:

1. The AgI particles produced by burning flares are of sizes around 0.1 μm in diameter.
2. The particles from AgI flares are good cloud condensation nuclei.
3. Most of the AgI particles produced by the acetone generators are smaller than 0.1 μm in diameter.

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POSSIBLE CHANGES IN AEROSOL PARTICLE DISTRIBUTIONS
CAUSED BY IN-CLOUD SCAVENGING

W. G. N. Slinn

Predictions are made about the evolution of the aerosol particle number and volume distributions caused by interaction with simple nonprecipitating, warm clouds. The rate of decrease of the Aitken nuclei concentration is shown, as is the influence of trace gas conversion on the size distribution of those particles which are active as cloud condensation nuclei.

A field program is being planned to look at the budgets of pollutants within simple, warm-cloud systems. It is planned that one aspect of the program will focus on the changes in the aerosol particle size distributions. It is the purpose of this note to estimate the expected magnitude of these changes.

It is simple to estimate the expected change in Aitken nuclei count. The rate of attachment of Aitken nuclei to cloud droplets is approximately $4\pi DRN$, where D is the diffusion coefficient of the Aitken nuclei, and R and N are the radius and number density of the cloud droplets. Then, if at the entrance to an orographic cloud there are $A_i da$ Aitken nuclei per unit volume whose radius is from a to $a + da$, at the exit there would be

$$A_x(a) = A_i(a) \exp\left(-\frac{4\pi DRNL}{U}\right) \quad (1)$$

where L is the clouds horizontal extent along the mean wind direction, and U is the mean wind speed.

The largest, N , wettable particles will act as cloud condensation

nuclei for the N cloud droplets. If the cloud is not precipitating, these aerosol particles will reappear at the exit of the cloud. However, they probably will be modified by the attachment of Aitken nuclei and by the conversion of trace gases to particles within each cloud droplet. For example, from the results of Scott and Hobbs,⁽¹⁾ the conversion of SO_2 to $SO_4^{=}$, alone, at typical atmospheric concentrations of SO_2 , will result in the formation of a sulphate particle, as a residue from each drop, of radius about $(1/50)^{th}$ the radius of the drop.

To obtain the number and volume distributions at the exit, we assume that the mass of each cloud condensation nucleus will be increased by the same amount $\frac{4}{3}\pi c^3$, where c is a constant. Then at the exit, the radius of each particle will be $a_x^3 = a_i^3 + c^3$. If the initial number of wettable particles per unit volume whose radius is from a_i to $a_i + da_i$ is $n(a_i)da_i$, then at the exit the number distribution will be⁽²⁾

$$n(a_x) = n(a_i) \frac{da_i}{da_x} \quad (2)$$

or

$$n(a_x) = \left(\frac{a_x^3}{a_x^3 - c^3} \right)^{2/3} n \left[(a_x^3 - c^3)^{1/3} \right] \quad (3)$$

For example, if for large particles the initial number distribution can be fit by a "power-law," a_i^{-n} , (3) yields

$$n(a_x) = \frac{a_x^2}{(a_x^3 - c^3)^{\frac{n+2}{3}}} , a_x > c \quad (4)$$

As expected, for particles large compared with c , there is little change to the distribution. However, for smaller particles, the number distribution steepens.

Figure 1 shows the results from Equations (1) and (3) for an orographic cloud 50 km in length, $\bar{U} = 10 \text{ m sec}^{-1}$, $R = 10 \mu\text{m}$, $N = 100 \text{ cm}^{-3}$ and $c = 0.1$ and $0.2 \mu\text{m}$. The initial size distribution of the particles, all presumed to be wettable, is shown. If the cloud were precipitating, the large particle end of the distribution would be reduced by a fraction depending on the rainfall rate, the lifetime of the cloud, etc. (i.e., depending on the in-cloud scavenging efficiency). Figure 2 shows the corresponding volume distributions.

With a view toward experimental design, it is noted that these effects should be detectable if they

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FIGURE 1. Number Distribution of Particles Versus Radius. The solid curve shows an assumed number distribution entering an orographic cloud of horizontal extent 50 km and $\bar{U} = 10 \text{ m sec}^{-1}$. The calculated reduction in Aitken nuclei and change in the large particle distribution are shown, the latter depending on the rate of conversion of trace gases to particles.

are real. The decrease in the Aitken nucleus concentration is not surprising. On the other hand, it would be very interesting to see if the predicted dip and then rise occurs in the number distribution around the point for which there are N larger particles present, since this effect would yield desired data on the efficiency of trace gas conversion to particles.

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FIGURE 2. Number Distribution of Particles Versus Radius (as for Figure 1), but the Volume Distribution Is Plotted, and Only for the Radius Range 0.1 to 1.0 μm

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TRACE METAL ANALYSIS BY ATOMIC ABSORPTION SPECTROMETRY USING A GRAPHITE FURNACE ATOMIZER

R. N. Lee

Atomic absorption spectroscopy using a carbon furnace technique has been evaluated to determine its applicability to atmospheric tracer and pollution studies. Observed analytical sensitivities for a number of metals are reported.

INTRODUCTION

The application of analytical techniques to environmental science has placed new demands on chemical instrumentation. The analytical laboratory must be capable of detecting trace quantities of pollutants as well as tracers introduced to the environment for the study of natural

processes. In addition to the ability to detect nanogram quantities, techniques are required which minimize sample manipulation, may be performed rapidly at a low cost and offer high accuracy. Although many techniques have been championed for their utility in environmental research, it appears that none are capable of handling the full range

of trace metals which are of interest to the environmental scientist.

In order to evaluate the capacity of flameless atomic absorption to meet the needs of precipitation and aerosol analysis, several metals have been investigated to determine detection limits, optimum experimental conditions and potential difficulties associated with this technique.

EXPERIMENTAL

A Perkin-Elmer model 306 atomic absorption spectrophotometer equipped with an HGA-70 graphite furnace⁽¹⁾ and model 56 recorder was employed for all analyses. The HGA-70 accessory allows a choice of seven different temperature programs. The drying and ashing temperatures are fixed by the program selected while the atomization temperature may be adjusted independently. The graphite furnace offers the advantage of high atomization efficiency and permits analysis of microliter quantities of sample. Aqueous solutions are generally most effectively handled although nonaqueous and solid samples may also be accommodated. In most instances, the instrument was operated with a deuterium background corrector.⁽²⁾ This accessory corrects for unwanted absorption and light scattering which would normally add to the recorder response yielding incorrectly high results. Although detection limits are generally increased by a factor of 2-5, use of the background corrector is suggested because of the high background interference commonly observed with environmental

samples. Background correction with this device is, however, limited to analyses using resonance lines in the ultraviolet and near ultraviolet regions.

It is recommended that aqueous samples be acidified with nitric acid. Such treatment enhances the stability of the solution and results in the formation of metal species of low volatility during the preatomization period. In each instance investigated, a 10^{-2} N nitric acid solution appeared to be effective in retarding loss of the metal to the container walls during storage. Standard solutions stored in glass did not suffer detectable deterioration for periods of at least 2 weeks. In the case of indium, solutions stored in polypropylene containers appear less stable. A solution 1N in nitric acid has shown a 10 percent loss of indium after 11 days of storage at room temperature in a Nalgene volumetric flask. In contrast, the stability of silver solutions is apparently independent of the container.

Although nitric acid solutions generally yield products of low volatility in the graphite furnace, the analyst should use caution in selecting conditions for the ashing stage of the heating cycle. Loss of the metal may occur either by carbon reduction of the metal oxide (formed by thermal decomposition of the nitrate) and subsequent vaporization of the metal, or loss of the oxide, or nitrate directly. In this connection, Fuller⁽³⁾ has reported significant loss of copper and nickel when a 1100°C ashing stage (program 7) was

employed. In our laboratory, we have failed to detect loss of either metal when this temperature was maintained for 60 seconds prior to atomization. We have, however, noted a 30 percent decrease in the lithium absorption signal when the ashing time was increased from 0 to 30 seconds at 1100°C.

In at least one instance, a prolonged ashing period results in a more intense absorption signal. Maximum response for indium is obtained when a nitric acid solution of the metal is heated to 490°C (program 5) for 5 minutes prior to atomization. This interval is apparently required to complete thermal decomposition of the nitrate to the oxide. It is assumed that the oxide must therefore yield a more uniform release of atoms to the vapor phase during atomization. Increasing the ashing time beyond 5 minutes produces no further change in the intensity of the absorption signal.

Table 1 lists those metals which have been studied. Although each may be detected at subnanogram levels, the application of this flameless atomic absorption technique may present a formidable problem when background interference exceeds the capacity of the background corrector. Reliable methods must therefore be developed to separate the metal of interest from interfering material if maximum utility is to be achieved.

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TABLE 1. Recommended Conditions and Detection Limits for Flameless Atomic Absorption Analysis of Various Metals Using the Perkin-Elmer HGA-70 Graphite Furnace Atomizer

Metal	Program	Wavelength, nm	Atom. Temp.	Sensitivity	Detect. Limit
Beryllium	6	235	2400°C	1.4×10^{-11} gm/1%	0.02 ng.
Chromium	7	358	2400	5.6×10^{-11}	0.04 ^(a)
Copper	7	325	2400	7×10^{-11}	0.05 ^(a)
Indium	5	304	2400	1.8×10^{-10}	0.1 ^(a)
Iron	7	248	2400	5×10^{-11}	0.1 ^(a)
Lead	6	283	2400	1.2×10^{-10}	0.08 ^(a)
Lithium	6	671	2400	8.3×10^{-11}	0.2
Manganese	7	280	2400	2.3×10^{-11}	0.02 ^(a)
Nickel	7	232	2400	5.2×10^{-10}	0.5 ^(a)
Silver	5	328	2250	1.3×10^{-11}	0.01
Zinc	5	214	2250	7×10^{-12}	0.01

a. With deuterium background corrector

TRACER STUDIES IN THE NATIONAL HAIL

RESEARCH EXPERIMENT

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

The National Hail Research Experiment (NHRE) is being conducted during summer seasons in northeastern Colorado. The main objective of NHRE is to determine the feasibility of preventing damaging hail by seeding with silver iodide. The main objectives of Battelle-Northwest's participation in NHRE are to determine precipitation scavenging efficiencies, aerosol and hydrometeor trajectories, and ice nucleation mechanisms using natural and artificial tracers.

During 1972 six inert tracers were released sequentially into the updrafts of convective storms from just below cloud base on July 24 and 26. Samples of rain and hail from these storms, collected using 250 rain-hail separators, are now being analyzed by neutron activation for the tracers, silver iodide, and other elements to determine scavenging efficiencies and trajectories. Measurements of the concentrations of the cosmogenic radionuclides ^{38}Cl ($t_{1/2} = 37$ min) and ^{39}Cl ($t_{1/2} = 55$ min) in precipitation from several convective storms were used to calculate the elapsed time between the beginning of coalescence and the arrival of the precipitation at the ground.

The National Hail Research Experiment (NHRE)* is being conducted during summer seasons in a 2500 square mile area in the northeastern corner of Colorado. The stated objectives of this study are to describe and understand hailstorms, to identify effective means of modifying damaging hailstorms, and to determine whether such methods are technically and economically feasible. Beginning in the summer of 1972 thunderstorms were seeded with silver iodide in a randomized experiment to determine statistically the effectiveness of silver-iodide seeding in the suppression of damaging hail.

Battelle-Northwest Laboratory personnel conducted two main experiments in NHRE during the summer of 1972. On July 24, six inert tracers--indium, ruthenium, gold, rhenium, europium and tantalum--were released sequentially at an altitude of 11,300 ft into the updraft of a convective cell located near the southwestern corner of the NHRE precipitation measuring network. To release these tracers, acetone-heptane solutions containing them were burned in six Brad-Patton aerosol generators mounted beneath the wings of the Battelle Cessna-411. They were released while the Cessna was flying below cloud base just below and approximately

* This work is partially supported by NSF Contract.

one quarter of a mile behind a silver iodide seeding aircraft. The tracers were released only during the burning of silver-iodide flares. The total burn time for each tracer was about 2 minutes. On July 26 several cells merged to form a squall line. The six tracers were released sequentially into the updraft of this system.

Following each tracer release, the rain and hail in all of the 250 NHRE rain-hail separators were collected and quickly frozen. Several precipitation samples were also collected utilizing containers lined with plastic bags. These samples are now being analyzed by neutron activation for silver iodide, for the inert tracers and for several other elements. When the analysis of the precipitation samples is complete, the results will be used to calculate scavenging efficiencies and trajectories for the silver iodide and the inert tracers.

The trajectories taken by the silver iodide could be different than those taken by the tracers, either because of possible differences in the scavenging mechanisms, or because the silver iodide causes droplets to freeze and the tracers do not. The relative scavenging efficiencies for the silver iodide and the inert tracers could provide information on the mechanism by which silver iodide acts as an ice nucleus.

The rain and hail in the rain-hail separators were also collected on 2 days on which convective storms were seeded with silver iodide. These sam-

ples will be analyzed by neutron activation to determine silver-iodide scavenging efficiencies and deposition patterns, and the background concentrations of the tracers in the precipitation.

The second experiment was the simultaneous measurement of the concentrations of short-lived cosmogenic radionuclides, nuclear-weapons-produced radionuclides, and radon daughters in large-volume rain samples. These rain samples were collected from the enameled, corrugated iron roof of a large building on a farm 10 miles north of Stoneham, Colorado. Rain draining from this roof was collected in a gutter and passed through 2 in. diameter polyethylene pipe into 55 gal polyethylene barrels. The water was then pumped through resin beds to remove dissolved radionuclides. The filters and resin beds were counted using germanium diode and multidimensional NaI(Tl) gamma-ray spectrometers.

Cosmogenic radionuclides are produced continuously in the atmosphere by cosmic radiation and immediately become attached to surrounding aerosol particles, cloud droplets, or ice particles. When these cloud droplets or ice particles begin to grow by coalescence, the volume-to-area ratio increases so rapidly that direct capture of cosmogenic radionuclides rapidly becomes negligible compared to the concentrations already in the hydrometeor. The cosmogenic radionuclide activities begin to decay when coalescence starts, and the ratios of the various radionuclides

change because of their differing half-lives. The time between the beginning of coalescence and the arrival of the precipitation at ground level, therefore, can be calculated from the ratio of $^{39}\text{Cl}(t_{1/2} = 55 \text{ min})$ to $^{38}\text{Cl}(t_{1/2} = 37.3 \text{ min})$ in the precipitation.

The calculated times showed a fairly consistent pattern for the storms in northeastern Colorado, averaging 72 minutes at the beginning and end of the storm, and decreasing to 6 to 29 minutes in samples in the middle of the storm. The longest calculated time was 126 minutes.

THE USE OF NATURAL AND MAN-MADE RADIONUCLIDES TO STUDY IN-CLOUD SCAVENGING PROCESSES

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

The concentrations of cosmic-ray-produced radionuclides, nuclear-weapons-produced radionuclides and radon daughters were measured simultaneously in rain and in ground level air samples at Quillayute, Washington in March 1972 and in northeastern Colorado in June 1972. The elapsed times between the beginning of droplet and/or ice crystal coalescence and the arrival of the precipitation at ground level, calculated on the basis of the $^{39}\text{Cl}/^{38}\text{Cl}$ ratios in the precipitation, averaged 50 minutes at Quillayute. In northeastern Colorado the calculated times averaged 70 minutes, except for a few samples near the middle of several storms which yielded calculated times of 6 to 29 minutes. The $^{39}\text{Cl}/^{24}\text{Na}$ ratios in the precipitation indicated that droplets growing by condensation experienced an increased rate of capture of newly formed cosmogenic radionuclides because of their increasing size and in northeastern Colorado because of their vertical displacement in the thunderstorm updrafts.

The cosmic-ray-produced radionuclides, nuclear-weapons-produced radionuclides and radon daughters were measured in rain and in ground level air samples on the west coast of the Olympic Peninsula at Quillayute, Washington in March 1972 and in northeastern Colorado in June 1972. These measurements provide information on precipitation scavenging rates and mechanisms.

The cosmogenic radionuclides are produced at relatively constant rates in the atmosphere by spallation reactions of cosmic rays with atmospheric gases. The atmospheric concentrations of the short-lived cosmogenic radionuclides are nearly independent of time and the ratios of their concentrations change little with location or altitude. The radon daughters are produced by the decay of

radon-222 gas, which emanates continuously from the earth's crust. The radon daughters are generally fairly close to equilibrium with radon in the atmosphere and their disintegration rates are nearly equal.

The cosmogenic radionuclides and radon daughters become attached to surrounding aerosol particles, cloud droplets or ice particles almost immediately after their formation. However, when the droplets or ice particles begin to coalesce, the surface area to volume ratios on the growing hydrometeors decrease sharply and the rate of capture of newly formed cosmogenic or radon daughter radionuclides rapidly becomes negligible compared to the concentrations in the hydrometeors. These radionuclides then begin to decay away and the activity ratios change with time due to differences in their half-lives. Therefore, the elapsed time between the beginning of coalescence and the arrival of the precipitation at ground level can be calculated from either the ratio of the cosmogenic radionuclides $^{38}\text{Cl}(t_{1/2} = 37.3 \text{ min})$ and $^{39}\text{Cl}(t_{1/2} = 55 \text{ min})$ or from the ratio of the radon daughters $^{214}\text{Pb}(t_{1/2} = 26.8 \text{ min})$ and $^{214}\text{Bi}(t_{1/2} = 19.7 \text{ min})$ in the precipitation.

The above calculations are complicated because the radionuclide ratios in the droplets or ice crystals may not be at the equilibrium values when coalescence starts. If the capture of newly formed cosmogenic or radon daughter radionuclides is governed by diffusion alone, the rate of capture should be proportional to the radius

of the particle. Therefore, if one aerosol particle of 0.1μ radius nucleated to form a water droplet and then grew to 30μ radius, the rate of capture of newly formed radionuclides by the 30μ droplet would be 300 times that of the original aerosol particle. This increased capture rate would cause the $^{39}\text{Cl}/^{38}\text{Cl}$ and $^{214}\text{Bi}/^{214}\text{Pb}$ ratios to decrease since the ^{39}Cl half-life is longer than that of ^{38}Cl , and because ^{214}Bi is formed by the decay of ^{214}Pb . In a cloud a large number of droplets grow at once; therefore, the competition among the growing droplets for the constant number of newly formed radionuclides would greatly reduce any increase in the capture rate by the droplets. In fact, if all of the aerosol particles nucleated and grew to water droplets at the same time and at the same rate, the growing droplets would experience no increase in capture rate.

In rain samples collected in Quillayute and in northeastern Colorado, an increased rate of capture of newly formed radionuclides by growing water droplets was indicated by high ratios of the activities of the radon daughters and short-lived cosmogenic radionuclides to those of the longer cosmogenic or nuclear-weapons-produced radionuclides. For example, the $^{39}\text{Cl}(t_{1/2} = 55 \text{ min})/^{24}\text{Na}(t_{1/2} = 15 \text{ hr})$ ratios in precipitation at Quillayute and northeastern Colorado averaged about five times that expected from equilibrium atmospheric concentrations, indicating that the amount of ^{39}Cl in the droplet increased to

about five times the amount in the original aerosol particle. The actual increase could have been significantly less than this at Quillayute, since the ^{24}Na concentrations in the atmosphere could have been considerably reduced by scavenging in the previous several hours. However, in northeastern Colorado this was not true for although the atmospheric ^{24}Na concentrations were probably near equilibrium, it had been several days since the previous thunderstorm. Since cosmogenic radionuclide production rates increase with altitude, the increase in the ^{39}Cl concentrations in the growing drops in the thunderstorms of northeastern Colorado was probably due at least in part to the vertical displacement of the droplets in the updraft. An increase in the rate of capture of newly formed radionuclides by growing droplets was also indicated by the values of the $^{39}\text{Cl}/^{22}\text{Na}$ ($t_{1/2} = 2.6 \text{ yr}$) and $^{39}\text{Cl}/^{137}\text{Cs}$ ($t_{1/2} = 30 \text{ yr}$) ratios in rain at Quillayute and in northeastern Colorado which were considerably higher than would be expected on the basis of ^{22}Na and ^{137}Cs concentrations measured in the air at ground level. However, interpretation of these ratios is complicated because ^{22}Na and ^{137}Cs concentrations at precipitation formation altitudes can only be estimated.

The increased rate of capture of newly formed cosmogenic radionuclides

by droplets growing by condensation will not seriously affect the times between the beginning of coalescence and the arrival of precipitation at ground level calculated on the basis of $^{39}\text{Cl}/^{38}\text{Cl}$ ratios in precipitation. However, since ^{214}Bi is formed by the decay of ^{214}Pb , the $^{214}\text{Bi}/^{214}\text{Pb}$ ratio in droplets at the beginning of coalescence will be considerably lower than the unit equilibration activity ratio if the droplets growing by condensation experienced increased rates of capture of newly formed radon daughters. The times between the beginning of coalescence and the arrival of the precipitation at ground level calculated on the basis of $^{214}\text{Bi}/^{214}\text{Pb}$ ratios in precipitation agree with those calculated on the basis of the $^{39}\text{Cl}/^{38}\text{Cl}$ ratio only if it is assumed that the $^{214}\text{Bi}/^{214}\text{Pb}$ activity ratio is about 0.5 rather than 1.0 at the beginning of coalescence. The times between the beginning of coalescence and the arrival of the precipitation at ground level, calculated on the basis of $^{39}\text{Cl}/^{38}\text{Cl}$ ratios in precipitation averaged 50 minutes for three storms at Quillayute. There did not appear to be any systematic variation in the calculated times throughout the course of the storms. In northeastern Colorado the calculated times averaged 70 minutes, except for a few samples near the middle of several storms which yielded calculated times from 6 to 29 minutes.

PRECIPITATION SCAVENGING OF INORGANIC POLLUTANTS
FROM METROPOLITAN SOURCES*

M. Terry Dana, Jeremy M. Hales and John M. Thorp

Rainwater samples from convective storms in the St. Louis metropolitan area were collected and analyzed for the chemical species SO_2 , NO_3^- , NH_4^+ , NO_2^- , SO_4^{2-} , and H^+ . Preliminary results indicate significant washout of these city-produced pollutants often occurs within a few miles of the down-storm side of the city.

A field research program was undertaken to measure the effectiveness of precipitation scavenging as a mechanism for removal of pollution originating in the St. Louis metropolitan area. Collection and analysis of rainwater samples took place during August, 1972, as part of the METROMEX (Metropolitan Meteorological Experiment) cooperative effort. Supporting measurements included several radiosonde flights, monitoring of weather radar, and aircraft soundings of the air concentrations of several air pollutants in the area. This report describes the conduct of the field program and current status of the data analysis.

EXPERIMENTAL PROCEDURE

To assess the impact of scavenging upon the metropolitan-source atmospheric pollution burden, it was required to sample precipitation at various locations around the city.

In general, this involved "upstorm" collection sites for background measurement, and two "downstorm" sampling lines. The latter, spaced at two distances from the city center, facilitated fulfillment of the objective of investigating the metamorphoses of pollutants as they react physically and chemically in time prior to deposition. The sampling network is shown in Figure 1. The locations of the sampling lines were dictated by the expected normal northwest to southeast movement of showers. Indicated on the map are the locations of the radiosonde launch point and weather radar (Lambert-St. Louis Airport), and the Battelle mobile laboratory (near Glen Carbon, Illinois).

The precipitation collectors, one of which is shown in Figure 2, consisted of plastic funnels with attached 500 ml bottles, the latter enclosed in insulated containers with dry ice. The dry ice froze the water soon after collection, thus fixing the trace constituents in the solid state and preventing their loss by desorption and reaction prior to analysis.

* This work was financially supported by the Environmental Protection Agency via a related services agreement with the Atomic Energy Commission.

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FIGURE 1. Precipitation Collection Network

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FIGURE 2. View of Freezing Rain Collector

Chemical analyses for SO_2 , NO_3^- , NH_4^+ , NO_2^- , and SO_4^{2-} were performed using Technicon Autoanalyzers. Hydrogen ion concentrations were determined from pH measurements. Most analyses were conducted on site at the mobile laboratory, but all samples were shipped frozen to Richland, Washington, where all SO_4^{2-} and NO_2^- measurements were made, in addition to repeated analysis for NH_4^+ and NO_3^- to assess possible effects of chemical reaction of these species during shipment.

The conduct of a typical run began with the field director--who monitored

the National Weather Service weather radar at Lambert Field--alerting the field crew (one man on each of the three sampling lines) as to expected timing and direction of travel of a storm. He specified the sample positions to be used (about 20 on each line), being guided by the direction and the desire for containment of the shower, or at least the portion of it which would pass over the city. The field director and the radiosonde operator conducted radiosonde flights during the time of sampling. At the end of the rain, the samples were secured and taken to the laboratory or to cold storage.

Table 1 lists sampling details of the five runs conducted. The position numbers correspond with those

of Figure 1. On only two of the runs were samples collected on all lines. Incompleteness of the initial setup affected Run 1, and the timing of the storm arrival forced doubling up on servicing just one downstorm line during Run 2. The storm of Run 5 stopped precipitating before it reached the downstorm lines.

Aircraft soundings were made on several occasions, measuring air content of particulates and SO₂ all around the city. Scheduling difficulties prevented such flights on run days except for that of Run 5, when a convective cell north of the city was investigated. The late arrival of aircraft NO-NO_x detection equipment prevented its use on this project.

TABLE 1. Summary of Washout Measurements

Date	Run No.	Sample Line	Position Numbers ^(a)	Inclusive ^(b) Collection Time (Time of Day-CST)	Storm Path	Rawinsonde Release Time (CST)
8/12/72	1	B	19-38	0912-1433	NW to SE	
8/17/72						1026
8/19/72	2	B	22-28	1421-1647	NW to SE	1402
8/20/72	3	C	61-74	1334-1700	NE to SW	0938, 1430
		B	1-5	1349-1649	NE to SW	
		A	146-153	1530-1840	NE to SW	
8/21/72	4	A	146-190	1530-2030	NW to SE	1417
		B	15-37	1455-1731	NW to SE	
		C	65-89	1503-2001	NW to SE	
8/25/72	5	A	154-168	0955-1705	W to E	1025, 1501
8/28/72						1053

a. Sample positions used at which measurable rain fell.

b. Total time from first collector out to last collector in.

INITIAL RESULTS

Rainfall amounts and the rainwater concentrations of NO_3^- , SO_4^{2-} , NH_4^+ , and H^+ for Run 4 are shown as bars on the area maps Figures 3 and 4. Rainfall was heavy, especially downtown (3.75 inches) and eastward. Rain collected at positions 25 to 35 (line B) exceeded the 30 mm capacity of our collectors, but was approximately 30 mm at position 36, indicating a decrease in the rain amounts to the south. The storm rainfall was contained by collectors on the second downstorm line C. Figures 3 and 4 show the

significant enhancement of the concentrations above background on line B, while concentrations on line C were back to near background levels.

SO_2 was not detectable in any of the samples, probably because of the acidity of the rain, which inhibits its affinity for SO_2 . The pH of the rain was quite uniform over the entire area (up- and downstorm), with the run-average values between about 3.9 and 4.4.

Further analyses and interpretations of the data are under way, and supporting data from other METROMEX

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FIGURE 3. Rain, NO_3^- , and NH_4^+ Deposition, Run 4. Arrows indicate rainfall in excess of 30 mm.

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FIGURE 4. Rain, H⁺, and SO₄⁼ Deposition, Run 4.
 Arrows indicate rainfall in excess of 30 mm.

contributors are being sought. Especially valuable in determining source inventory estimates will be city and state air pollution measurements and aircraft data. Full meteorological assessment of the run days will be

performed, and other data will be gathered to aid in deducing the extent of precipitation scavenging's influence upon the atmospheric pollution output of the city.

TRACER STUDIES IN THE METROPOLITAN METEOROLOGICAL EXPERIMENT

The Metropolitan Meteorological Experiment (METROMEX) is a study of convective storms in the St. Louis area. Scientists from the Radiological Chemistry Section conducted two main experiments during METROMEX in 1972. The first was the sequential release of six inert tracers from 10 to 13.5 thousand feet near the upwind edge of convective towers to measure the amount of air entrained which reached regions of precipitation scavenging. Samples of rain from these convective storms were collected using a network of 122 samplers. They are now being analyzed by neutron activation for the tracers and for other elements to determine scavenging efficiencies and tracer trajectories. In the samples analyzed, those which were collected 10 to 20 miles downwind of the gold, iridium, and tantalum releases, measurable quantities of these tracers were generally present. The second experiment was the measurement of the trace element composition of air filter samples to determine the contribution of the St. Louis metropolitan area to the contamination of the atmosphere.

TRACER AND POLLUTION STUDIES DURING METROMEX

J. A. Young, T. M. Tanner,
C. W. Thomas and N. A. Wogman

The METROMEX experiment is a study of convective storms being conducted for five summer seasons beginning in 1971 in a 2000 square mile area centered around St. Louis, Missouri. Special attention is being given in this experiment to determine the effects of heat and contaminating materials added to the atmosphere on precipitation processes and on aerosol scavenging in convective storms.

Scientists from the Radiological Chemistry Section of Battelle-Northwest conducted two main experiments during METROMEX in the summer of 1972. In the first experiment begun on August 3, six inert tracers (Eu, Ru, Ta, Ir, Re and Au) were sequentially released at 10 to 13.5

thousand feet near the upwind edge of convective towers to measure the amount of air penetrating them and reaching the region of precipitation scavenging (Figure 1).

The tracers were released by burning acetone-heptane solutions containing the elements in six Brad-Patton aerosol generators mounted underneath the wings of the Battelle Cessna-411. Each tracer was released over a period of 2 minutes.

The six inert tracers were released into a storm system which consisted of a series of convective cells partially merged to form a squall line. Europium, ruthenium, and tantalum were released near the western part of a cell located near the center of the Battelle precipitation network. Iridium, rhenium, and gold were then released near the western edge of a second cell located

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FIGURE 1. Tracer Release Locations and Precipitation Collection Network During METROMEX

near the southwest corner of the network. The release of the six tracers took 65 minutes. The pilot of the Cessna reported that there were measurable inflows into the convective towers throughout the burn of the six tracers. The tracers, indium and lithium, were also released into the updraft of this storm at cloud base by Illinois State Water Survey personnel.

Following the tracer release, samples of precipitation were collected using a 25 mile by 36 mile network of 122 rain collectors. The rain samples were frozen as quickly as possible and are now being analyzed by neutron activation. Aliquots of all of the samples will be sent to Illinois State Water Survey for indium and lithium analysis.

The analysis of gold, iridium and tantalum has been completed for several of the water samples. The seven samples analyzed were collected from 0 to 10 miles downwind of the gold release and did not contain measurable amounts of gold. However, most of the samples collected 10 to 20 miles east and northeast of this release contained gold concentrations ranging from 0.8 to 5.0 ng per sample. Two of the four samples analyzed from the eastern edge of the sampling grid, 20 miles downwind of the release, contained gold. The deposition pattern for the iridium was much the same as that for gold.

Tantalum was released approximately 10 miles north of the gold release, but none of the samples collected directly downwind and east have been analyzed. Some of the samples collected from 7 to 19 miles southeast have been analyzed and they contained measurable quantities of Ta. Two of these samples contained 110 and 165 ng.

The concentrations of a few other trace elements have been obtained from the neutron activation analysis of the rain samples; amounts ranged per sample from 41 to 260 ng cobalt, 71 to 4400 ng chromium, 72 to 426 μ g iron, 7 to 84 ng hafnium, 81 to 780 ng lanthanum, 15 to 116 ng scandium, 11 to 125 ng samarium, and 0.004 to 45 ng thorium.

Aerosol samples were also collected to chemically characterize the aerosol in the St. Louis area and these are now being analyzed for trace elements by X-ray fluorescence and neutron activation analysis to determine the contribution of the

St. Louis metropolitan area to the trace element contamination of the atmosphere. On August 7, 8, and 9 mobile Hurricane air samplers were used to collect samples upwind, in, and downwind of St. Louis. In addition, stationary air samplers were operated continuously at four locations for over a week. Lundgren impactors were also operated at these locations to obtain trace element concentrations as functions of particle size and time.

The Hurricane air filter samples have all been analyzed by X-ray fluorescence, but not by neutron activation. Perhaps the most interesting trace elements measured by X-ray fluorescence are lead and bromine produced primarily by automobile exhaust. The lead concentrations several miles upwind of St. Louis ranged from 35 to 71 ng/m^3 . In downtown St. Louis concentrations varied from 140 to 800 ng/m^3 . Concentrations downwind were generally higher than those upwind. On August 8 concentrations of 360 and 510 ng/m^3 were measured 20 miles east and downwind of the city. These concentrations were considerably higher than some measured closer to St. Louis the same day. Samples collected 40 to 50 miles downwind on August 7 contained 36 to 130 ng/m^3 , as compared to 35 ng/m^3 upwind. The highest lead concentrations (3500, 1690, and 1460 ng/m^3) were measured five to twenty miles east of the city center near Interstate Highway 70. As expected, the bromine concentrations were generally about one quarter of the lead concentrations.

AN AEROSOL GENERATOR SYSTEM FOR
SEQUENTIAL TRACER RELEASE

C. W. Thomas H. G. Rieck, Jr.,
J. A. Young and N. A. Wogman

An aerosol generator system was developed for the sequential release of up to 12 different tracers into storm systems to study several time dependent storm processes. The generator system consists of two banks of four Brad-Patton aerosol generators mounted beneath and to the rear of the engine nacelles of the Battelle Cessna-411 aircraft (Figure 1). The generators release submicron-sized particles of the tracers by burning

acetone-hexane solutions containing the elements. The eight generators burn a total of about 1 gal of acetone-hexane per minute. Ram pressure resulting from the aircraft motion is used to supply air to the burners. Acetone-hexane is fed into the burners by nitrogen gas pressure from twelve 1-1/3 gal aluminum feed tanks mounted in the baggage compartments in the engine nacelles (Figure 2). The six tanks in each baggage compartment are mounted on a common base plate which is bolted to the floor to prevent movement of the tanks during flight (Figure 3).

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FIGURE 1. Aerosol Generators Mounted Beneath the Engine Nacelle of the Battelle Cessna-411

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FIGURE 2. Feed Tanks Mounted in the Baggage Compartment of the Engine Nacelle of the Battelle Cessna-411

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FIGURE 3. The Feed Tank and Hardware

Some plating of the tracers on the walls and corrosion of the walls was experienced, so the tanks have subsequently been coated with teflon. During tracer releases, the tanks are pressurized to 50 psi with nitrogen fed through stainless steel lines from a cylinder located in the cabin. The line from each tank is equipped with a one-way spring-loaded valve to prevent acetone-hexane from flowing between tanks.

Acetone-hexane is fed to the burners through stainless steel lines connected near the base of the tanks. There is a wire gauze filter in each line which prevents small particles from reaching the burners and fouling the burner nozzles. Each line is equipped with a solenoid valve which is kept closed except during release. The solenoid valves are controlled from a switch box operated by the copilot. At the present time the switch box is wired so that only one solenoid on each wing may be opened at one time.

For the 1972 releases, each individual tracer was placed in one tank on one wing and in the corresponding tank on the other wing so that a single tracer could be released simultaneously from the burners on both wings. The solenoid valves had to be fitted with neoprene seats and polyethylene bushings because the original valve seats and bushings were not acetone resistant.

Each burner is equipped with an igniter similar to a spark plug. The igniter can be left on throughout the burn of the tracer, although it was found that once the acetone-hexane was ignited, it would continue to burn after the igniter was turned off.

Following a tracer release, the pressure in the tanks is removed through bolts equipped with O-ring seals mounted in the lids. The tanks are generally filled with tracer solution through these bolt holes, even though the lids can be removed.

SIZE DISTRIBUTIONS OF ST. LOUIS AEROSOL PARTICLES

A. J. Alkezweeny

The size distributions of aerosol particles were measured during the month of August, 1972, in St. Louis. The result shows that for a day with low-level temperature inversion, the size distribution is characterized by high concentrations of larger particles. The size distribution averaged over the entire period of measurement approximately fit the equation $\Delta N/\Delta D = 6.5D^{-4}$.

INTRODUCTION

It is well recognized that air pollution has several harmful effects to our life. Degradation of visibility, and damage to our health, properties and vegetation have been documented. Also, evidence has accumulated in recent years that suggests air pollution is changing the weather in certain localities. Since aerosol particles are a major pollutant, it is important that their physical and chemical properties be determined. Knowledge of the concentration and size distribution of aerosol particles and their variation in time and space, is essential to our understanding of their formation, removal, and effects.

In this report the concentration and size distribution of aerosol particles measured in St. Louis during the month of August, 1972, are presented. The measurements were conducted as part of the METROMEX program.⁽¹⁾

EXPERIMENT

The aerosol monitoring instrumentation for this study was located on

the second floor, southwest corner, of the Holiday Inn in Edwardsville, Illinois, about 15 miles east of St. Louis. This location is about 10 miles from the nearest major industrial source of aerosol particles, but is less than a mile north of the major interstate highway. A sampling tube was extended to the roof of the building, and was predominantly upwind of the building exhaust. The sampling system consisted of a Royco model 220 optical counter, pulse converter, pulse height analyzer, and a digital printer. This system is capable of simultaneously measuring the size distribution in the range 0.3 to 5 microns in about 100 channels. Dry, filtered air was used to dilute the aerosol sample. The amount of dilution was set so that the total number of particles counted was less than 15,000 particles per minute. Dilution was necessary because at higher counting site coincidence loss became significant.

RESULTS

A total of 26 size distributions were obtained over the period from August 14, to August 24, 1972. Each

distribution was an average of several distributions. A portion of the results are presented here; the complete results will be reported later.⁽²⁾

Comparison of the particle-size distributions obtained at 1330 for August 16, 17, 18, and 22, show a definite difference as well as similarities. For example, on the 17th and the 18th the size distributions were nearly the same. On these two days the temperatures were 93°F and 92°F, relative humidity was 39 percent and 38 percent and the height of the temperature inversion were 845 mb and 709 mb, respectively. The average of the two distributions is plotted in Figure 1, curve a. On the 16th and 22nd the temperatures were 80°F and 80°F, the relative humidities were 75 percent and 61 percent, and the heights of the temperature inversions were 968 mb and 945 mb, respectively. The particle-size distributions were characterized by higher concentrations of large particles. An average of the two size distributions is plotted in Figure 1, curve b.

On August, 19, concentrations and size distributions of aerosol, temperature, and relative humidity were measured every two hours starting at 0830 to 1600 local time. Throughout the day the wind was calm, the temperature continued to increase, with a decrease in the relative humidity until about 1430 and the trend was reversed. The changes in the concentrations of particles in the size ranges 0.3 to 1 μm and greater than

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FIGURE 1. Size Distributions of Particles During Temperature Inversion and Noninversion Conditions

1 μm are shown in Figure 2 along with the variations in temperature and relative humidity. It was noted that at the beginning of the measurements, the sky was clear, but two hours later scattered low clouds and haze appeared in the sky. At 1430 a large cumulus formation was overhead. Half an hour later, precipitation started.

The peak in the particle concentration around noon probably was caused by coagulation of particles formed earlier by photochemical reaction. Averages of all the particle size distributions obtained during the entire

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FIGURE 2. The variation in Temperature, Relative Humidity and Particle Concentration with Time

period of measurement, except for one day, are shown in Figure 3. The data points approximately fit the following equation:

$$\frac{\Delta N}{\Delta D} = 6.5D^{-4},$$

where $\Delta N/\Delta D$ is the number of particles per cm^3 per micron, and D is the particle diameter in microns.

CONCLUSIONS

From the data presented here the following conclusions may be made:

1. There is a definite effect of the height of the temperature inversion on the particle size distribution.

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FIGURE 3. Average of Particle Size Distribution

2. Average of the particle size distributions obtained over a long period of time, can be approximated over the particle size range measured, by a Junge distribution for an aged aerosol.

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COMMENTS ON THE DISCUSSIONS OF "RESIDENCE TIME
OF PARTICLES IN URBAN AIR"*

W. G. N. Slinn

A critique is made of recent discussions in the literature about the relationship between turbidity variations and the residence time of particles in an urban atmosphere.

In their reply⁽¹⁾ to Fischer's comments⁽²⁾ on their original paper,⁽³⁾ Esmen and Corn seem to have made a mechanical error. To find the ratio of the turbidity at time zero, $K(0)$, to the turbidity at a later time, $K(t)$, they have set up the ratio, cancelled, and then integrated, rather than integrate first; see their Equation (4). Conceptually this amounts to neglecting the time change of the extinction cross section, accounting only for the time change of the total number of particles. In effect they have not followed the instructions of their reference:⁽⁴⁾ "For a polydisperse suspension $n\alpha E$ has to be integrated over the particle-size distribution" The purpose of this report is to recalculate the turbidity and thereby respond to Fischer's comments.

Using the same notation as Esmen and Corn, we are to find the time dependence of the turbidity:

$$K(t) = \int_0^\infty \alpha E \& n(r,t) dr. \quad (1)$$

We assume that the size distribution can be approximated for the size-range of interest by the time-dependent Junge density function

$$n(r,t) = n_0 \left(\frac{r_0}{r} \right)^{1+\beta} \exp \left(- \frac{rt}{r_1 \tau} \right) \quad (2)$$

where they suggest $r_1 = 1 \mu$ and $\tau = 75$ hours. Based on his observations of the diurnal variation of turbidity, Fischer suggests that the "dry removal" time constant, τ , is an order of magnitude smaller.

We approximate the extinction cross section by the analytically convenient expression

$$\alpha E = \pi r^2 \frac{4}{\alpha^{1/6}} \exp \left(- \frac{1}{3\alpha} \right) \quad (3)$$

where $\alpha = kr$ and $k = 2\pi/\lambda$ is the wave number of interest. Substituting (2) and (3) into (1), performing the integrations and then dividing, we obtain

$$\frac{K(t)}{K(0)} = \frac{2}{\Gamma(v)} \left(\frac{p}{3} \right)^{v/2} K_v \left(2\sqrt{\frac{p}{3}} \right) \quad (4)$$

where $v = \beta - \frac{11}{6}$, K_v is a modified Bessel function of order v and $p = t/(r_1 k \tau)$.

To evaluate (4), an exact specification of β is needed. Our recent

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measurements* have shown considerable variability in β , but they tend to be in the range between 3 and 4 in urban environments. If, for analytical convenience, we take $\beta_1 = 3-1/6$ and $\beta_2 = 4-1/6$, then upon expanding the Bessel functions for small values of the arguments, we obtain

$$\frac{K(0)}{K(\tau)} = 1 + x + \frac{1}{2} (\ln x)^2 + (1 - 2\gamma) x + O(x^2), \quad \beta = \beta_1 \quad (5a)$$

$$\frac{K(0)}{K(\tau)} = 1 + x + O(x^2 \ln x), \quad \beta = \beta_2 \quad (5b)$$

where $x = p/3$ and γ is Euler's constant, $\gamma \approx 0.577$. Equations (5) are to be compared with Esmen and Corn's Equation (5).

For the case $\lambda = 5000 \text{ \AA}$, $r_1 = 1 \mu$ and Esmen and Corn's suggested value for $\tau = 75$ hours, then (5) yields

$$K(0) = 1.015 K(7.5 \text{ hr.}), \quad \beta = \beta_1 \quad (6a)$$

$$K(0) = 1.002 K(7.5 \text{ hr.}), \quad \beta = \beta_2 \quad (6b)$$

These results, which predict of the order of a 1 percent reduction in turbidity are to be compared with their results (6) which predicts a 70 percent reduction, 7.5 hours after sunset. If we use an order of magnitude faster removal rate (i.e., $\tau = 7.5$ hours) as suggested by Fischer,

$$K(0) = 1.092 K(7.5 \text{ hr.}), \quad \beta = \beta_1 \quad (7a)$$

$$K(0) = 1.024 K(7.5 \text{ hr.}), \quad \beta = \beta_2 \quad (7b)$$

which is to be compared with their prediction of a sixteen-fold reduction in turbidity.

What is seen from the above is that neither Fischer's nor Esmen and Corn's estimate of the "dry removal" rate fits with Esmen and Corn's model to predict a noticeable change in turbidity. From our own experience, we expect that noticeable variations in turbidity occur because of changes in air masses and/or changes in humidity as well as changes in source strength. The influence of condensation and subsequent evaporation of vapors on aerosol particles is probably especially significant and is a topic under current investigation.

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CONCENTRATIONS OF AEROSOL AND TRACE GASES OVER ST. LOUIS

A. J. Alkezweeny

Two aircraft flights were made over St. Louis to determine the vertical and horizontal distributions of aerosol particles, SO₂, and ice nuclei. It was found that under calm wind conditions the aerosol concentrations were nearly invariant with altitude, but a considerable variation across the city occurs. The SO₂ concentration was in the range between 0.3 to 0.6 ppm. High concentrations of ice nuclei active at -20°C were observed.

During the month of August, 1972, two aircraft flights were made over St. Louis to determine the horizontal and vertical distribution of aerosol and SO₂ concentrations. The investigation was conducted as part of the METROMEX program.⁽¹⁾ The measuring instruments were mounted onboard Battelle's Cessna 411 aircraft. The sampling probe was a stainless tube, 0.5 inches in diameter, expanded to 2 inches over 2 inches in length and then bent at right angle to the aircraft fuselage with 2.5 inches radius of curvature. It was located on upper left side of the aircraft, just behind the pilot's seat, and its intake is about 10 inches from the aircraft skin. The outlet of the probe is connected to various instruments with polyethelene tubing.

The concentrations of particles were measured with a Royco model 220 optical counter. The pulses from the Royco sensor were fed into a two-channel pulse height analyzer, and the concentrations of particles in the size range 0.3 to 1 μ m and greater than 1 μ m in diameter were registered on a Rushtrack recorder. SO₂ concentration was recorded from the output of a Sign X SO₂ Monitor.

A Mee Ice Nuclei Counter was used to determine the concentration of ice nuclei active at -20°C.

The first flight was made on August 28. The sky was clear and the wind speed was estimated at less than 5 mph from the NE. The flight paths were across the city along the west-east direction at three locations, north, central, and south of the city. Each crossing was about 50 miles long. The flight started at an altitude of 1500 feet MSL, and after making three passes, the aircraft climbed to 2000 feet MSL and repeated the flight path. The second flight was made on August 29. Sky and wind conditions were the same as the previous day, except that a temperature inversion was present at about 3000 feet MSL. The same flight paths were made except that each crossing was reduced to about 25 miles long which allowed the measurements to be made at four altitudes 500 feet apart, at 1500, 2000, 2500, and 3000 feet MSL.

Figures 1, 2, and 3 show an example of the data collected on August 29, across the central part of the city; completed data will be reported elsewhere.⁽²⁾ The results were plotted as a function of time, zero

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FIGURE 1. Particle Concentrations at Different Altitudes over St. Louis

time corresponding to a point on west edge of the city, and each minute representing two miles from this reference point (see Alkezweeny⁽²⁾ for map of the flight path). Figure 1 shows the concentrations of particles in the two size ranges for four altitudes. It can be seen that the concentrations are almost the same, for a given point, with height, but a considerable variation exists across the city for the same altitude, which

indicates good vertical mixing, but limited horizontal transport caused by the calm wind condition. The concentrations are higher than those measured on August 28, which should be expected since a much lower temperature inversion base was present.

The results of the SO₂ measurement are shown in Figure 2. The concentration varies from 0.3 to about 0.6 ppm, which is in general agreement with ground sampling network measurements.

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FIGURE 2. SO_2 Concentrations at Different Altitudes over St. Louis

The concentration seems to vary considerably with both vertical and horizontal position. A possible explanation for this behavior is that the sources of SO_2 are located mainly above the ground, in contrast to particle sources which are located both on and above the ground.

The concentration of ice nuclei active at -20°C shows an interesting feature. At 1500 and 2000 feet MSL, two strong peaks are shown at the beginning and end of each path, but they disappeared at levels between 2500 and 3000 feet MSL with a strong peak merging in the center of the path. This peak was also observed on the August 28th flight. At this time, no satisfactory explanation can be offered for this behavior. However, the results indicate that the city is a significant source of ice nuclei.

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FIGURE 3. Ice Nuclei Concentrations over St. Louis

Future work should include measurement of the Aitken nucleus concentration (sizes below $0.1\ \mu\text{m}$), NO_x concentration, and hydrocarbon under different meteorological conditions. This kind of information is useful for the verification of urban pollution models, and may shed light on the mechanisms of gas-to-particle conversions.

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TRACE ELEMENT CHARACTERIZATION OF URBAN AEROSOLS BY X-RAY FLUORESCENCE AND NEUTRON ACTIVATION ANALYSIS

J. A. Cooper, L. A. Rancitelli and T. M. Tanner

Atmospheric aerosols are derived from a large variety of natural and man-made sources. The anthropogenic sources usually dominate in urban locations and are often harmful to man and his environment. The measurement of a wide range of elements in aerosol samples taken under an appropriate variety of conditions can provide very useful information on the origin, movement and interaction of these atmospheric components. Instrumental neutron activation analysis has demonstrated high sensitivity for the direct analysis of a large number of atmospheric elements on air filters but it lacks adequate sensitivity for important elements such as S, Cu, Ni, and Pb. The potential of X-ray fluorescence for measuring these elements and others has been demonstrated, but it has not been applied to an ex-

tensive quantitative analysis of atmospheric aerosols. A highly sensitive and precise instrumental technique employing X-ray fluorescence and neutron activation has been developed which is capable of measuring over 40 elements on air filters used to sample urban aerosols at ground level.

The X-ray fluorescence technique is based on the direct excitation of the elements in the filter with high-intensity radioisotopic sources and their analysis using a Si(Li) detector. The use of 100 mCi ^{55}Fe , ^{109}Cd and ^{241}Am sources provides for the rapid and inexpensive measurement of S, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ge, Br, Sr, Zr, Mo, and Pb in air filters mounted in 35 mm slides. Quantitative results are obtained both by comparing the air filter results to thin film standards and by using those

elements measured by neutron activation as internal standards. The neutron activation procedure involves a one-minute irradiation of the filter samples together with the appropriate standards at a neutron flux of 10^{13} to 10^4 n/cm²/sec. The sample and standards are allowed a 10 minute decay interval to reduce interference from ²⁸Al. They are then counted for 5 minutes on a Ge(Li) gamma-ray spectrometer. This permits the simultaneous measurement of Na, Mg, Al, Cl, Ti, V, Mn and Br. The samples and standards are then reirradiated to an integral neutron exposure of 10^{17} n/cm², allowed to decay for 5 and 20 day periods and counted on a Ge(Li) gamma-ray spectrometer for 30 and 100 minutes, respectively. The 30 minute count permits the measurement of Na, K, Ca, As, Br, La, Sm, and U, while the 100 minute count measures the concentration of Sc, Cr, Fe, Co, Zn, Se,

Rb, Ag, Sb, Ta, Cs, Hf, Eu, Tb, Hg, and Th.

The filter samples which are placed in clean polyethylene capsules for the irradiation are 30 cm² sections taken from a total area of 400 cm² of a Whatman 41 filter paper. The volume of air passed through these filters ranged from 1000 to 1500 m³, while the total aerosol deposited ranged from 0.1 to 1 mg/cm².

Several important observations concerning the origin of elements in urban aerosols can be inferred from the ratio of the trace element concentrations to the Sc concentration. From a comparison of these ratios in granite and diabase with those of the filter, it is evident that Zn, As, Se, Sb, Hg, and Pb levels have been increased by several orders of magnitude while Al, Co, La, Eu, and Th appear to exist at levels compatible with an earth's crust origin.

RADIOACTIVE FALLOUT RATES AND MECHANISMS

The concentrations of trace elements, cosmogenic radionuclides and nuclear-weapons-produced radionuclides were measured in air, precipitation, and seawater to study the rates of atmospheric and oceanic mixing, the rates of air-sea interchange, and the rates and mechanisms of precipitation scavenging. Radioactive debris from the Chinese nuclear test on March 18, 1972 was measured in air and in grass samples collected on two arcs from San Francisco to Seattle and from Texas to Chicago to determine the path of the debris, the rate of deposition on the earth's surface, and the ^{131}I dose to man. The concentrations of plutonium isotopes resulting from nuclear tests and from SNAP-9A nuclear generator burnup were measured in air samples collected from 1963 to 1972 at Richland, Washington.

BEHAVIOR AND CHARACTERISTICS OF RADIOACTIVE DEBRIS FROM CHINESE NUCLEAR WEAPONS TESTS

C. W. Thomas, W. B. Silker and
C. E. Jenkins

Radioactive debris from the Chinese nuclear test of March 18, 1972 was measured in ground level air at Richland, Washington and at Great Falls, Montana in 61 grass samples collected on two arcs extending from San Francisco to Seattle and from Dallas to Chicago, and in milk samples obtained from cows grazing near Richland and Seattle, Washington. The measured radionuclide distributions and deposition patterns were typical of debris injected into the troposphere, although a large fraction of the debris probably entered the stratosphere. Tropospheric debris generally circles the globe in about eleven days and is deposited on the earth's surface within a few weeks. Variations in wind patterns

and precipitation scavenging cause sudden, often dramatic changes in air concentrations and can result in random areas of high contamination on the earth's surface.

The debris from the Chinese test of March 18, 1972 was first detected in ground level air at Richland late in the afternoon on March 21, 1972. Air concentrations increased to a maximum on March 23 and 24 and then decreased. Analysis of the grass samples indicated that the center of the cloud crossed the west coast just north of the Washington-Oregon border. The radioactivity concentrations were generally highest in grass collected in Washington, although local hot spots were observed in Oregon and California. Radioactivity measurements at Great Falls and at other locations by other investigators indicate that after crossing the coast, the cloud moved northeastward penetrating into Canada and then turned southeastward.

In the midwest, activities were generally highest in the grass collected from northern Arkansas to Chicago.

The ^{131}I concentrations in milk and grass were used to calculate the ^{131}I dose to man (Figure 1). The maximum ^{131}I doses to the thyroid calculated from the concentrations in grass were 236 mrem for the west coast and 102 mrem for the midwest. These doses are well below the value of 10 rem recommended as the maximum permissible exposure by the Protective Action Guide for the general population.

The activities of the grass samples were probably only fair indicators of the average deposition, since the activity in each sample probably resulted from a few highly radioactive particles. One grass sample was divided into seven fractions. The seven fractions contained 0.7, 14.4, 0.05, 0.8, 0.05, and 0.05 dpm/g wet weight, respectively.

Several individual particles collected at Richland were isolated and analyzed separately (Table 1). The ratios of the various radionuclides varied considerably from particle to particle. Some particles were enriched in radionuclides having volatile precursors, while others were enriched in radionuclides having refractory precursors. The elemental compositions of two translucent, pale yellow, birefringent particles of 8 to 9 μ diameter were 13 to 18% iron, 14% aluminum, 2% magnesium,

and less than 1% calcium, silicon and manganese.

The air filter samples contained large amounts of 6.7 day ^{237}U produced primarily in thermonuclear devices by a neutron reaction which has a 5.9 MeV threshold. These samples also contained large amounts of 2.35 day ^{239}Np formed by a ^{238}U (n, γ) ^{239}U reaction followed by beta decay. The large amounts of ^{237}U and ^{239}Np present in the debris indicate that the device was a fission-fusion-fission weapon which employed a ^{238}U blanket to enhance its yield.

Studies of the physical and chemical properties of the particles from this nuclear test indicated that the activation products were associated with the surface of the particles, whereas the fission products were incorporated within the particle matrix. The fission products ^{97}Zr , ^{132}Te , ^{131}I and ^{99}Mo were largely insoluble in water, while the activation product ^{237}U was slightly soluble, and ^{239}Np was readily soluble. The fractions of the radionuclides which did dissolve were passed through resin beds to help define their chemical forms. Uranium-237, ^{239}Np , ^{140}Ba , ^{95}Zr , and ^{143}Ce were retained by the cation bed, while ^{99}Mo , ^{131}I and ^{132}Te were retained by the anion bed. The radionuclides present in the milk obtained from cows grazing near Richland, Washington were also in the above ionic forms.

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FIGURE 1. ^{131}I Dose to Humans - Pacific Coast and Midwestern USA - March 1972

TABLE 1. Concentration of Fallout Debris in Individual Particles from Chinese Nuclear Test of March 18, 1972

dpm/particle corrected to March 18, 1972, 0600 GMT

(a) Radionuclide Concentrations in Individual Particles							
Particle Number	^{237}U	^{99}Mo	^{239}Np	^{143}Ce	^{131}I	^{132}Te	^{97}Zr
1	15,000	30,000	81,860	146,000	670	---	773,000
2	17,000	26,600	27,800	131,000	---	---	483,000
3	32,900	155,000	20,000	410,000	770	---	110,000
4	31,800	40,200	190,000	128,000	570	---	---
5	11,000	43,400	164,000	61,000	1,980	---	725,000
6	51,700	34,000	754,000	110,000	1,140	---	---
7	10,900	24,900	362,000	72,200	1,200	2,000	336,000
8	16,500	34,700	281,000	123,000	1,210	4,400	---
9	19,300	39,000	285,000	108,000	2,180	4,560	---
10	44,400	37,800	603,000	74,000	4,300	7,000	---
11	---	---	38,400	---	---	---	---
12	4,930	47,300	99,200	101,000	500	1,350	350,000
13	18,500	18,550	307,300	23,600	3,200	6,430	---
(b) Radionuclide Ratios in Individual Particles							
Particle Number	$^{99}\text{Mo}/^{131}\text{I}$	$^{99}\text{Mo}/^{143}\text{Ce}$	$^{99}\text{Mo}/^{237}\text{U}$				
1	45	0.20	2.0				
2	---	0.20	1.6				
3	201	0.38	4.7				
4	70	0.31	1.3				
5	22	0.71	4.0				
6	30	0.30	.7				
7	20	0.34	2.3				
8	29	0.28	2.1				
9	18	0.35	2.0				
10	9	0.51	.9				
11	---	---	---				
12	95	0.46	9.5				
13	5.8	0.6	1.0				

PLUTONIUM CONCENTRATIONS IN SURFACE AIR AT RICHLAND, WASHINGTON

C. W. Thomas

The plutonium isotopes released by atmospheric nuclear testing are a combination of unreacted plutonium in the device and isotopes of plutonium formed by neutron capture in plutonium and any ^{238}U in the device. The most abundant isotope of plutonium is ^{239}Pu , with lesser amounts of ^{238}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu . The ^{238}Pu is produced by either the reaction of fast neutrons with ^{239}Pu or by the decay of ^{238}Np or ^{242}Cm . Plutonium-238 to ^{239}Pu ratios vary from detonation to detonation depending upon the burst conditions and the material used.

In April 1964, a transit navigational satellite was launched from Vandenberg containing a System for Nuclear Auxiliary Power (SNAP-9A) generator. The satellite failed to achieve a stable orbit and subsequently burned upon re-entry, releasing approximately 17 kCi of ^{238}Pu (~1 kg) at 46 km over the Indian Ocean. This ^{238}Pu almost tripled the global atmospheric ^{238}Pu inventory. Plutonium from this device was first detected in a sample collected by balloon at 33 km over Australia in August 1964. Since that time, it has been detected in stratospheric and tropospheric air from both hemispheres.

The atmospheric concentrations of ^{238}Pu and ^{239}Pu were measured in surface air samples collected at Richland, Washington from 1963 to 1972 as a part of a program to define the

rates of long term stratospheric processes in the northern hemisphere as well as the rates of interhemispheric mixing (Figure 1). The seasonal variations in the concentrations of ^{238}Pu and ^{239}Pu in surface air at Richland were similar to those of other nuclear-weapons-produced radionuclides of stratospheric origin; maximum concentrations occurred in the late spring and minimums occurred in the winter. The rate of decrease in the ^{239}Pu concentration from 1963 through 1967 corresponded to a stratospheric half-residence time of 10 to 11 months, which is the same as the half-residence times calculated from measurements of other radionuclides of stratospheric origin. The ^{239}Pu concentrations remained fairly constant from 1967 to 1972, primarily because of yearly injections of plutonium by thermonuclear tests conducted by the Chinese at Lop Nor (35°N); the contribution from the French tests in the South Pacific (23°S) may also have been significant. Fission product yields indicate that the Chinese used ^{235}U as the fissile material in their high-yield tests of 1967 through 1970. However, large amounts of ^{237}U , ^{239}Np , and ^{239}Pu were produced, indicating that ^{238}U was used as a component part of the devices.

From 1962 through 1965 both the ^{238}Pu and the ^{239}Pu in surface air at Richland came primarily from the

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FIGURE 1. ^{238}Pu and ^{239}Pu Concentration in Surface Air at Richland, Washington

1961-62 U.S.-Russian test series (Table 1). The $^{238}\text{Pu}/^{239}\text{Pu}$ ratio averaged 0.015 in 1964. The ratio increased slightly in 1965, and by the spring of 1966 it had increased to 0.042, indicating that ^{238}Pu from the SNAP-9A burnup was present. The amount of SNAP-9A ^{238}Pu present was

calculated from the ^{238}Pu concentrations and the $^{238}\text{Pu}/^{239}\text{Pu}$ ratios, assuming that the ratio in debris from nuclear weapons tests was 0.015. These calculations indicate that the ^{238}Pu in Richland air from 1967 to 1971 came primarily from SNAP-9A. From 1967 to 1969 the concentrations

TABLE 1. The Average Yearly Ratios of ^{239}Pu , ^{238}Pu , and ^{90}Sr in Surface Air at Richland, Washington

	dpm/KSCM					
	^{90}Sr	^{239}Pu	^{238}Pu		$^{239}\text{Pu}/^{90}\text{Sr}$	$^{238}\text{Pu}/^{239}\text{Pu}$
			Weapons at 1-1/2%	Other		
1962	90.6	1.15 ± 0.169	0.0166	0.0324	0.0127	0.044
1963	119	1.20 ± 0.02	0.0125	0.0015	0.010 ± 0.0043	0.019
1964	80.5	1.02 ± 0.015	0.0127	0.0005	0.0127 ± 0.012	0.015 ± 0.0015
1965	40.0	0.704 ± 0.012	0.0108	0.003	0.0176 ± 0.0072	0.019
1966	22.9	0.325 ± 0.007	0.0038	0.0069	0.0142 ± 0.0055	0.042
1967	7.08	0.125 ± 0.002	0.0023	0.0608	0.018 ± 0.0049	0.415
1968	10.3	0.230 ± 0.003	0.0027	0.0375	0.022 ± 0.0068	0.223
1969	11.4	0.310 ± 0.004	0.0028	0.0511	0.027 ± 0.0076	0.293
1970		0.284 ± 0.004	0.0023	0.0295		0.2066
1971		0.275 ± 0.003	0.0025	0.0073		0.063

of SNAP-9A plutonium at Richland remained fairly constant, indicating that the ^{238}Pu was being transferred across the equator into the northern hemisphere at a rate comparable to the rate at which ^{238}Pu was being deposited on the earth's surface. The ^{238}Pu concentrations showed seasonal variations typical of radio-nuclides of stratospheric origin, so the transfer was probably taking place primarily in the stratosphere.

Concentrations of SNAP-9A plutonium at Richland have decreased rapidly since that time. These results indicate that the stratospheric debris injected into the high stratosphere of one hemisphere will produce high concentrations of the debris in ground level air in the other hemisphere in the spring 1 to 2 years later. These concentrations will then remain constant for about three years before beginning to decrease.

ATMOSPHERIC RADIONUCLIDE CONCENTRATIONS AT RICHLAND, WASHINGTON
AND AT POINT BARROW, ALASKA

C. W. Thomas and J. A. Young

The radionuclide concentrations in surface air are being measured continuously at Richland, Washington (45° N) and at Point Barrow, Alaska (71° N) by filtering large volumes of air through membrane filters and then analyzing the filters by gamma-ray spectrometry. A few of the radionuclides must be separated chemically before analysis. The variation of the concentrations with time depends upon the locations, size and nature of atmospheric nuclear tests. Debris from detonations in the kiloton range generally is not carried above 15 km and will reside in the atmosphere only a few weeks. A large fraction of the debris from thermonuclear weapons in the megaton range is carried above 15 km; the half-residence time for stratospheric debris is about 11 months.

Essentially all of the radionuclides measured in the troposphere from 1962 to 1966 were formed in the 1961-62 test series. Since 1966, the contribution from Chinese and French nuclear tests has increased; at the present time essentially all of the nuclear weapons debris in the atmosphere is from the Chinese and French tests.

The atmospheric concentrations of fallout radionuclides with half-lives greater than 1 year decreased 2 to 10-fold during 1972 at Richland and at Point Barrow, reflecting the lack of

high-yield-weapons testing in the northern hemisphere during 1971 (Figure 1). The only atmospheric test reported in the northern hemisphere during 1971 was a 20 kiloton device tested by the Chinese in November. However, the Chinese detonated 20 kiloton and 200 kiloton devices in January and March of 1972, respectively, and the atmospheric concentrations of short-lived fallout radionuclides increased after March 1972 to levels comparable to those of the past 5 years.

The atmospheric concentrations of the cosmogenic radionuclides ^7Be ($t_{1/2} = 53$ d) and ^{22}Na ($t_{1/2} = 2.6$ yr) have been measured continuously at Richland and at Point Barrow since 1964. Sodium-22 has been produced in significant quantities by nuclear weapons tests, but ^7Be has not, so the ^7Be concentrations have remained fairly constant from year to year. Beryllium-7 production rates in the lower stratosphere at mid-latitudes are several times those at ground level; therefore, ^7Be concentrations at Richland and at Point Barrow have exhibited seasonal variations typical of radionuclides of stratospheric origin. In order to determine the average seasonal ^7Be variations, the average concentration for each month of the year was calculated for the period 1964 through 1972 (Figure 2). At Richland, the concentration varied relatively sinusoidally with July

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FIGURE 1. Ground Air Concentration of ^{155}Eu at Richland, Washington

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FIGURE 2. Monthly Averages of Atmospheric ^7Be Activities from 1964 Through 1972

having the maximum average concentration of 272 dpm/1000 standard cubic meters (KSCM) and December having the minimum average concentration of 102 dpm/KSCM. Average concentrations at Point Barrow were always lower than those at Richland, possibly because the majority of the radionuclide transfer from the stratosphere to the troposphere occurs at mid-latitudes in the region of the tropopause discontinuity. In January the ^7Be concentrations averaged only slightly less at Barrow than at Richland. Concentrations at Barrow increased from January through April at a slower rate than those at Richland, reaching a maximum of 129 dpm/KSCM in April. Concentrations at Barrow then decreased regularly, reaching a minimum of 33 dpm/KSCM in August, possibly because of decreased mixing between middle and high latitudes in the summer. Concentrations at Barrow then increased regularly, climbing to values nearly equal to those at Richland in December. The fall increase may have been due to the increase of meridional mixing.

The trace element concentrations in the air filter samples collected at Richland in 1972 were determined using X-ray fluorescence and neutron activation analysis in order to determine background concentrations and the sources of the trace elements. The ratio of the concentration of

each element to that of iron was calculated and compared to the ratio in diabase (basalt) to determine if the atmospheric concentrations of the element resulted primarily from material from the earth's crust (Table 1). The calcium, titanium, chromium, and manganese in the air probably were due primarily to basaltic dust. Nickel, zinc, arsenic, bromine, and lead demonstrated large contributions from other sources. The bromine to lead ratio of 0.16 is about what would be expected from automobile exhaust.

TABLE 1. Trace Element Concentration Iron Concentration

	<u>Air</u>	<u>Dibase</u>	<u>Air/Dibase</u>
Ca	0.38	1.01	0.377
Ti	0.114	0.082	1.39
Cr	0.0034	0.0015	2.24
Mn	0.018	0.017	1.05
Fe	1	1	1
Ni	0.032	0.001	31.7
Cu	0.005	0.0014	3.57
Zn	0.018	0.0011	17.1
As	0.0018	0.000028	62.9
Br	0.0184	0.000006	3066
Sr	0.007	0.0023	3.17
Mo	0.0003	0.0000006	533
Pb	0.113	0.0001	1130

PRELIMINARY FINDING OF THE CONCENTRATION OF ^{99}Tc
IN ENVIRONMENTAL SAMPLES

C. W. Thomas

Considerable interest has recently been given to the distribution and source of ^{99}Tc in environmental samples mainly because the reactions of Tc are similar to those of iodine in the human body and like iodine, Tc is concentrated in the thyroid. Preliminary results on the concentration of ^{99}Tc in surface air taken October 1970 at Richland, Washington indicated a value less than $0.012 \text{ dpm}/10^3 \text{ scm}$. The fission yields for ^{99}Tc are slightly greater than 6% from both ^{239}Pu and ^{235}U fission, however the specific activity of ^{99}Tc is extremely low due to its half-life of $\sim 210,000$ years. If we assume ^{99}Tc and ^{137}Cs are present in

air at ratios similar to their fission yields, we would estimate the ^{99}Tc concentration in surface air to be $0.0017 \text{ dpm}/10^3 \text{ scm}$. This preliminary measurement would indicate that the ^{99}Tc concentration in surface air could possibly be as much as 7 times higher than would be predicted from fission. Such a value would seem entirely reasonable since substantial fractionation between the 99 and the 137 fission yield chains may occur shortly after detonation. Additional samples are presently being analyzed to determine actual rather than upper limit values for ^{99}Tc .

QN EDDY DIFFUSIVITY AND COSMOGENIC RADIONUCLIDES

W. G. N. Slinn

An experiment is proposed to use vertical profiles of certain cosmogenic radionuclides to infer the vertical profile of the eddy diffusivity in stable layers of the troposphere.

Many authors have attempted to infer the vertical variation of eddy diffusivity from vertical profiles of various trace contaminants; for example, see Roedel,⁽¹⁾ Birot et al,⁽²⁾ Reiter and Sladkovic,⁽³⁾ Bhandari

et al,⁽⁴⁾ Carnuth,⁽⁵⁾ Israël and Hobert,⁽⁶⁾ Bakulin et al,⁽⁷⁾ and Selezneva.⁽⁸⁾ Isotopes of radon and Aitken nuclei are the trace constituents that have been used most extensively but the difficulties with

these are that the sources of the radon isotopes are not uniformly distributed nor is the exhalation rate independent of soil and meteorological conditions, and with Aitken nuclei, the volume production rate from the conversion of trace gases is unknown but is probably a strong function of meteorological conditions.

The purpose of this report is to suggest that an attempt be made to infer tropospheric eddy diffusivity profiles, within stable air masses, from the vertical variation of cosmic-ray-produced (cosmogenic) radionuclides; ^{39}Cl (55 minutes half life), ^{24}Na (15 hours) and ^7Be (53 days) seem to be especially suitable candidates. The major advantage to their use is that the production rates are known;⁽⁹⁾ in fact, they are probably known more accurately than the available data indicates, because in its reduction, apparently no attempt was made to relate the data scatter to the diffusive properties of the atmosphere.

The proposed observational program is the following. An initial time would be identified when a "specific" air mass experienced a significant event which would distort the equilibrium distribution of the radionuclides. For example, the event could be wide-spread frontal precipitation or it could be deep ($\sim 20,000$ feet) fair weather convection. After this event occurs, the vertical profiles of the radionuclides would be measured for a time period as long as possible, in a (Lagrangian) coordinate system moving with the air mass,

in an attempt to follow the equilibration of the profiles.

The calculation of the eddy diffusivity from the results seems to be reasonably straightforward. For the case that cross-wind variations and downwind diffusion are ignorable, and especially, provided that the scale length for the diffusion is small compared with the scale length of the flow (e.g., stable flow not near the surface) then the concentration χ of a specific nuclide is governed by

$$\frac{D\chi}{Dt} = \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial \chi}{\partial z} \right] + P(z) - \lambda \chi, \quad (1)$$

where $\frac{D}{Dt} = \frac{\partial}{\partial t} + U \frac{\partial}{\partial x}$ is just the time derivative of the concentration measured in Lagrangian coordinates; $\epsilon(z)$ is the eddy diffusivity; $P(z)$ is the production rate (nuclei $\text{m}^{-3} \text{sec}^{-1}$); λ is the decay rate and where it is assumed that there is negligible vertical velocity of the air, and negligible settling velocity of the aerosol particles to which the nuclides are attached. Integrating Equation (1) over z we obtain

$$\begin{aligned} \epsilon(z) \frac{\partial \chi}{\partial z} &= \frac{D}{Dt} \int_0^z \chi(z', t^*) dz' \\ &+ \int_0^z [\lambda \chi(z', t^*) - P(z', t^*)] dz' + g(t^*) \end{aligned} \quad (2)$$

where g is an arbitrary function of time and the asterisk on t has been used to emphasize that the z - integration is not necessarily vertical (unless U is independent of height) but it is over that (presumably distorted) air column that has the same

(Lagrangian) initial coordinates. To obtain the arbitrary function $g(t^*)$ we notice that for $z=0$, Equation (2) yields that g is the negative of the flux of the nuclide to the surface. Probably this can be ignored; it can be estimated by equating it to the negative of the surface air concentration multiplied by a deposition velocity of about 1 cm sec^{-1} . Dividing Equation (2) by $(\partial\chi/\partial z)$ at each height gives the desired expression for the eddy diffusivity.

The above technique would seem to be applicable so long as $(\partial\chi/\partial z) \neq 0$. In fact $(\partial\chi/\partial z)$ could be zero in an unstable air mass, after only as little as 10 minutes.⁽¹⁰⁾ For example, if there existed updrafts and downdrafts of 1 m sec^{-1} , then a 1 km unstable air mass would be well mixed in ~ 20 minutes. For this case, even ^{39}Cl could not regain its equilibrium vertical profile, since this requires 0(1 hour). However, this restriction to layers for which $(\partial\chi/\partial z) \neq 0$, is not at all serious for the practical value of the proposed study since the objective is to determine the eddy diffusivity in those layers of the atmosphere where diffusion could impede the dispersal of contaminants, i.e., especially in stable layers. Unstable layers will disperse contaminants relatively rapidly, depending of course on the degree of instability and on the vertical extent. Further, this restriction does not distract from the theoretical value of the results since for an unstable air mass the eddy diffusion concept is inappropriate because the

characteristic length for the diffusion is the same as the characteristic length for the flow which is the depth of the unstable layer.⁽¹⁰⁾

The proposed choice of radionuclides ^{39}Cl , ^{24}Na , and ^7Be , follows not only because of their detectability, but also because of the range of diffusivities to which their concentration profiles would "respond." To see this more clearly, we nondimensionalize time in Equation (1) by λ , and z by a characteristic length of the initial disturbance, say h . For example, h might be the depth of an initially stable layer which was overturned by nonadiabatic lifting. In these nondimensional variables, it is then seen that the relative importance of diffusion is governed by the magnitude of the dimensionless group $\epsilon/(\lambda h^2)$.

In Figure 1 are plotted the lines $\epsilon/(\lambda h^2) = 1$ for the three radionuclides of interest. At distances of an order of magnitude or more below these lines in (ϵ, h) parameter space, diffusion dominates the nuclide profiles and their concentrations would be essentially uniformly distributed over the height h . At distances of an order of magnitude above these lines, diffusion can be ignored and for $\lambda t \gg 1$, the concentration profiles would be simply $P(z)/\lambda$.

From Figure 1 it is seen that the ^{24}Na profile would be most informative in the region of reasonable sizes of initial disturbances (100 m to 3 km) and typical values of the diffusivity in stable layers (0.1 to $10 \text{ m}^2 \text{ sec}^{-1}$). On the other hand the ^{39}Cl profile would almost always

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FIGURE 1. Lines of $\epsilon/(\lambda h^2) = 1$. In the upper region of this (ϵ, h) parameter space, diffusion is ignorable for all three radionuclides, whereas in the lower right corner, diffusion dominates. In the region indicated, ^{24}Na would be most informative for the inference of the diffusivity from the radionuclide's vertical profile.

possess its equilibrium profile in any stable layer. Although it appears from Figure 1 that the ^7Be profile would be sensitive to typical values of diffusivity, and therefore it appears as if it would be a useful tool to analyze the diffusivity, we recall that its equilibration requires a time interval of the order of its half life, which is 53 days. Thus even in a very stable layer, the ^7Be profile would almost always, only reflect any atmospheric instability or cleansing which had occurred several days prior to the measurement period. In spite of this insensitivity, however, it would be valuable to

measure its profile in conjunction with ^{24}Na and ^{39}Cl since it would assist in marking the air mass under study as it pursued its complicated trajectory.

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VERTICAL PROFILES OF AITKEN NUCLEI

A. J. Alkezweeny

Vertical profiles of Aitken nuclei in the atmosphere were determined over several locations in Washington State. It was found that the nuclei concentration either decreases or is nearly the same with altitude up to about 8000 feet. Above this level, the concentration continuously increases, even exceeding its value near the ground in some instances.

Aitken nuclei refer to particles of size below 0.1 microns.⁽¹⁾ Particles in this size range play an important role in cloud and aerosol-physics processes. Cloud condensation nuclei, which are essential for

the formation of natural cloud, are in the size range 0.01 to 0.1 μm .^(2,3,4) Although Aitken particles have a negligible effect on visibility, particles of significant sizes are formed through coagulation.

The first vertical profile of Aitken nuclei was reported by Wigand⁽¹⁾ in 1919. His data shows a decrease in nucleus concentration with altitude. Wigand's data was obtained from one or two balloon flights over central Europe. Similar results were found by Weickmann⁽¹⁾ from an aircraft during 12 flights. Auer⁽⁵⁾ performed similar measurements in the vicinity of Fort Collins, Colorado. The average of four flights he made shows a decrease in the nucleus concentration with altitude up to about 8,000 feet MSL, and then a tendency to an increase above this level. The decrease of nuclei with altitude was also observed by Selezneva⁽⁶⁾ over the USSR. Recently, Kocmond and Mack⁽⁷⁾ investigated the vertical distribution of cloud and Aitken nuclei upwind and downwind of Buffalo, New York. They observed a considerable decrease in the concentrations of both types of nuclei above the temperature inversion layer.

The concentrations of Aitken nuclei, reported here, were measured with a General Electric Aitken nucleus counter. The counter was mounted onboard a Cessna 411 aircraft. The sampling probe used was a stainless tube, 0.5 inches in diameter expanded to 2 inches in diameter over 2 inches in length, and then bent at right angle to the aircraft fuselage with 2.5 inches radius of curvature. The sampler intake was about 10 inches from the aircraft skin. A polyethylene tubing was connected from the counter to the sampling probe.

Several flights were made over several locations in Washington State during the period from March to September, 1972. Examples of the results are presented here. The results of all the flights are reported elsewhere.⁽⁸⁾

On April 19, the vertical profile of Aitken nuclei over the Tri-Cities area was measured at 0830 local time. During the measurement, high cloud was present, estimated at an altitude of 20,000 feet. Figure 1 shows the results of this flight. The nucleus concentration was nearly constant up to about 3000 feet and then decreased with altitude. At about 8000 feet the concentration increased continuously with altitude. Above 10,000 feet the concentration exceeded that near the ground. This characteristic of the profile is typical of most of the measurements. Another example of the increase in the concentration of nuclei with altitude above 8000 feet is demonstrated in Figure 2. The data were collected during a flight started at 14,500 feet MSL above Wenatchee, descending down to about 2000 feet heading toward Pasco on April 25th. The concentrations of Aitken and cloud condensation nuclei (CCN) were recorded. The CCN concentration was measured with an automatic CCN counter⁽⁹⁾ operated at 1 percent supersaturation. Each data point was obtained with the aircraft flying in a straight line and constant altitude. During the flight there was scattered clouds in the area, with cloud base at about 10,500 feet MSL.

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FIGURE 1. Aitken Nuclei Concentration and Temperature as a Function of Altitude

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FIGURE 2. Aitken and Cloud Condensation Nuclei Concentration as a Function of Altitude

The concentrations of nuclei are not corrected for change in altitude. However, the correction would have increased the concentrations reported for higher altitudes.⁽¹⁰⁾ The shape of the nucleus concentration profile suggests two sources of Aitken particles in the atmosphere. One of these arises from nuclei which are transported upward by turbulent diffusion from the atmospheric boundary layer. This contribution to the nuclei vertical distribution is dependent upon the vertical temperature structure. The second source of nuclei is formation within the atmosphere. These nuclei may form from the gas phase by photochemical reaction, or the evaporating portion of a cloud.⁽¹¹⁾

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AIRBORNE MEASUREMENT OF AEROSOL PARTICLE SIZE DISTRIBUTIONS*

A. J. Alkezweeny

The concentration and size distributions of aerosol particles were measured over Seattle and over the ocean. It was found that the concentrations of particles of all sizes decrease considerably above the temperature inversion level.

INTRODUCTION

Knowledge of the concentration and size distribution of aerosol particles with altitude is required to understand their production, removal and transport in the atmosphere. Particles of sizes greater than 0.1 μm are responsible for the attenuation of radiation in the earth's atmosphere. Because of this importance an investigation was performed to determine aerosol particle size distributions at different altitudes over Seattle and over the Pacific Ocean. In this report, results of the investigation are presented and discussed.

The concentration and size distribution of aerosol particles in the range 0.3 to 5 μm in diameter were measured using a Royco Optical System.** This system employs a Royco Optical Sensor Model 220 and a Pulse Height Analyzer (PHA). The pulses from the optical sensor are simultaneously counted and stored in the

memory of the PHA, according to their height, in over 100 channels.

Through a laboratory calibration, the channel numbers are related to particle size. After a given sampling period, the count in each channel can be printed on a paper tape for analysis. The system was mounted onboard a B-23 aircraft operated by the University of Washington. The size distribution obtained in this study should be considered accurate only up to 2 μm particle diameter. Above this size, the concentration is very small and cannot be determined with adequate statistical accuracy with the sample volumes obtained during this study.

The first flight was made on September 29, 1972. The first measurement was taken over west Seattle which is a fairly well developed industrial area. The sky was clear with a temperature inversion base at about 2000 feet MSL (Figure 1). Table 1 shows the concentration of aerosol particles, in the size ranges 0.3 to 1 μm and greater than 1 μm , at different altitudes. It can be seen that the concentration of particles of both sizes decreased sharply above the inversion layer. The size distributions at 1000 and 2000 feet MSL are shown in Figure 2.

* The data presented here is a portion of a study conducted in cooperation with the University of Washington. The complete results of the study will be reported later by Alkezweeny and P. V. Hobbs.

** "A System for Measuring Particle Size Distributions," this report.

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FIGURE 1. Temperature and Dewpoint Versus Altitude over South Seattle

TABLE 1. Concentration of Aerosol Particles at Different Altitudes over Seattle, Washington

Altitude, ft	Concentration of Particles (0.3 - 1) μm	No./cm ³ in the Size Range > μm
1000	6.8	0.35
2000	5.3	0.27
3000	0.07	0.013
3500-4000	0.24	0.037
4000-4500	0.04	0.0028
5000-5500	0.014	0.00035
5500-6000	0.0038	-0-
6000-6500	0.0007	-0-
7000	0.00028	-0-

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FIGURE 2. Particle Size Distributions over Seattle

The second flight was made over the Pacific Ocean about 30 miles west of Hoquiam, Washington. The ocean was quite calm, with bright clear sky. The measurement was taken at several altitudes and started at 7000 feet. Figure 3 shows the size distributions at four different altitudes, and in Table 2 the concentrations of particles of two size ranges are shown for these altitudes.

During the measurements, a strong temperature inversion was present below 2000 feet (Figure 4). The con-

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FIGURE 3. Particle Size Distributions over the Ocean

centration of particles of all sizes seemed to decrease above this level. Comparison of the particles concentration at 200 and 1000 feet indicated the concentration of small particles to be about the same; however, the large-particles concentration decreased by a factor of about 3.6. The reduction in the number of large particles may have been caused by the calm condition of the ocean surface, and hence small production of larger particles (sea spray).

In conclusion, the data presented in this report clearly demonstrate the effect of temperature inversion on the concentration of aerosol particles.

TABLE 2. Concentration of Aerosol Particles at Different Altitudes over the Ocean

Altitude, ft	Concentration of Particles (0.3 - 1) μm	No./cm ³ in the Size Range >1 μm
200	2.5	0.14
1000	2.3	0.055
2000	0.57	0.017
7000	0.023	0.0014

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FIGURE 4. Temperature and Dewpoint Versus Altitude over the Ocean

SOME MEASUREMENTS OF ATMOSPHERIC AEROSOL PARTICLES

W. G. N. Slinn*

Results are shown for number and volume distributions, in the diameter range from 0.3 to 8 μm , of atmospheric aerosol particles measured at ground level sites in the Blue Mountains, Oregon; Monterey Bay, California; Basalt, Colorado and Edwardsville, Illinois.

During a two-week period in the summer of 1972, aerosol particle size distributions were measured at a number of ground level sites in an attempt to gain some appreciation of the characteristics of atmospheric aerosols. These ground level (or actually 2 to 3 m level) measurements were taken at an elevation of 4000 feet in the Blue Mountains of Oregon, on the north shore of Monterey Bay, California, at 6000 feet in the Rocky Mountains near Aspen, Colorado and at Edwardsville, Illinois. The size distributions were obtained with a Royco particle counting system, whose features and calibration are described elsewhere.** This report summarizes some of the observations.

Figure 1 shows the average size distributions measured at three sites. The dependence of $\Delta N/\Delta D$ on D varies from steeper than D^{-4} at the Blue Mountains' site, where we suspect there was considerable contribution from pollen emitted by a nearby dry-grass field, to about D^{-4} at the Monterey Bay site, where we expect

there was considerable influence from nearby pollution sources, to about D^{-2} in the very clean air in this region of the Colorado Rockies.

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FIGURE 1. Number Distribution as a Function of Particle Size. 1. Monterey Bay, California, 8/1/72, 900 hours, 45°F, 80% rh, 5 to 10 mph winds from NE; 2. Meacham National Weather Station, Oregon, 7/27/72, 1300 hr., 75°F, 27% rh, 5 mph winds from NNE; and 3. Basalt, Colorado, 8/3/72, 1900 hr., 85°F, 60% rh, 5 mph winds from NNE.

* Supported by Battelle Institute's Physical Sciences Program.

** See "A System for Measuring Particle Size Distributions" this report.

Figure 2 shows the corresponding volume distributions.

An interesting series of events was monitored on August 3 and 4 at the Colorado site. Figure 3 shows the number distributions measured under calm conditions at 1800 hours local time. The temperature was 85°F and the relative humidity was 60 percent. At about 2000 hours that evening, a large cumulonimbus cloud passed about 5 miles to the north of the sampling site and strong wind gusts (presumably the updraft) buffeted the site. Figure 3 shows the high concentration of 1 to 10 μ particles that were resuspended by the 30 to 50 mph gusts. Figure 3 also shows the background concentration at 1200 hours the next day; temperature, 80°F, relative humidity, 25 percent, wind speeds 5 to 15 mph. At 1930 hours, light rain fell at the sampling site, slightly dampening the

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FIGURE 3. Number Distribution as a Function of Diameter. See text for details. The "downdraft" curve represents the average of 24 independent measurements; "updraft," average of 8.

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FIGURE 2. Volume Distribution as a Function of Particle Size. Same data as for Figure 1.

soil. The winds intensified up to 30 mph and appeared to represent the downdraft from a Cb about 10 miles south of the site. Figure 3 shows that relatively little resuspension occurred, presumably because the soil was damp, and that the concentration of submicron particles was reduced by a detectable amount. Perhaps this represents the influence of precipitation scavenging, although it could be indicative of concentrations in the upper air, and perhaps the concentration would be significantly lower at all sizes were it not for the occurrence of some resuspension. Figure 4 shows the same data as Figure 3, plotted as volume distributions.

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FIGURE 4. Volume Distribution. Same as Figure 3.

Figure 5 shows an interesting evolution of the number distribution of aerosol particles that was monitored during the night of August 12 at Edwardsville, Illinois. The reduction in the concentration of sub-micron particles was expected with the reduction of human activities. The wind was fairly steady in magnitude and direction throughout the night (5 to 15 mph from the SE). The significant rise in the concentration of particles larger than one micron was unexpected.

As a concluding remark, the following should be mentioned. It was apparent during the measurement period that ground level sampling leaves much to be desired if the objective is to obtain characteristic properties of the background atmospheric aerosol. The influence of a dry-grass field here, a campsite there,

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FIGURE 5. Variations in the Number Distribution Observed During the Night of 8/12/72 at Edwardsville, Illinois

or even a sand pile over there is extremely hard to account for. It would be much preferable if the sampling were done by aircraft flying low in the mixing layer. On the other hand, if specific phenomena are to be investigated, such as resuspension or diurnal changes near highways, it is, of course, necessary to choose a ground site and account as best as one can for the perturbations caused by other phenomena.

A COMPARISON OF THE PARTICULATE POLLUTANT LEVELS AT PROPOSED
BASELINE MONITORING STATIONS

J. D. Ludwick, J. A. Young, L. A. Rancitelli and W. E. Davis

It is important that one or more sophisticated atmospheric "baseline" monitoring stations be established at locations where there is a minimum of interference from local sources in order (1) to assess the biological cost of environmental pollution increases from industrial and nuclear operations, (2) to determine the effectiveness of control measures, (3) to assist in identifying contributions from a particular source and (4) to provide data for establishing an acceptable environmental pollutant level among nations.

The site selected should be free from local pollution so comparable measurements can be expected for years to come. Our purpose is to find, instrument and operate a survey station at such a location. At this station, the spectrum of radionuclides and inorganic and organic pollutants will be studied as a function of time and of local and hemispheric meteorological parameters.

A program was initiated to determine the most suitable site for the location of a baseline monitoring station in the northwestern section of the contiguous United States, including Vancouver Island. The criteria for such a location are that it be under the influence of onshore air flow for the maximum possible time during the year and that it preferably have long time periods of constant, clean baseline air. This is particularly important in view of the extended sampling times necessary for the measurement of certain atmospheric pollutants.

Two approaches were taken to this problem as the first step in reducing the number of possible sites. They are the seasonal analysis of 18-hour atmospheric back trajectories for more promising locations, and the quantitative measurement of the particulate pollutant levels at several interesting sites. In order to arrive at definitive conclusions in a

minimum of time, simultaneous measurements were made of the condensation nuclei count at three locations using three Environment One particulate monitors (Model Rich 100). In this way, the influence of possible local origin pollutants could be observed during similar atmospheric conditions at these sites.

In general, the presence of pollutants of local origin has been detected with frequent regularity. Daily fluctuations in particulate pollutant levels often exhibit increases in the morning hours, decreasing in late evening. However, during obvious and prolonged periods of onshore air flow, relatively clean air (very low particulate count) has been observed.

Three sites, located near the Pacific Ocean in western Washington have been compared in some detail. A fourth site is now under study in southwestern Oregon. The locations of the sites follow.

	Station	Location	Elevation, feet	Distance from Pacific Ocean, mile
1	Quillayute, Wash.	47°50'N - 124°34'W	195	4.3
2	Mt. Octopus, Wash.*	47°45'N - 124°10'W	2500	12
3	Kalaloch, Wash.	47°36'N - 124°21'W	200	<1
4	Cape Blanco, Ore.	42°51'N - 124°32'W	200	<1

* Air samples were made from a 40-foot lookout tower on Mt. Octopus.

On August 30, 1972, stations 1 and 2 were put into operation. Under typical summer weather conditions, the Mt. Octopus site was somewhat cleaner. It had occasional 6-hour periods when only 300 nuclei/cm³ were present as compared to double that level at Quillayute airfield. In addition to generally lower levels of particulate pollution, fewer short term, very high pollution sources were evident. These were attributed to occasional motor vehicle traffic or nearby personnel activity. With the increase in air mass changes with increased frontal passages and associated precipitation in late September, both stations exhibited markedly reduced particulate levels. For almost a full week, station 2 remained below the 100 nuclei/cm³ level; station 1 was occasionally that low but it exhibited the ever present very high local fluctuations. With autumnal increase of precipitation which resulted in a reduced forest fire hazard, slash burning operations were permitted in the surrounding forested areas. This immediately raised the nuclei count at station 2 to a level as high or higher than that at station 1. Extended periods of 5,000 to

15,000 nuclei/cm³ were common until frontal passages and associated air mass changes reduced the levels to those previously observed in the minimally polluted onshore flow. These low particulate conditions prevailed throughout September 1972.

Station 3 at Kalaloch, Washington was established on September 18, 1972, for comparison with the existing sites. A complete comparison was not obtained because instrumental difficulties were encountered, particularly at remote station 2. For several weeks, however, station 1 and 3 showed nearly identical particulate pollution levels in continuous background readings and in occasional periods of very high levels lasting for several hours. This was probably due to the atmospheric stagnation conditions that persisted during this period. Baseline levels were about 2000 nuclei/cm³ but rose to 15,000 to 30,000/cm³ for several hour intervals. High surface pressure prevailed over the Pacific Northwest for much of October 1972. Resultant frequent offshore flow, nocturnal temperature inversions and air mass stagnation characterized the predominant weather at sites 1, 2 and 3. Surface mixing

assuming adiabatic movement upward of air parcels would include site 2 on 9 of 31 days. The correlation between 1, 2 and 3 during these periods of mixing in October has not been verified due to instrumental failure at site 2. A further analysis will be performed with meteorological data taken with particulate measurements.

Due to instrumental leasing and modification commitments, little monitoring was accomplished after November 5 at sites 1, 2 and 3. Station 4, at the Oregon site, was however, put into operation on November 22, 1972, and data will shortly be available from that location.

In addition to the particulate nuclei monitor at station 1, a 600 cfm high volume air pump has been established at the airbase. Radioisotope information will shortly be available from filter samples taken using this large air flow system. A variety of other pollutants are scheduled for analysis at station 1 using equipment which will be set up during the next 3 months. These include nitrogen oxide analysis, particulate sizing information and stable element analysis, wet and dry fallout analysis and gaseous iodine-131, tritium and radon analysis.

A SYSTEM FOR MEASURING PARTICLE SIZE DISTRIBUTIONS

A. J. Alkezweeny

A modified Royco particle counting system is described which is capable of simultaneously measuring the concentration and size distribution of particles in the size range 0.3 to about 5 microns diameter in about 100 channels.

INTRODUCTION

Single-particle optical scattering instruments have been used for the determination of aerosol particle concentration and size distribution for many years. Several of these instruments are now available commercially. Size distributions are normally measured in 10 to 15 size ranges. In each size range, the number of particles are counted for

a period of time, then the instrument is sequenced, manually or automatically, to other size ranges. A considerable error is produced in this method if the particle concentration and size distribution are changing during the period of measurement. To avoid this error, the particles must be counted simultaneously in each size range. Available commercial instrumentation of this kind is limited to two to four size ranges.

In this report a system is described which is capable of measuring particle size distribution in the range between 0.5 to 5 μm particles diameter in about 100 channels simultaneously.

INSTRUMENTATIONS

The aerosol particles are drawn into a Royco Model 220 optical sensor (Royco Instruments, Inc., Menlo Park, California), and the 90-degree light scattering from the individual particles is detected by a photomultiplier tube and preamplifier, in the form of a voltage pulse. The voltage pulses are fed into a modified pulse converter Model 171 manufactured by TII (Technical Instruments, Inc., North Haven, Connecticut), then to TII Model 401 D pulse height analyzer (PHA) operated in the Mossbauer mode, and to TII Model 500 A paper tape printer. The sensor is also connected to a two channel PHA with a display and a two channel recorder where, the particle count is displayed and recorded in two size ranges, 0.3 to 1 μm and greater than 1.0 μm . A block diagram of the system is given in Figure 1.

The function of the pulse converter is to provide an interface between the Royco sensor and the PHA. The pulses from the sensor have a rise time of about 100 μsec . The pulse converter, upon receiving the pulse, will amplify it and hold it at its peak, then it will issue a gate pulse to the PHA causing it to read the voltage. With the original design of the converter several prob-

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FIGURE 1. Block Diagram of the Royco System

lems were encountered. These problems are:

1. When the rate of incoming pulses is high, multiple gating pulses per incoming pulse occurred at the shorter delay time setting.
2. When the base line is adjusted, the output signal changes.
3. The converter amplifies the low level signal higher than necessary, and therefore broadens the pulse spectrum.

The first problem was solved by diluting the air sample with a dry filtered air. This dilution was also necessary to avoid coincidence loss. Coincidence loss is the result of more than one particle simultaneously passing through the illuminated volume in the optical sensor. However, if the count rate is less than 15,600 counts per minute, the loss is very small. The last two problems were solved by modifying the converter.⁽¹⁾ The modification was made in accordance with the suggestions of Professor Benjamin Liu of the University of Minnesota.

CALIBRATION

In order to use this system for the determination of particle size distributions, a calibration is needed to establish the relationship between the particle size and the channel number in the PHA, in which the counts are stored. Calibrations were performed in two steps. In the first step, pulses of known voltages were fed into the pulse converter, and the channel numbers, in which these pulses were registered, were observed. The value of these pulses were selected to correspond to particles of size greater than 0.3 microns. The relationship between the pulses heights and channel numbers is shown in Figure 2, curve a. Using this curve and a curve supplied by the manufacturer, relating the output voltage to the particle diameter, the relation between the particles diameter and channel numbers was calculated and plotted as shown in Figure 2, curve b. In the second step, nondispersed polystyrene latex spheres, produced by a Royco Aerosol Generator, Model 256, were introduced into the optical sensor. The locations of each peak, in the PHA channels were plotted on

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FIGURE 2. Relationship Between Output Voltage, Particle Diameter and Channel Number

Figure 2, curve b. The agreement between it and the calculated curve is quite good.

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SEQUENTIAL PROCEDURE FOR MEASURING ^{239}Pu , ^{238}Pu , ^{241}Am ,
 ^{90}Sr and ^{55}Fe IN AIR SAMPLES

C. W. Thomas

To determine the atmospheric transport of the radioisotopes of plutonium, americium, strontium and iron, it is necessary to separate them from filters prior to their specific analysis.

A method was therefore developed for their sequential removal from a large membrane filter. The filter specimen is transferred to a 100 ml platinum dish where ^{236}Pu , ^{85}Sr and ^{243}Am are added as tracers for yield determination along with a 100 mg Sr carrier and sufficient acetone to cover the sample. After evaporation to dryness twice with acetone, the residue is charred. The resulting charcoal is removed by igniting it with a Meeker burner at temperatures below 500°C . Fifty grams of sodium carbonate are added to the residue in the platinum dish and the sample fused at $\sim 900^{\circ}\text{C}$ until the melt is clear (about 10 min). After dissolving the powdered melt using concentrated nitric acid, the solution is refluxed for 1 to 2 hours near boiling temperature. Fuming HNO_3 is added to precipitate the Sr as the insoluble nitrate. After 12 hours the precipitate is filtered and the separated strontium nitrate is dissolved from the residue by washing twice with water. The aqueous wash is then analyzed for Sr using standard radiochemical methods. The filtrate is passed through

a Dowex 1-X8, Cl^- form, 200 to 400 mesh, 1 cm diameter by 20 cm long column that has been equilibrated with 7.7N HNO_3 . The flow rate is adjusted to about 1 ml/min; after passage the column is washed with 7.7N HNO_3 combining wash with filtrate and saving the resulting solution for Am and Fe analysis.

Plutonium is eluted from the column with 6M HCl containing 0.024% HI . Hydrochloric and iodic acid are removed from the eluate containing Pu by evaporating to near dryness with concentrated HNO_3 . The residue is dissolved in H_2SO_4 and NH_4OH is added to the acid endpoint of thymol blue. Plutonium is electrodeposited onto stainless steel or Pt discs from this solution at 1.1 amps for 2 hours. The plutonium isotopes are then measured using alpha energy analysis.

Following dilution of the filtrate which contains Am and Fe, the solution is made basic with NH_4OH . The resulting precipitate is dissolved in HCl , reprecipitated three times with NH_4OH to permit removal of insoluble silica and finally dissolved in 1M HCl . This solution is then passed through a 1 cm diameter by 20 cm ion exchange column (Dowex 50-X8, H^+ form, 200 to 400 mesh, equilibrated with 1M HCl). The column is washed with 3M HCl and the combined wash and filtrate are saved

for ^{55}Fe analysis using standard radiochemical methods. The Am is eluted from the column with 7M HCl and analyzed using standard radiochemical methods. Analytical yields

for ^{90}Sr , $^{238-239}\text{Pu}$, ^{55}Fe and ^{241}Am measured on 70 samples were $77 \pm 12\%$, $73 \pm 17\%$, $85 \pm 10\%$, and $65 \pm 20\%$, respectively.

A MODEL FOR NEAR SURFACE COMPUTATION OF MONTGOMERY STREAM FUNCTIONS

W. E. Davis

A model was developed to compute Montgomery stream functions from bilinear interpolated soundings from four radiosonde sites. The results of comparing the model sounding with observed sounding indicates that within a standard deviation of $\pm 0.05 \times 10^7 \text{ cm}^2 \text{ sec}^{-2}$ accurate stream functions could be computed over a range of 1000 mb to ~925 mb. Implications of these results as well as proposed future work are included.

INTRODUCTION

Macroscale trajectory techniques are being used to predict air-mass transport in conjunction with analyses of deposition, resuspension and scavenging.⁽¹⁾ This report describes results of a project to develop a mesoscale trajectory model that will give improved estimates for following regional as well as point-source air pollution.

Attempts have been made to refine trajectories to a time and distance scale of <12 hours and <400 to 500 nautical miles, respectively. In the last annual report a technique was described which would allow for hourly computations of accurate Montgomery stream functions to be used in an hourly trajectory analysis over

scales ≥ 100 km. In order to make use of this technique, an estimation of the vertical profile of temperature and humidity is needed. The following discussion will describe the model developed to estimate indirectly only the temperature profiles, and will provide a comparison with observed soundings to test the accuracy of the corresponding predictions.

THE MODEL

The model is based on a bilinear interpolation of the pressures of the potential temperature surfaces for two time periods twelve hours apart at four radiosonde sites. The data from the radiosonde sites are first interpolated linearly on

a north-south basis to a grid corner point. The data are then adjusted for linear time changes to the desired hour. At this time the bilin-

ear interpolation is used to construct soundings for each of the hourly stations within the grid (Figure 1).

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FIGURE 1. Grid Coverage of Model. Note SEA (Seattle) and SMP (Stampede Pass) in upper left quadrant.

The model incorporates three alternative modes of calculation. The first uses the calculated surface potential temperature at Stampede Pass, Washington, in the Cascade Mountains, as a vertical point to adjust the sounding at the pass. This is done by moving the bilinear interpolated sounding for Stampede up or down to fit the surface potential temperature at Stampede Pass. Once this is done, a re-calculation is made by using the sounding at Stampede Pass as a fifth sounding.

The second mode is to adjust the sounding at each station to fit the calculated surface potential temperature. This modification will be most useful when a front moves through the grid. To date there has been only limited comparison made between predicted and observed data for this feature. No results on this will be presented.

The third mode of calculation is to divide the grid into two sections: one section east of the Cascade Mountains and the other on the west side of the mountains. The reason for this is that frequently the mountains would block the movement of marine air inland. Generally, the marine air would have lower potential temperatures than the air masses on the east side of the mountains. A comparison of both the east and west calculations with observations has not yet been done.

CASE STUDY

August 1972 was selected as a test situation for two reasons; first, it

was a summer month and second, soundings in conjunction with air pollution studies were available at 1400Z and 2000Z at Seattle.

The result of the comparisons are shown in Figure 2 and Table 1. As can be seen in Figure 2, the errors exceeded $0.05 \times 10^7 \text{ cm}^2 \text{ sec}^{-2}$ between 925 to 900 mb, for the bilinear interpolation. In the case where Stampede Pass was included, the range was 950 to 940. The initial indication is that the bilinear approach is better than the one including Stampede Pass. In looking at Table 1, the reader will note that the method using Stampede Pass actually gives a closer approximation to the sounding; i.e., difference in pressure is less

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FIGURE 2. Mean and Standard Deviation of the Error in Stream Function as a Function of Pressure

TABLE 1. Results of Comparing Montgomery Stream Functions and Pressures of Potential Temperature Surfaces

14Z mb	Mean Difference In Pressure (mb) (P Calculated - P Observed)		Standard Deviation		Mean Difference in Stream Function ($\times 10^{-7}$ cm ² sec ²) (ψ Calculated - ψ Observed)		Standard Deviation	
	Stampede	Bilinear	Stampede	Bilinear	Stampede	Bilinear	Stampede	Bilinear
1000	-4	19	16	19	Assumed Zero	Assumed Zero	Assumed Zero	Assumed Zero
950	4	29	16	22	-0.015	0.010	0.036	0.020
900	8	32	13	25	-0.030	0.022	0.040	0.031
850	4	28	28	35	-0.030	0.040	0.050	0.050
20Z								
mb								
1000	15	11	23	25	Assumed Zero	Assumed Zero	Assumed Zero	Assumed Zero
950	51	45	30	29	0.00	0.00	0.02	0.02
900	37	34	32	41	0.025	0.025	0.05	0.04
850	5	1	44.8	52.6	0.035	0.041	0.088	0.088

between observed and the predicted. The reason for the discrepancy is that the model does not include the effects of water vapor. When an estimate of the water vapor is included, it will cause a positive increase in the mean differences and produce a closer approximation to the observed sounding by the modification using Stampede Pass at 1400Z. The inclusion of water vapor in the calculations will cause problems in the 2000Z data since any inclusion will cause the mean to be increased which will result in a decrease in the height range over which the calculations can be made for both the bilinear as well as the modification using Stampede Pass. At present, methods are being developed which may improve the data at 2000Z; i.e., diabatic effects from surface heating may be

estimated, resulting in a better approximation of the observed sounding.

DISCUSSION OF RESULTS AND CONCLUSIONS

The error limits on the model results will allow estimation by the model of the geostrophic wind within ± 5 m/sec for a 100 km interval at 45° latitude or 1 m/sec for a 500 km interval. Thus a model which can produce accurate stream function estimates over a layer from 1000 mb to 925 mb, or ~ 800 m above surface, has been developed. At present, alterations are under way to include water vapor as well as initial reduction of data for November, 1972. Further changes may be warranted in order to better estimate the surface heating at 2000Z.

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1972 MICROCLIMATOLOGICAL MEASUREMENTS ON THE ARID LANDS ECOLOGY RESERVE

John M. Thorp

Daily and monthly microclimatology measurements on the Arid Lands Ecology Reserve continued on schedule during 1972. Micrometeorological and microclimatological data from various ALE Reserve study plots are being summarized by computer and distributed monthly to Reserve investigators.

Measurements of climatological parameters continued through 1972 on the Arid Lands Ecology Reserve (Figure 1). Monthly measurements of precipitation, maximum and minimum air temperature and 6 dm soil temperature were made at 26 sites comprising the network of ALE Reserve microclimatological stations. Bioyear (Oct., 1971-May, 1972) precipitation totals (Figure 2) were generally the lowest observed in four years of record. Four sites at the east margin of the Reserve did receive slightly higher amounts of precipitation than they did in bioyear 1971. Totals for

bioyear 1972 ranged from 11 cm of water in upper Cold Creek Valley to nearly 20 cm at station 10 just north of the Rattlesnake Hills crest.

The micrometeorological and microclimatological data from various study plots on the ALE Reserve provide additional measurements of air temperature, relative humidity and soil temperature. All such data are summarized by computer and transmitted monthly to interested investigators. Table 1 shows the current microclimatological instrumentation on the Reserve.

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FIGURE 1. Arid Lands Ecology Reserve

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FIGURE 2. Arid Lands Ecology Reserve, October 1971-May 1972, Bioyear Precipitation (CM of Water)

TABLE 1. Climatological Instrumentation of the ALE Reserve -- 1972

Station	No.											
		-60 cm Soil Temp.	-40 cm Soil Temp.	-20 cm Soil Temp.	-10 cm Soil Temp.	-5 cm Soil Temp.	1 ft. Air Temp. Max-Min.	4 ft. Air Temp. Max-Min	4 ft. Relative Humidity	Precipitation 5" capacity gage	Precipitation Standard Gage	Precipitation Heated, Recording Gage
D Daily W Weekly M Monthly												
Upper Cold Creek	1	M					M			M		
Rattlesnake Springs	2	M					M	D	D	M		M
Lower Snively	3	M	D		D		M			M	M	W
Upper Snively	4	M	D	D	D		M	D	D	M	M	W
Benson Ranch	5	M	D		D		M	D	D	M	M	
Uper Benson	6	M					M			M		
Alluvial Fan.	7	M					M			M		
Lower Lark Spur	7a	M	D		D		M	D	D	M	M	
Lark Spur	8	M					M			M		
Benson Basin	9	M					M			M		
Anderson Rim	10	M	D		D		M	D	D	M	M	
Anderson Field	11	M					M			M	M	
Hopsage	12	M					M			M		
Telemeter	13	M					M			M		
Rattlesnake Crest, NE	14						M			M		W
Rattlesnake Crest,	15						M			M		
Rattlesnake Crest, SW	16						M			M		
Winterfat	17	M					M			M		
Calochortus	18	M					M			M		
Beetle Plot 7	19	M					M			M		
Lower Cold Creek	20	M					M			M	M	
Beetle Plot 3	21	M					M			M		
Beetle Plot 4	22	M					M			M		
ERC Y	23	M					M			M		
Beetle Plot 5	24	M					M			M		
Beetle Plot 6	25						M			M		
I.B.P. Plot 1	114	D			D			D	D			W
I.B.P. Plot 2	115			D		D						
I.B.P. Plot 3	116	D			D							
I.B.P. Plot 4	117			D		D						
Rattlesnake Irrigation	123		D		D							
Rattlesnake Irrigation	124		D		D							
Rattlesnake Irrigation	125		D		D							
Lower Rattlesnake	126		D		D							
Rattlesnake Springs 1E	127		D		D			D	D			W
Rattlesnake Observatory	113							D	D			

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Technical Information (5)
Technical Publications (2)