PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1974 TO THE
USAEC DIVISION OF BIOMEDICAL
AND ENVIRONMENTAL RESEARCH
PART 3 ATMOSPHERIC SCIENCES

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

FEBRUARY 1975

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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1974
TO THE
USAEC DIVISION OF BIOMEDICAL AND ENVIRONMENTAL RESEARCH

PART 3 ATMOSPHERIC SCIENCES

By

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

February 1975

On January 19, 1975, research and development programs of the U.S. Atomic Energy Commission (AEC) became part of the newly formed Energy Research and Development Administration (ERDA). In this report, since it refers to work done in 1974, most references are to AEC programs.

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PREFACE

The Annual Report for 1974 to the U.S. Atomic Energy Commission's Division of Biomedical and Environmental Research is organized by major program categories according to our schedule-189 submissions. Each part is directed toward a particular DBER Research and Development Program: Part 1 to Biomedical Programs, Parts 2 and 3 to Environmental Programs, and Part 4 to Physical and Technological Programs. Each part of the Annual Report comprises project reports authored by scientists from several research departments, reflecting the interdisciplinary nature of the research effort. The Annual Report consists of four parts:

Part 1	Biomedical Sciences	Program Manager: R. C. Thompson Editor: D. L. Felton
Part 2	Ecological Sciences	Program Manager: B. E. Vaughan Editor: J. L. Engstrom
Part 3	Atmospheric Sciences	Program Manager: C. E. Elderkin Editor: E. L. Nichols
Part 4	Physical and Analytical Sciences	Program Manager: J. M. Nielsen Editor: G. M. Garnant

Reports previously issued are as follows:

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1951	HW-25021, HW-25709
1952	HW-27814, HW-28636
1953	HW-30437, HW-30464
1954	HW-30306, HW-33128, HW-35905, HW-35917
1955	HW-39558, HW-41315, HW-41500
1956	HW-47500
1957	HW-53500
1958	HW-59500
1959	HW-63824, HW-65500
1960	HW-69500, HW-70050
1961	HW-72500, HW-73337
1962	HW-76000, HW-77609
1963	HW-80500, HW-81746
1964	BNWL-122
1965	BNWL-280, BNWL-235, Vol. 1-4
1966	BNWL-480, Vol. 1, BNWL-481, Vol. 2, Pt. 1-4
1967	BNWL-714, Vol. 1, BNWL-715, Vol. 2, Pt. 1-4
1968	BNWL-1050, Vol. 1, Pt. 1-2, BNWL-1051, Vol. 2, Pt. 1-3
1969	BNWL-1306, Vol. 1, Pt. 1-2, BNWL-1307, Vol. 2, Pt. 1-3
1970	BNWL-1550, Vol. 1, Pt. 1-2, BNWL-1551, Vol. 2, Pt. 1-2
1971	BNWL-1650, Vol. 1, Pt. 1-2, BNWL-1651, Vol. 2, Pt. 1-2
1972	BNWL-1750, Vol. 1, Pt. 1-2, BNWL-1751, Vol. 2, Pt. 1-2
1973	BNWL-1850, Pt. 1-4
1974	BNWL-1950, Pt. 1-4

W. J. Bair
Program Director, Life Sciences

FOREWORD

With expanding energy development and use, serious associated air pollutant levels are anticipated on local and regional scales, exceeding ambient air quality standards for periods of hours to days depending on their distribution and accumulation in the atmosphere. Consequently, further understanding of atmospheric transport processes in those time and space scales producing the pollutants is needed. This knowledge is the basis on which assessment methodologies must be developed and applied.

The most urgent air pollution problems must be identified and continually reassessed to set priorities for more detailed investigations. To accomplish this, first of all pollutant distributions in the atmosphere are being measured in regions of potentially significant pollution, as well as in "background" regions, to evaluate hemispheric levels of both fossil fuel effluents and nuclear contaminants. The detailed airflow and dispersion behavior of pollutants distributed locally as well as over several states is being investigated to determine dependence on meteorological conditions and terrain features. Also under investigation are the significant alterations of physical and chemical characteristics of pollutants which occur during transport, varying as a function of atmospheric conditions. These modifications can strongly influence the effectiveness of pollutant removal from the atmosphere as well as the seriousness of the ultimate health or environmental impact. The removal of the pollutants by wet means in clouds and rain, and by dry means through direct interaction with vegetation and surface elements, is being studied extensively. Delivery of pollutants damaging to ecosystems and the budgets of pollutants remaining in the air as inhalation hazards depend strongly on the effectiveness of these removal processes. The deposition of contaminants from nuclear energy facilities is of continuing concern, as is the resuspension potential of deposited, long-lived radionuclides like plutonium, and continued emphasis is being placed here.

As attention has turned to investigation of larger scale processes such as precipitation scavenging by cloud systems and evolution of urban pollution plumes downwind of large industrial cities, experiments conducted from PNL's Cessna 411 aircraft have increased in both frequency and complexity. Experiments planned for next year require even greater aircraft capability, incorporating concurrent measurements of a number of physically and chemically interacting aerosols and precursor gases in addition to related meteorological and cloud physics variables. The experiments will require the use of a DC-3 which was recently acquired and is presently being equipped with the necessary instrumentation.

The past year was particularly productive with regard to reporting research results. More open literature publications were written and more scientific conference papers were presented than in any previous year for PNL in Atmospheric Sciences. An international symposium, "Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants - 1974" was organized and held at PNL; staff members contributed ten papers on results from the Atmospheric Sciences Program. PNL staff also assisted in planning the American Meteorological Society's "Symposium on Atmospheric Diffusion and Air Pollution" and seven PNL papers were presented there. Strong participation in the "Precipitation Scavenging Symposium-1974," where six PNL papers were presented, reflected the continued leadership in this important research area, which has gained growing interest since the first such symposium was organized and held at PNL in 1970.

Significant results obtained during the year include experimental determination of some overall efficiencies for removing particulates from the atmosphere by both frontal storms and convective storms. Initial model descriptions of the in-cloud removal process were made for the convective storm case. The changes in pollutants character occurring during transport prior to removal were described. It is particularly important in describing the effectiveness

of the removal mechanisms to specify the changing aerosol budgets and the reactive products of the pollutants as they interact with one another, with clouds and precipitation, and with surface elements.

The dry deposition of particulates has been modeled as a function of particle size, surface roughness, and wind characteristics from wind tunnel experimentation. Limited comparison of the model, projected to environmental conditions, with full scale plume diffusion/deposition experiment results showed reasonable agreement. Future work should extend these comparisons for verification over a wide range of surface roughness conditions for selected particle size ranges and meteorological conditions.

The resuspension work has broadened considerably in the past year, with experiments conducted over forests as well as desert and plains vegetation. A particularly significant accomplishment is the initial modeling work which permits gross estimates of resuspended airborne concentrations downwind of various sources of surface contamination and provides a framework for evaluating experimental results.

The synthesis of results from experimental and modeling investigations of the various processes acting during atmospheric transport was also begun during the past year. An overall model for assessing consequences on a mesoscale or multistate scale is being assembled. This model will play an increasingly significant role in integrating our research results, to solve serious air pollution problems and identify priorities for future research.

C. E. Elderkin, Associate Manager
Atmospheric Sciences Department

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CHARACTERIZATION OF SOURCES AND AMBIENT POLLUTANTS

The nature and concentrations of radioactive and chemical airborne contaminants must be known before their consequences to the environment can be assessed objectively. Knowing the pollutant characteristics is essential to estimate downwind concentrations of particles and gases, rates of deposition on the earth's surface, possible resuspension, and the biological significance to the receptor of interest. During the past year, Atmospheric Sciences and Radiological Sciences have used an array of very sensitive, low-level analytical methods to measure the particle size distributions and atmospheric concentrations of trace elements, chemical compounds, and radionuclides; these measurements have been made downwind of point and area pollution sources, and also at an uncontaminated site on the Olympic Peninsula. The relationships between contaminant concentrations and meteorological conditions are also being studied. Even more sensitive analytical techniques are being investigated, since many contaminants still cannot be analyzed with sufficient accuracy.

- **SURVEY OF RADIOACTIVITY AND CHEMICAL POLLUTANTS***
- **RADIOACTIVE FALLOUT RATES AND MECHANISMS**
- **ATMOSPHERIC AEROSOLS AND TRACE GASES**

**This list, both here and in the following sections, denotes the sponsoring programs for the research reports in the section.*

QUILLAYUTE AIR REFERENCE STATION

J. D. Ludwick, C. W. Thomas, L. L. Wendell, and T. D. Fox

A large number of radioactive and non-radioactive airborne constituents are being measured continually at our coastal air-reference station located in northwestern Washington State. Important correlations have been observed between many of these materials and also with atmospheric parameters. The apparent relationships between changing concentrations and meteorological parameters are being tested using more thorough techniques of investigation, including back trajectory computer analyses.

INTRODUCTION

The permanent air sampling and monitoring station located at the Quillayute Air Base in northwest Washington State is in its twenty-first month of operation and has completed its first full year of data collection.^{1,2} Considerable expansion and improvement of this facility has taken place during the past year. The continuous air monitoring program which was initiated with the condensation nuclei detector and ozone monitor has been expanded to include real-time measurement of NO and NO_x, CO as well as CH₄ and total hydrocarbons.

Last year, the improved results obtained in going to a 15-m air intake stack prompted our decision to construct a permanent 32-m-high by 15-cm-diameter sampling stack. An in-line filter housing was designed to fit near the bottom of the stack for sampling a variety of stable elements and radionuclides. A high-volume pump (2.25 m³/min) established a linear velocity of 760 m/min across the face of an IPC filter for high-efficiency particulate collection. A "Y" joint located just upstream of this filter drew air for other major monitoring and sampling purposes.

Samples are collected over a 6.5-cm-diameter filter for this high-volume system. This has greatly increased our sensitivity for daily measurements of stable elements by x-ray fluorescence techniques. In addition, these daily samples contain sufficient ⁷Be and some other radioisotopes to allow their accurate measurement for some of our daily correlation analysis. Generally some 15-20 stable elements are measured by x-ray analysis on a daily basis. Selected samples of particular interest are analyzed for an additional 10 to 15 elements by neutron activation.

Our high-volume air sampling unit consisting of a 17 m³/min, IPC filter system is used for some 30 radionuclides. This system is also used for other special studies. A cost saving of several thousand dollars yearly was realized when the large membrane-type filter head was replaced by an in-line IPC filter holder similar to the one just described. A 12-m stack was mounted on top of this system and may be seen to the right of the 32-m stack and retainer tower in Figure 1.

In order to better coordinate certain pollutant sampling with wind

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FIGURE 1. Quillayute Air Reference Station.

information, a small wind station was mounted at a 22-m height at the air reference station. This provides supplemental as well as comparative information with the hourly surface values reported by the U.S. Weather Bureau at the Quillayute Airport site.

There has been essentially no land use change in the western fetch to the ocean, some 3 miles away, since the establishment of the air-reference station at its present location. In general, no land use change is noticeable within 10 miles of the site in any direction.

A considerable effort was expended this year in improving the method of handling the huge amount of data coming from measurements of the many natural and anthropogenic pollutant materials on the daily and weekly analytical programs. Most of the data is presently punched onto cards which are then available for comparison or individual readout in simplified computer programs. Occasional instrumental failures have resulted in incomplete data for reactive gases. This may be a recurring problem; however, we expect to minimize failures. A description of the particulate sampling equipment is given in the following table.

TABLE 1. Particulate Air Sampling Devices at Quillayute.

<u>Materials Measured</u>	<u>Collector</u>	<u>Collection Interval</u>
Radionuclides (~ 30)	17 m ³ /min Pump	5 day
Radionuclides (trajectory analysis)	2-1/4 m ³ /min Pump	1 day
Radionuclides Size distribution	1.13 m ³ /min Andersen impactors	7 day
Stable Elements	2-1/4 m ³ /min Pump	1 day
Stable Element Size Distribution	0.028 m ³ /min Andersen impactors	7 day
Special Studies (⁵⁵ Fe, organics)	17 m ³ /min Pump	1 day

STABLE ELEMENTS IN AIR SAMPLES AT QUILLAYUTE

Airborne particulates are collected for daily analysis at Quillayute using the 2.25 m³/min pump that pulls air down the 32-m stack. This provides information useful in identifying aerosol sources, tracing air masses, evaluating potential health hazards and establishing a reference level to evaluate the effects of man's future activities. Daily x-ray fluorescence analysis of each sample, along with supplementary neutron activation of selected samples provides useful information on over 30 stable elements.

Figures 2-4 illustrate the results of x-ray fluorescence analysis from selected daily samples while Figure 5 indicates the minimum detectable levels of the elements routinely undergoing analysis. The minimum detectable levels obtained are well below the clean air values observed from remote locations around the world (see 1973 annual report, p. 116). This illustrates the adequate sensitivity of the sampling and analytical method. Figure 2 is typical of anthropogenic and natural sources of air pollution and was obtained during meteorological conditions conducive to sampling locally polluted air. Characteristic results obtained dur-

ing such atmospheric conditions include high lead, sulfur, iron and related material values and low relative chloride values.

Figures 3 and 4 illustrate typical air concentrations during periods of clean-air influx, evidenced meteorologically by corresponding periods of on-shore air flow. The lead value and general elemental pattern are very low while values of chloride indicate the relative amount of lower level sea air present at the sampling site. In general, low lead values are accompanied by high chloride values due to ocean trajectory air. In these cases, Na/Cl ratio is characteristic of sea salt. In some specific instances, however, low lead and other materials are accompanied by relatively low chloride values, possibly the result of subsiding clean air masses.

Excellent correlations have been obtained between meteorological parameters indicating clean-air sources and low values of elements of anthropogenic origin. Correlations are apparent between the natural aerosol composition and air masses whose shorter range trajectory has been northerly from remote land regions. The absolute values obtained for almost all of the materials measured during periods of on-shore flow from

Neg 747594-16

FIGURE 2. Quillayute Aerosol.

Neg 747594-17

FIGURE 3. Quillayute Aerosol.

Neg 747594-18

FIGURE 4. Quillayute Aerosol.

Neg 747594-19

FIGURE 5. Minimum Detectable Limits.

short range oceanic trajectory air were comparable to the lowest values reported throughout the world. Figures 6-12 contain average daily air-lead concentrations at the Quillayute reference station, determined by x-ray fluorescence analysis. Values less than about 1.5 Ng/m^3 can be considered as representing very clean air. Several interesting features of the stable element values and their comparative ratios are presently under study. These relate to

the ultimate source of the air mass origin and its upwind history. Low lead values are almost always associated with the influx of a fresh air mass preceded by frontal activity. Very high lead values are apparent only during stagnation conditions at the sampling site. The average lead value for a clean air mass at Quillayute is about 0.8 Ng/m^3 , and this may be considered representative of the hemispheric values at this latitude.

Neg 747594-7

FIGURE 6. Average Daily Lead Values at Quillayute, Washington, March 1974.

Neg 747594-4

FIGURE 8. Average Daily Lead Values at Quillayute, Washington, May 1974.

Neg 747594-1

FIGURE 7. Average Daily Lead Values at Quillayute, Washington, April 1974.

Neg 747594-5

FIGURE 9. Average Daily Lead Values at Quillayute, Washington, June 1974.

Neg 747594-6

FIGURE 10. Average Daily Lead Values at Quillayute, Washington, July 1974.

Neg 747594-3

FIGURE 11. Average Daily Lead Values at Quillayute, Washington, August 1974.

Neg 747594-2

FIGURE 12. Average Daily Lead Values at Quillayute, Washington, September 1974.

RADIONUCLIDE CONCENTRATIONS IN AIR SAMPLES AT QUILLAYUTE, WASHINGTON

The concentrations of a wide spectrum of fallout and natural radionuclides were measured in surface air at the Quillayute, Washington reference station during 1974. Aerosol samples were obtained by passing large volumes of air ($17 \text{ m}^3/\text{min}$) from a 12-m-high stack through IPC filter paper for 7 day periods. IPC filters had been tested and found to be essentially absolute for removing the airborne radionuclides at filtering face velocity of greater than 670 linear m/min. The radionuclide concentrations measured at Quillayute during 1974 were about 1/3 the concentrations measured at Richland, Washington. This relatively large difference is believed due to the orographic lifting of the Olympic and Cascade mountains with subsequent vertical mixing of higher altitude and surface air prior to sampling at Richland. Since the fallout radionuclide concentration increases with altitude, the vertical mixing caused by movement over the mountains could result in higher radionuclide concentrations at Richland. The concentrations of radionuclides associated with weapons debris observed in surface air in 1973 at Quillayute, Washington were the lowest recorded in the northwestern United States since measurements began in 1961. Previous data has shown that material inserted into the stratosphere during one year will be measured in surface air the following year at the mid-northern latitudes. During 1974 the concentration of weapons-associated radionuclides in air at Quillayute, Washington increased 5-20 times over that measured during 1973 as a result of the June 1973 nuclear tests (see Figure 13). In June 1974, a similar Chinese nuclear test was conducted. Most of the debris was injected into the stratosphere, and the fresh debris in the troposphere from this test was about one-fifth of that measured after the June 1973 nuclear test.

Two high-volume Andersen Cascade impactors with a total air flow of $1.13 \text{ m}^3/\text{sec}$ are operated at Quillayute and allow our measurements of the radionuclide distribution versus

FIGURE 13. The Concentration of ^{144}Ce in Surface Air at Quillayute, Washington.

particle size. The impactors separate the atmospheric aerosol into five fractions: greater than 7 microns, 3.3 to 7.0 microns, 2.0 to 3.3 microns,

1.1 to 2.0 microns, and a backup filter of less than 1.1 microns. For the radionuclide particle size studies 8 weeks of samples were combined. Results from measurements of these fractions are shown in Table 2. Measurements made during 1973 show 83% of the ^7Be activity, 58% of the ^{137}Cs activity, and 44% of the fresh medium-lived nuclear debris (from the June 1973 Chinese nuclear test) to be associated with particles less than 1.1 micron. During 1974 our measurements showed almost identical results for ^7Be but a much smaller size range for ^{137}Cs . About 82% of the ^7Be activity and 71% of the ^{137}Cs activity was associated with particles less than 1.1 micron. This reflects the fact that the ^{137}Cs was almost entirely of stratospheric origin. These data together with concentration data indicate that most of the ^{137}Cs in surface air was from stratospheric nuclear debris inserted in 1973 by the Chinese test. Thus, the stratospheric input completely overwhelmed the small contribution of fresh nuclear debris from the 1974 nuclear test, which would be associated with relatively large tropospheric aerosol particles.

TABLE 2. Distribution of Atmospheric Aerosols at Quillayute, Washington.

		SEPT OCT 73	NOV DEC 73	JAN FEB 74	MAR APR 74	MAY JUN 74	JULY AUG 74	$\bar{x}$
^7Be	1	3.84	8.31	1.21	3.39	3.08	1.87	3.61
	2	3.13	2.17	2.08	2.28	3.25	5.94	3.14
	3	2.97	2.20	3.44	3.46	3.19	5.58	3.47
	4	7.51	4.54	9.40	7.66	9.33	11.3	8.29
	5	82.6	82.8	83.9	83.0	81.3	75.6	81.5
^{95}Nb	1	7.99	11.9	1.45	3.65	2.31	2.83	5.02
	2	10.7	5.61	3.14	2.64	3.78	6.50	5.40
	3	12.1	3.63	4.22	4.34	3.80	5.19	5.55
	4	8.28	12.5	12.8	12.2	12.7	11.9	11.7
	5	61.0	66.3	78.4	77.1	77.5	73.4	72.3
^{137}Cs	1	7.10	15.2	2.94	3.08	2.97	3.88	5.85
	2	11.8	9.60	4.14	2.79	3.90	5.70	6.32
	3	15.6	6.78	5.71	4.21	4.80	5.25	7.06
	4	17.2	7.91	20.2	15.0	13.4	14.0	14.6
	5	48.3	60.5	67.0	74.9	74.4	71.1	66.0
^{144}Ce	1	10.0	11.4	2.01	3.05	2.30	2.37	5.18
	2	10.8	13.2	5.66	2.31	3.14	4.58	6.62
	3	14.3	5.97	4.02	4.54	3.94	5.42	6.37
	4	9.93	6.96	12.7	12.2	12.6	12.5	11.1
	5	54.9	62.5	75.6	77.9	78.2	75.3	70.7

THE CONTINUOUS DATA ACCUMULATION PROGRAM

Daily comparisons between the various parameters measured showed several interesting features (see Figures 14-21). Where obvious correlations exist in these data, more rigorous meteorological analyses were conducted to relate these correlations back to trajectory, or other information.

A great deal of conjecture was, of course, possible from this information; however, we attempted to follow up in a firmly directed manner.

For example, ^{95}Nb , ^{144}Ce and ^7Be , measured on a daily basis, showed a fairly good correlation. The tropospheric fallout is usually minimal and one can relate fluctuating levels to stratospheric input. About 30% of ^7Be is produced in the troposphere. Upon close examination of the data it appears that the ^{95}Nb and ^{144}Ce concentrations follow each other almost exactly, whereas the ^7Be behaves similarly but not identically. In

the September data, very high values of ^7Be were observed, in comparison to low levels of fallout radionuclides. This latter fact can be explained by the annual decrease in airborne stratospheric mixing at this latitude. This is attributed to the rather rapid removal by deposition of particulate material from stratospheric injection during the late spring and early summer. The high ^7Be values result from tropospheric production and perhaps its concentrating in the lower atmosphere during stagnating conditions, which prevail during a good deal of September at Quillayute, Washington.

Another very distinct feature is the correlations of radionuclide and the maximum ozone concentrations during the same time period. At Quillayute, these are attributed to the subsidence of upper level air, whose origin can eventually be traced to the stratosphere. It is interesting that these particulate materials are of a sufficiently conservative nature to show a good correlation with gaseous ozone.

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FIGURE 14. Chemical and Physical Comparisons with Meteorological Data, February 1974.

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FIGURE 15. Chemical and Physical Comparisons with Meteorological Data, March 1974.

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FIGURE 16. Chemical and Physical Comparisons with Meteorological Data, April 1974.

Neg 747594-12

FIGURE 18. Chemical and Physical Comparisons with Meteorological Data, June 1974.

Neg 747594-11

FIGURE 17. Chemical and Physical Comparisons with Meteorological Data, May 1974.

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FIGURE 19. Chemical and Physical Comparisons with Meteorological Data, July 1974.

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FIGURE 20. Chemical and Physical Comparisons with Meteorological Data, August 1974.

An additional observation is that clean air masses, identified by their low lead and natural pollutant values and low levels of ozone and radionuclides, are therefore indicative of lower level air masses. This is further verified by the high chloride values associated with these systems and a Na:Cl ratio typical of sea salt. Thus such an air mass must have had considerable contact with ocean sprays.

The effects from precipitation scavenging are also evident in the accumulated data. Many new air masses moving into the Quillayute area bring upper air whose radionuclide and ozone levels are high. This situation is apparent beginning on March 25, 1974. One can see the ozone and radionuclide levels increase with inception of frontal passage. The ensuing precipitation and winds are illustrated in

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FIGURE 21. Chemical and Physical Comparisons with Meteorological Data, September 1974.

Figure 15. Although the high ozone levels persist essentially throughout the remainder of the month, unusually low values for the radionuclides were observed for March 28 and 29. The continuous precipitation throughout this period evidently depleted the particulate radionuclides from the incoming air mass. This effect is difficult to observe from the condensation nuclei values since they are often affected by local contamination sources (which predominate when south to easterly winds persist).

In general, the wealth of data accumulated by the continuous operation of the Quillayute air reference station will provide unique opportunities for study of the correlations of large scale meteorological processes with natural and anthropogenic tracer pollutants.

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METEOROLOGICAL CONSIDERATIONS IN THE INTERPRETATION
OF THE TIME VARIATION OF RADIOACTIVITY AND
CONCENTRATION OF OTHER POLLUTANTS

T. D. Fox, L. L. Wendell, and J. D. Ludwick

Atmospheric analysis is being performed in association with ongoing pollution concentration measurements at Quillayute, Washington. Tentative relationships have emerged from comparisons of concentration data and meteorological data. These relationships are being tested using more thorough techniques of investigation, including back trajectory computer analyses.

INTRODUCTION

A permanent air monitoring station has been established at Quillayute, Washington, for continuous monitoring of trace atmospheric materials.^{1,2} Analysis of large scale atmospheric processes is being performed in association with these measurements, to attempt to define and further understand the variations which are seen in the concentrations of the various materials.

To facilitate data interpretation and analysis, computer graphics capabilities have been developed to plot concentration and meteorological data as time series. Comparison of these time series has suggested some strong correlations between meteorological conditions and concentration variations.

It must be stressed that these findings are preliminary, and will be tested in further and more thorough investigations.

STRATOSPHERIC SOURCE MATERIALS

Observations have shown that the major source region for atmospheric radionuclides and ozone is the stratosphere. If there is no significant low level source, such as a nuclear device detonation, high concentrations at the ground can occur only if there is a vertical atmospheric transport mechanism which moves these materials from high levels through the depth of the troposphere.

The data analyzed so far suggest the following relationships and mechanisms of downward transport:

- a) Maxima in the daily averaged records of the radionuclides tend to occur following a frontal passage. This may be due to, or facilitated by a wide area of sinking air behind the front. Another possible mechanism of downward transport could be local deep convective mixing associated with the front.
- b) Maximum concentrations tend to occur when the station is under the leading edge of an upper level trough. In these situations, the reference station may be downstream of a region of possible upper tropospheric cyclogenesis. These regions have been shown to be sometimes associated with tropospheric intrusions of stratospheric air.³
- c) As would be expected, radionuclide concentrations vary similarly with daily maximum ozone concentrations. However, there are a few cases when distinctly dissimilar behavior can be seen. Further more detailed analyses will help to explain these situations.
- d) There is some indication that scavenging by rain induced by a front may reduce radionuclide concentrations.
- e) Longer term periods (on the order of 5 days) of high concentrations

have been recorded. Often in these situations, surface pressure patterns lacked organization, and pressure gradients at Quillayute were weak. Gradients were weak aloft also, and upper level flow was divergent west of the station. Further data and investigation will help to define the processes involved in these situations.

An example which illustrates several of these conditions is the case

of March 10, 1974. The radionuclides (Figure 1) all showed distinctive maxima that day. The hourly ozone concentrations (not shown), reached a higher maximum on the 10th than on either the preceding or the following day. Strong precipitation (see Figure 2) preceded the front as it replaced an earlier system with the frontal passage, but then dropped off during much of the 10th. This strongly suggests the possibility of scavenging.

FIGURE 1. Quillayute, Washington, Air Concentrations, March 1974.

FIGURE 2. Surface Winds and Hourly Precipitation.

Figure 3 and Figure 4, respectively, show the surface and 500 mb charts at 0400 PST on the 10th. The surface front had just passed the reference station at that time. The position of the 500 mb trough is shown, with Quillayute under its eastern side.

The strong maximum which appears on March 20th in Figure 1 coincided with the arrival of a continental air mass following the north-to-south passage of an Arctic front east of the reference station. There was no precipitation associated with this system at Quillayute. This is an extreme example of the somewhat greater concentrations which are sometimes associated with more northerly storm trajectories.

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FIGURE 3. Surface Weather Map, 0400 PST, March 10, 1974.

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FIGURE 4. 500 mb Height Contours, 0400 PST, March 10, 1974.

EARTH SOURCE MATERIALS

A second group of atmospheric trace materials to be considered includes those whose source region is the earth. In studying the variations in concentrations of these materials, horizontal transport is the prime mechanism under consideration, although vertical movement may play a role. The variations in hourly condensation nuclei concentrations, and in daily lead (Pb) concentrations, are the records examined to date. A coarse examination of the data suggests the following relationships:

- a) Lowest Pb and condensation nuclei concentrations are usually observed in a "new" air mass within two days of a frontal passage.
- b) Significant reductions in condensation nuclei concentrations corresponding to precipitation episodes indicate removal by scavenging.

A good example of low Pb concentrations in a "new" air mass occurred in April 1974 (Figure 5). The daily concentration values on April 10th had become low following a frontal passage on the 9th. A second front, which can be seen on the surface map for 0400 PST on March 12 (Figure 6), passed Quillayute early on the 11th, with strong westerly flow behind it as a surface high developed to the west. This high (see Figure 6) was responsible for the continuing northwest surface flow through the 12th, the day of extremely low Pb concentrations.

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FIGURE 5. Average Daily Lead Values at Quillayute, Washington, April 1974.

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FIGURE 6. Surface Weather Map,
0400 PST, April 12, 1974.

PRESENT AND FUTURE ACTIVITIES

The preceding discussion presents a number of possible relationships which are suggested by the data analyzed so far. These working ideas

will be tested using back trajectory analysis techniques, which are now being adopted for use with NMC gridded temperature, height, and wind data at standard levels between the surface and 10 mb. Presently, constant pressure trajectory analyses are being performed on significant cases.

Future plans include performing of isentropic trajectory analyses. These will allow the addition of vertical motion considerations to the analyses, for the investigation of: a) paths of air movement from regions of suspected stratospheric-tropospheric air interchange; b) vertical motions associated with dynamic atmospheric conditions, which may help to explain anomalies in concentration variations; and c) air movement histories, including scavenging from passage through precipitation regions.

The Quillayute reference station observations present a great opportunity, not only to study the behavior of many atmospheric contaminants and constituents, but to study large scale atmospheric processes as well.

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TRANSURANIUM ELEMENTS IN THE ATMOSPHERE

C. W. Thomas and R. W. Perkins

The chemical and radiochemical composition of atmospheric aerosols is being measured to aid in assessing the present and future hazards to the environment presented by radioactive and nonradioactive contaminants. The rates and mechanisms by which airborne particles are mixed in the atmosphere and transported to and deposited on the earth's surface are also under study. This report describes 1) past introduction of radionuclides to the atmosphere resulting from nuclear weapons testing, the burnup of a navigational satellite, and by natural processes, and 2) the behavior of these radionuclides after release.

On November 1, 1952 the first thermonuclear device was detonated¹ and injected significant amounts of debris into the stratosphere. However, it was not until the U.S. and USSR nuclear weapons testing program of 1961-62 that large amounts of transuranium elements were injected into the atmosphere. In addition to the weapons-produced transuranium elements, major injection of ^{238}Pu into the atmosphere occurred in April of 1964 when a navigational satellite failed to achieve a stable orbit.¹ The 17 kCi of ^{238}Pu which was added to the atmosphere was relatively small compared with the 350 kCi of $^{239,240}\text{Pu}$ which has been added by nuclear weapons testing.²

These injections result in the long-term, relatively slow deposition of radioactivity over the entire surface of the earth. Empirical box models which describe the movement of radioactivity from the upper to the lower stratosphere, between hemispheres, from the stratosphere to the troposphere, and the deposition rate on the earth's surface have been developed and appear to be reasonably satisfactory. As a first approximation, the transuranium elements appear to behave in their atmospheric transport in essentially the same manner as other fission products. It has been shown, for example, that the ratio of ^{90}Sr to $^{239,240}\text{Pu}$ was reasonably constant in fallout from sev-

eral locations on the earth during the years following the U.S. and USSR nuclear tests of 1961 and 1962.³

The concentrations of ^{238}Pu and ^{239}Pu from 1962 to the present show that seasonal variations in the $^{238,239}\text{Pu}$ were similar to those of other nuclear-weapons-produced radionuclides of stratospheric origin. The rate of decrease in the $^{239,240}\text{Pu}$ concentrations from 1963 through 1967 corresponded to a stratospheric half-residence time of 10 to 11 months. Concentrations remained fairly constant from 1967 to 1972, primarily because of thermonuclear tests conducted by the Chinese at Lop Nor (44°N). The $^{238}\text{Pu}/^{239}\text{Pu}$ ratio which averaged about 0.015 in 1964 increased slightly in 1965, and by the spring of 1966 it had increased to 0.042, suggesting that ^{238}Pu from the SNAP-9A burnup was present. The amount of SNAP-9A ^{238}Pu present was determined 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. From 1967 through 1970 the concentrations of SNAP-9A plutonium at Richland remained fairly constant, indicating that the ^{238}Pu was being transferred into the northern hemispheric lower stratosphere at a rate comparable to the rate at which ^{238}Pu was being deposited on the earth's surface. This suggests that a substantial amount of ^{238}Pu was retained in the upper stratosphere and

its slow movement into the lower stratosphere maintained a nearly constant level for 3 to 4 years. The fact that the ^{238}Pu concentrations showed the usual seasonal variations typical of radionuclides of stratospheric origin, indicates that the transfer was taking place primarily from the stratosphere to the troposphere and finally surface air.

Although not formed directly in the nuclear weapons detonation, considerable amounts of ^{241}Am are present in fallout debris. Based on the amount of ^{239}Pu in the atmosphere and an estimated radioactivity ratio of ^{239}Pu to ^{241}Pu of 0.015 (based on the Ivy Mike concentration), one can calculate their approximate ratios as a function of time in the atmosphere. These ratios are plotted in Figure 1 together with our observed concentrations of ^{241}Am and show a very good correlation. Based on the yields of transuranium elements which were observed in the Ivy Mike tests and the $^{239,240}\text{Pu}$ inventory established by HASL, the total amounts of the other transuranium elements which have entered the atmosphere can be estimated. These values are shown in Table 1. For isotopes of mass greater than 244, the total atmospheric injections are in the range of hundredths to tens of curies and the total alpha decay ac-

tivity of all the transuranium elements of mass greater than 241 is only about one percent of the $^{239,240}\text{Pu}$.

FIGURE 1. Estimated Concentration of ^{241}Pu and ^{241}Am as Compared to the Measured Concentration of ^{241}Am in Surface Air Since 1962 at Richland, Washington.

TABLE 1. Relative Abundance of Transuranium Elements
Based on Ivy Mike and Estimated Atmospheric Injection.

<u>Isotope</u>	<u>Activity Abundance</u>	<u>Total Injection, kCi¹</u>
²³⁹ Pu	1	150
²⁴⁰ Pu	1.35	203
²⁴¹ Pu	63	9450
²⁴¹ Am	--	327 ²
²⁴² Pu	1.2×10^{-3}	.18
²⁴³ Am	6.9×10^{-3}	1.04
²⁴⁴ Pu	3.5×10^{-7}	5.1×10^{-5}
²⁴⁵ Cm	3.6×10^{-4}	5.4×10^{-2}
²⁴⁶ Cm	2.4×10^{-4}	3.6×10^{-2}
²⁴⁷ Cm	6.2×10^{-9}	8.6×10^{-7}
²⁴⁸ Cm	8.4×10^{-8}	1.3×10^{-5}
²⁴⁹ Bk	3.2×10^{-3}	.48
²⁵⁰ Cm	6.5×10^{-8}	8.6×10^{-6}
²⁵¹ Cf	3.8×10^{-7}	5.6×10^{-5}
²⁵² Cf	9.3×10^{-6}	1.4×10^{-3}
²⁵³ Cf	2.5×10^{-4}	3.8×10^{-2}
²⁵⁴ Cf	7.4×10^{-6}	1.1×10^{-3}
²⁵⁵ Es	9.1×10^{-6}	1.4×10^{-3}

¹Assumes kCi ^{239,240}Pu atmospheric injection.

²Americium-241 formed on total decay of ²⁴¹Pu.

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ATMOSPHERIC FALLOUT DURING 1974 AT RICHLAND, WASHINGTON
AND POINT BARROW, ALASKA

C. W. Thomas

Concentrations of ^{238}Pu , ^{239}Pu , ^{240}Pu , and ^{241}Am have been measured in air samples taken at Richland, Washington, and Point Barrow, Alaska, since 1962, and the concentration of several other transuranium radionuclides estimated. During 1974, the concentrations of nuclear-weapons-produced radionuclides were 3 to 10 times higher than those of 1973, as a result of the June, 1973, Chinese nuclear test.

The concentration of radionuclides associated with nuclear weapons testing and natural processes were measured in ground-level air samples collected at Richland, Washington and Point Barrow, Alaska during 1974. In June 1973 the Chinese tested a nuclear device of a few megatons, inserting relatively large amounts of radioactive debris into the stratosphere. Data from previous years had shown that debris inserted into the stratosphere during one year would be mainly observed in surface air the following year. This was well demonstrated in measurements made in 1974 where debris from the 1973 nuclear test increased the surface air concentration by 3 to 100 times over those observed in 1973.

Figure 1 shows the ^{144}Ce concentration in surface air measured at Richland, Washington since 1962. The large scale testing by the USSR and USA in 1962 was responsible for the ^{144}Ce concentration during the period 1963 through 1967. During this period the stratospheric concentration decreased with a residence half-time of about 11 months,¹ typical of material of stratospheric origin. In 1967 the Chinese detonated their first high-yield device and in 1968 the debris from this device was responsible for the relatively large increase measured during that year. With single high-yield nuclear tests occurring in 1968, 1969, and 1970 the surface air concentration of ^{144}Ce was governed by the yearly stratospheric insertion from these devices

combined with a radioactive decay and a stratospheric residence half-time of 11 months, the combination of which resulted in a near constant surface air concentration from 1968 through 1971.

During the period 1971 through 1973 the atmospheric concentration of ^{144}Ce decreased in a manner similar to that of 1963-1967, reflecting the lack of high-yield testing during 1971-1972. In 1974 the relatively large increase of ^{144}Ce surface air concentration over the 1973 values was similar to that in 1968 which followed a relatively long period without testing. In June of 1974 the Chinese tested a device of near 1 megaton inserting most of the radioactive debris into the stratosphere. This debris will be apparent in the spring maximum of 1975 with probable surface air concentrations similar to those measured during 1969.

Yttrium-88 was not detected in early air samples that contained fresh debris from the June 1974 Chinese test but was easily detected in samples taken after July 1974. This, coupled with the large amounts of ^{88}Y measured in ocean water samples taken near the Panama Canal would indicate the source of this isotope was from French testing in the southern hemisphere.

The concentrations of short-lived debris in the troposphere from the 1974 test (see Table 1) measured at

FIGURE 1. Radionuclide Concentration in Surface Air at Richland, Washington.

TABLE 1. Atmospheric Concentrations of Fallout Debris from the Chinese Nuclear Test of June 17, 1974 in Surface Air at Richland, Washington.¹

Date	Air Volume	dpm/10 ³ SCM										
	10 ³ m ³	¹⁴⁷ Nd	⁹⁹ Mo	¹⁴¹ Ce	²³⁷ U	²³⁹ Np	¹⁴³ Ce	¹³¹ I	⁹⁷ Zr	¹³² I	⁹⁵ Zr ²	¹⁴⁰ Ba
6/24-25	14.0	3.8±0.5	31±0.3	4.5±0.2	33±2	147±7	--	4.6±0.5	--	4.9±1.3	--	3.4±0.3
6/29-30	17.3	13±2	--	10.5±0.2	177±2.5	752.±69	--	9.8±0.5	--	3939±5	26±0.6	16.5±0.5

¹Decay corrected to June 17, 1974.

²Combined fallout from June 23, 1973 and June 17, 1974 Chinese tests.

Richland, Washington were about one-fourth of those measured at our laboratory from the June 1973 Chinese nuclear test and were approximately two orders of magnitude lower than airborne concentrations observed from the March 1972 test. Radioactivity levels on grass samples collected at approximately 40-mile intervals between Los Angeles, California, and the Canadian border were surveyed.

The results indicated concentrations below detection levels which gave ^{131}I radiation exposure values for the grass-milkman pathway for the U.S. west coast of <0.042 mrem, based on a 2-g child thyroid. These ^{131}I thyroid exposures were two to four orders of magnitude lower for U.S. west coast residents than those from the March 1972 low yield Chinese nuclear test.

RADIONUCLIDES FROM NUCLEAR POWER REACTORS

C. W. Thomas

Because of the anticipated use of nuclear power reactors as a power source, the relative concentrations of transuranium radionuclides expected to be present in radioactive wastes from these reactors were calculated. This will aid in predicting the environmental hazards posed by accidental release of radioactive wastes.

Over the next 30 years the nuclear power industry is expected to accumulate approximately a quarter of a million tons of radioactive wastes. To aid in evaluating the impact from unintentional and inadvertent releases to the environment, it is essential to identify those radionuclides that will constitute a major potential environmental hazard.

In the past most of the radionuclides entering the environment have been generated in connection with nuclear detonations or with production of weapons grade plutonium. The radionuclide spectrum in wastes produced by the nuclear industries will differ significantly from those accumulated during prior production and nuclear detonation. In nuclear waste from weapons grade plutonium production, the long-lived isotope ^{239}Pu was a major alpha emitter; however in nuclear power generation, ^{238}Pu together with isotopes of americium and curium are the important alpha emitters.

To provide perspective as to the relative amounts of the radionuclides in nuclear power industry wastes their concentrations have been compared to either ^{137}Cs or ^{239}Pu as shown in Tables 1 and 2. The relative concentrations of the fission products are somewhat similar to what we have been concerned with from fallout and ^{239}Pu production; however, the longer exposure does favor the long-lived products and also the second-order reaction products. The very long decay of fission product mixture will result in the relatively rapid loss of the more common short-lived fission products and will leave

a fission product spectrum containing ^{93}Zr , $^{93\text{m}}\text{Nb}$, ^{151}Sm , ^{99}Tc , ^{79}Se , ^{126}Sn , ^{135}Cs and ^{129}I . The activation products will vary widely dependent on changes in construction material. However, it is believed that the activation radionuclides of greatest significance will be ^{55}Fe , ^{54}Mn , ^3H , ^{63}Ni , ^{60}Co and ^{14}C .

The most striking difference in radionuclide ratios in wastes from nuclear energy generation will be in the heavy element radionuclides. It is evident that with the high fuel exposure for the nuclear industry, the Am and Cm radionuclides and ^{238}Pu will be of far greater significance in nuclear wastes, both in abundance and in radiological hazard, than is ^{239}Pu .

The ratios of the transplutonium elements to ^{239}Pu in nuclear fuels will, of course, be less than those in nuclear wastes. However, as burnup and recycling continues, the Cm isotopes will approach the values in the waste. In Table 3 the relative concentrations of the transuranium elements in 3.3% enriched ^{235}U fuel and first generation recycled 3.5% enriched ^{239}Pu fuel is shown both before and after exposure. It is apparent that as this fuel is recycled, the relative concentrations of the higher atomic number plutonium isotopes compared to ^{239}Pu increase dramatically. After several recycles of the fuel, the higher isotopes of plutonium, as well as ^{238}Pu , become responsible for most of the actual weight of the plutonium and their relative abundance approaches that in the waste material.

TABLE 1. Activity of Fission Products in Accumulated Waste for Entire Nuclear Industry.

	NORMALIZED TO 1 FOR ^{137}Cs								
	6.2×10^6	4.3×10^7	1.8×10^8	4.5×10^8	8.6×10^8	1.5×10^9	3.4×10^9	6.2×10^9	1.9×10^{10}
	1972	1974	1976	1978	1980	1982	1986	1990	2000
^3H	0.0064	0.0063	0.0061	0.0060	0.0060	0.0061	0.0059	0.0056	0.0049
^{85}Kr	0.10	0.099	0.096	0.095	0.093	0.091	0.084	0.087	0.091
^{89}Sr	0.19	0.12	0.088	0.067	0.052	0.046	0.030	0.025	0.024
^{90}Sr	0.72	0.72	0.70	0.71	0.71	0.70	0.67	0.69	0.68
^{91}Y	0.73	0.23	0.16	0.13	0.097	0.086	0.056	0.047	0.044
^{95}Zr	0.66	0.44	0.31	0.24	0.19	0.17	0.11	0.091	0.094
^{95}Nb	1.4	0.88	0.63	0.48	0.38	0.34	0.23	0.18	0.18
^{103}Ru	0.13	0.085	0.061	0.046	0.037	0.034	0.022	0.019	0.031
^{106}Ru	2.8	2.2	1.8	1.6	1.3	1.3	0.97	0.72	0.66
$^{110\text{m}}\text{Ag}$	0.015	0.011	0.0087	0.0071	0.0060	0.0059	0.0045	0.0030	0.0011
^{125}Sb	0.067	0.060	0.055	0.051	0.047	0.046	0.040	0.034	0.035
$^{127\text{m}}\text{Te}$	0.023	0.015	0.011	0.0087	0.0069	0.0064	0.0044	0.0036	0.0038
$^{129\text{m}}\text{Te}$	0.035	0.0023	0.0016	0.0012	0.00096	0.00089	0.00059	0.00048	0.00061
^{134}Cs	1.7	1.5	1.3	1.2	1.1	1.0	0.79	0.69	0.39
^{137}Cs	1	1	1	1	1	1	1	1	1
^{141}Ce	0.069	0.045	0.032	0.024	0.010	0.017	0.011	0.0095	0.013
^{144}Ce	4.9	3.7	3.0	2.4	2.0	1.8	1.3	1.1	0.73
^{147}Pm	0.82	0.74	0.66	0.61	0.56	0.53	0.44	0.38	0.42
$^{148\text{m}}\text{Pm}$	0.0051	0.0033	0.0024	0.0018	0.0015	0.0014	0.00092	0.00070	0.00011
^{151}Sm	0.012	0.012	0.012	0.012	0.012	0.012	0.013	0.013	0.016
^{154}Eu	0.064	0.063	0.061	0.061	0.061	0.062	0.061	0.059	0.043
^{155}Eu	0.051	0.044	0.038	0.034	0.030	0.029	0.024	0.021	0.040
TOTAL CURIES	1.5×10^8	8.5×10^8	3.0×10^9	6.5×10^9	1.1×10^{10}	1.8×10^{10}	3.3×10^{10}	5.3×10^{10}	1.4×10^{11}

TABLE 2. Activity of Heavy Elements in Accumulated Waste for Entire Nuclear Industry.

Isotope	Normalized to 1 for ^{239}Pu								
	1972	1974	1976	1978	1980	1982	1986	1990	2000
^{237}Np	0.22	0.21	0.22	0.21	0.20	0.18	0.15	0.14	0.076
^{239}Np	11.5	11.2	11	10.8	14.5	26.5	43.1	31.7	7.84
^{236}Pu	0.00092	0.00076	0.00075	0.00067	0.00056	0.00061	0.0036	0.010	0.012
^{238}Pu	36.1	44.1	49	50	55.6	80.6	167	283	361
^{239}Pu	1	1	1	1	1	1	1	1	1
^{240}Pu	1.54	1.65	1.7	0.58	1.89	2.55	4.45	5.33	3.14
^{241}Pu	315	306	300	287	295	337	403.3	367	255
^{242}Pu	0.0043	0.0043	0.0044	0.0042	0.0047	0.0065	0.0099	0.011	0.011
^{243}Pu	--	--	--	--	--	--	--	--	--
^{241}Am	96.1	95.3	96.5	93.6	103	137	178	141	77.2
$^{242\text{m}}\text{Am}$	5.69	5.64	5.5	5.4	7.23	12.2	19.5	14.2	5.68
^{242}Am	5.69	5.64	5.5	5.4	7.23	12.2	19.5	14.2	5.68
^{243}Am	11.5	11.2	11	10.8	14.5	26.5	43.1	31.7	7.84
^{242}Cm	5637	3887	3020	2255	2624	3876	3183	953	278
^{243}Cm	2.31	2.23	2.25	2.16	2.34	3.16	4.03	3.00	2.16
^{244}Cm	1561	1511	1510	1427	2441	5671	9813	6464	919
^{245}Cm	0.23	0.22	0.23	0.22	0.50	1.43	2.64	1.92	0.33
^{246}Cm	0.046	0.045	0.046	0.046	0.072	0.26	0.49	0.35	0.063
^{247}Cm	--	--	--	--	--	--	--	--	--
TOTAL CURIES	7.2×10^5	3.9×10^6	1.4×10^7	3.0×10^7	7.9×10^7	2.7×10^8	9.3×10^8	1.2×10^9	1.8×10^9

TABLE 3. Heavy Element Activities in Nuclear Fuels.¹Normalized to 1 for ^{239}Pu

Isotope	3.3% Enriched ^{235}U		3.5% Enriched ^{239}Pu	
	Charge ²	Discharge	Charge	Discharge
^{234}U	23.2	.0023	.00022	.00038
^{235}U	1	.000052	.00001	.00001
^{237}Np		.001	--	.00014
^{238}U	4.55	.00086	.00022	.00047
^{238}Pu		8.62	8.84	21.0
^{239}Pu	--	1	1	1
^{240}Pu		1.49	1.46	2.45
^{241}Pu		324	291	662
^{242}Pu		.0043	.0042	.016
^{241}Am		.264	.762	1.26
$^{242\text{m}}\text{Am}$		.0127	--	.072
^{243}Am		.055	--	.276
^{242}Cm		114	--	668
^{243}Cm		.017	--	.129
^{244}Cm		8.05	--	73.1
^{245}Cm		.00087	--	.015
^{246}Cm		.00019	--	.003

¹PWR, Power = 30 MWT Burnup = 33,000 MWDT.²Normalized to 1 for ^{235}U .

COLLECTION EFFICIENCY CALCULATIONS
FOR AN ASPIRATING, AXISYMMETRIC PROBE
FOR MEASURING ATMOSPHERIC AEROSOLS

Ronald L. Drake

The proper design of inlet probes and ductwork is essential to the validity of data obtained in airborne measurements in the Aerosols and Trace Gases program. Because of its importance a small, but continuing research effort is being conducted to improve design of this equipment. This report describes one phase of the work that has been completed during the past year.

INTRODUCTION

Collection probes for atmospheric aerosols usually consist of four major components, namely: the platform on which they are mounted, an external entrance port, internal plumbing, and a final collection device or measuring device. The platform may be a balloon, a rocket, a glider, or an airplane traveling through the air with a speed equal to U . The shape, location and type of the external port of the collector varies greatly, and the variations depend on whether the sample is taken isokinetically or anisokinetically, and on whether the flow through the port is due to ram pressure or due to a suction created by a pump. The final collection or measuring unit may be a filter, a bag sampler, or some type of electronic or optical measuring device.

An "undisturbed" population of particles undergoes many modifications from the time it first experiences the effects of the flying platform to the time it is finally collected or measured by the probe. And if the collected samples are removed from the probe to be further analyzed in a laboratory, the population is modified further. Since the undisturbed air is forced to flow around the flying platform, the particles embedded in this air are also disturbed. Gravitational effects and aerodynamic drag cause the larger particles to follow trajectories different than those of the fluid particles. How-

ever, the smaller particles will tend to follow the general motion of the air, except possibly in the immediate neighborhood of the entrance port.

As the particles enter the sampling port, the changes in direction, mean shear characteristics, and turbulent structure cause some of the particles to be deposited on the walls of the port. As the particles and air move on through the internal plumbing, some particles will be deposited and some will be reentrained depending upon the particle size, turbulent character of the flow, number and type of pipe bends, and the number and type of expansions and contractions in the flow lines. Finally, as the particles are collected or enter the measuring chamber, there will be still other modifications of the population. Usually, these modifications are size dependent. Hence, if we want to correlate the measured aerosol spectrum with the particle population in the undisturbed atmosphere, we must analyze the entire time-history of a set of particles from the undisturbed atmosphere to their final destination.

In this report we will only consider the external collection efficiency of the inlet port. In addition, we will assume that the probe is so located that the flying platform only influences the collection procedure by moving the probe through the air with speed U . Thus, the

analysis consists of the determination of the flow field around the probe, the trajectories of the particles embedded in that flow field, and the calculation of the collection efficiency.

FLOW FIELD

The flow field around the probe is assumed to be a steady, inviscid, incompressible and irrotational field; that is, it is a perfect flow field. The aspirating probe is "constructed" from a solid axially symmetric body by "drilling" a sampling hole of radius r_0 along the axis of symmetry (see Figure 1). In these figures the dimension "a" is a pseudo-semimajor axis and the dimension "b" is pseudo-semiminor axis. Point P is a point with axial coordinate x and radial coordinate r. The velocity field at P is given by (u,v) , where u is the axial component and v is the radial component. When we are sufficiently far upstream, the velocity field is uniform and is denoted by $(U,0)$. That is, our coordinate system is fastened to the body and the air moves past it at a speed equal to the flying platform. Flow fields of this type can be represented by a Stoke's stream function.

The highlights of our flow field computations are as follows:

1. The system is nondimensionalized using U as a characteristic velocity and "a" as a characteristic length.
2. The Stoke's stream function satisfies an elliptic, partial differential equation which must be solved numerically, at least in the immediate neighborhood of the inlet hole.
3. To reduce the amount of numerical computation, χ is determined for the solid body, Figure 1a, by analytic or psuedo-analytic means.
4. From the results in Item 3 and from the specifications of the inlet hole, we can write down the boundary conditions for χ for the aspirating probe in Figure 1b.
5. For numerical computations it is convenient to have a rectangular

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FIGURE 1. A Schematic of an Axially Symmetric, Aspirating, Aerosol Probe.

domain of computation. Thus, we search for a transformation

$$\begin{aligned}\xi &= \xi(x,r), \\ \eta &= \eta(x,r),\end{aligned}\quad (1)$$

that will map the domain around the physical probe onto a rectangle in the (ξ, η) -plane. We can use the techniques of conformal mapping to aid in the selection of Equation (1). An example of this mapping is given in Figures 2 and 3, where $\xi \equiv$ constants are ellipses in the (x,r) -plane and $\eta \equiv$ constants are hyperbolas. The $\eta = \eta_0$ hyperbola intersects the sampling hole and $\xi = \xi_2$ is the elliptical body.

6. The governing equation for the stream function $\phi(\xi, \eta)$ in the (ξ, η) -plane is obtained by applying Equation (1) to the governing equation for χ . This new equation is then solved numerically in the (ξ, η) -plane and the results are transformed back to the (x,r) -plane.

The procedure outlined above is valid for rather arbitrary probes (as

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FIGURE 2. Elliptic Coordinate Grid Around the Probe.

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FIGURE 3. The Transformed Flow Field.

long as they are axially symmetric) and for isokinetic or anisokinetic probes. In addition, the flow through the sampling hole may be forced by ram pressure or by a suction created by a pump.

PARTICLE TRAJECTORIES

For the trajectory calculations, we use a rectangular coordinate system with components (x, y, z) . In Figure 1b, the r -axis is resolved into a vertical z -axis (positive upward) and a horizontal y -axis. The outline of the computational procedure is given below.

1. The flow field around the probe is given from the previous section and we assume that the particles do not affect this field.
2. The particles are assumed to be perfect spheres; however, we can relax this restriction.
3. As the particle moves through the flow field, it experiences the effects of gravity and aerodynamic drag. The empirical drag coefficients which are used in our work are found in Schlichting (1955) and McDonald (1960). Some improvements in these coefficients are given in Beard and Pruppacher (1969).
4. Our computations start with a particle at a location (x_0, y_0, z) such that its velocity is given by $(u, 0, -v_s)$, where v_s is the terminal velocity of the particle.
5. The equations of motion for the particle are integrated numerically from (x_0, y_0, z) to the inlet hole $(-a, y, z)$, where $y^2 + z^2 \leq r_0^2$.
6. The technique of integration is an Adams-Moulton variable-step procedure with time steps varying from 10^{-8} seconds to 10^{-2} seconds. The smallest time steps are used in the neighborhood of the sampling hole.

COLLECTION EFFICIENCIES

The purpose of this work is to calculate collection efficiencies for an aerosol probe. That is, we want to know how large a region of a cloud of particles is being sampled by the probe as the platform moves along. The collection efficiency (C.E.) of the probe is defined as the ratio of area A , to area A_0 . The area A_0 is the area of the sampling hole $A_0 = \pi r_0^2$, and A , is the area in some vertical plane in front of the probe through which pass only those particles that eventually enter the probe. A particle not passing through A , will not enter the probe. This vertical plane must satisfy two requirements:

1. The vertical plane must be sufficiently far in front of the probe so that the probe does not influence the flow field; $x = -6a$

is a good candidate for the vertical plane.

- The area A is dependent upon the particle radius; as the radius r_p decreases, A , approaches a value $(A_1)_* = \pi r_*^2$; and as r_p increases, A tends toward A_0 . The value of A_* , or r_* , is determined from the fact that the quantity of flow between streamlines is proportional to the difference of the values of χ . Hence, $r_* = r_0[S/2u]^{1/2}$, where the mean velocity through the sampling hole is $s/2$ (see Figure 4). In Figure 4, the velocity

profile in the hole is assumed to be parabolic.

The area A , in the $x = -6a$ plane is obtained by employing a search technique along the lines $y = 0$, $z = z_0 \pm y$. If the center of the particle eventually enters the sampling hole, it is declared a "hit," otherwise it is a "miss." A sufficient number of initial positions $(-6a, y_0, z_0)$ are taken along each line to ensure an accurate value for A .

A more detailed discussion of this work will be in the forthcoming BNW report.

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FIGURE 4. A Schematic Showing the Maximum Collection Efficiency of the Probe.

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TRANSPORT, DIFFUSION AND TURBULENCE

Research summarized in the following reports considers and models the physical behavior of the atmosphere and its contaminants. These reports reflect the widening range of scales in Battelle's transport, diffusion and turbulence research.

Work has begun on a regional transport, dispersion and removal model embracing a horizontal area of 1400 x 1400 km, and extending vertically beyond the planetary boundary layer through the depth of the troposphere. A regional study over a more modest 400 x 500 km Columbia Basin area generates graphical display techniques which could significantly aid analysis of accidental or routine effluent releases. Improvements in a model to compute low-level Montgomery stream functions are reported; the stream function data can be used in trajectory calculations in the lowest kilometer of the atmosphere.

As emphasis in transport and diffusion studies moves from micro to meso and regional scales, emphasis in field sampling also moves from fixed to mobile sampling platforms. Contributions in this section deal with an airborne sampling technique and some of the problems associated with mobile sampling.

At the microscale end of the spectrum, the concentrations of a pollutant released in low wind speed situations are examined. Mean concentrations and movement of diffusing plumes are modeled and the frequency distribution of short-period concentrations is examined.

Finally, turbulent wind data are analyzed as a potential input to microscale diffusion modeling. The extension of constant-flux-layer models to a height of 60 m is discussed, and Hanford data are compared with models based on data from other locations.

- **ATMOSPHERIC DIFFUSION, DEPOSITION AND TRANSPORT PHENOMENA**
- **FUNDAMENTAL TURBULENCE**
- **FALLOUT PHENOMENOLOGY**
- **REGIONAL TRANSPORT OF EFFLUENT FROM LMFBR RELATED FACILITIES**
- **HANFORD REGIONAL METEOROLOGICAL STUDIES — AEC/RL**

REGIONAL TRANSPORT, DISPERSION AND REMOVAL
OF ATMOSPHERIC POLLUTANTS

L. L. Wendell and D. C. Powell

Work has begun on a regional scale transport and dispersion model to assess the environmental impact of current and future levels of pollution from industrial and power generating facilities. Preliminary work involved examining the effects of time and space scales for a 1400 x 1400 km area on transport and diffusion considerations used in micro and meso scale work (less than 100 km). The vertical diffusion process with regard to the mixing depth concept is critically reviewed. Removal processes such as wet and dry deposition as well as chemical and radiological transformations are also considered for incorporation into the model.

INTRODUCTION

During the past several months the AEC (soon to formally become ERDA) has been encouraging a substantial effort in the investigation of regional pollution problems. The term "regional" refers to a space scale of 100 to 1000 km (multi-state). The concern here is that, while great effort is expended in urban and industrial pollution problems for a local area, little concern has been shown for the problems caused several hundred kilometers away. For example, stacks may be tall enough that pollution standards are met close to an industrial or power generating plant, but the effluent may be causing an acid rain in a distant location. Because the problem is most acute in the northeastern United States, the modeling effort has begun for this area of the country. We are trying to keep one eye on state-of-the-art developments in all areas involved in this problem and the other on the growing need for this type of assess-

ment as soon as possible. Our effort, begun only recently, is to assemble an assessment model to include the elements of transport, dispersion and removal processes in as realistic a manner as is practicable. The model will permit evaluation of the regional environmental impact of present and increased levels of industrialization and power production.

Because the time periods involved in an assessment model of this type are long, i.e., seasonal to annual, certain economic measures will have to be considered in the initial stage of the work. One such measure is the use of historical wind data for transport calculations. Since we are not concerned here with a prediction of the flow field from some initial condition, we can forego the expense of generating a time dependent solution to a system of hydrodynamic equations. However, results of time dependent flow models should provide insight for development of objective schemes in space data regions of complex topography or meteorological conditions.

The diffusion portion of the initial model is patterned after a puff-approximated plume approach used previously in mesoscale work.¹ The initial selection of this scheme, as opposed to a particle-in-cell technique, is based mainly on economy for long term calculations.

The primary objective during the first phase of this work is to assemble a working model and make some attempts at validation. This will involve collecting whatever emission and sampling data are available from the Environmental Protection Agency and selecting some cases suitable for model validation runs.

EMISSION INVENTORIES

Regardless of the sophistication and validity of a model for transport diffusion and removal of atmospheric pollutants, a correct specification of the source term is essential to a realistic result. The source term to be used in the model will be determined by compilation of data, collected and presented by others, in such areas as plutonium and transplutonium products from LMFBR related facilities and pollutants from conventional power production facilities and large urban industrial complexes. Since only annual emission inventories are required by the EPA, a time history of finer resolution will have to be estimated using any available related factors. The composition of the urban plumes will be checked with data from aircraft samples obtained in an associated program.

There is a great volume of air sampling data available from the National Air Sampling Network of the EPA. Work is now progressing to determine if the data is suitable for model validation purposes and comparison with the emission inventories.

TRANSPORT

Boundary Layer Flow

Atmospheric pollutants will be mainly confined to a layer less than one kilometer deep during very stable periods, when relatively minor topographic variations can cause complex variations in the flow.² During these periods transport in the boundary layer will have to be determined from

surface wind measurements over a region, and from a very few low level wind determinations from pilot balloons and flights. The hourly surface wind measurements obtainable from the National Weather service of NOAA are visual quick look averages but should be representative under steady wind conditions of high stability. One of our main objectives in this phase of the work is to determine how much influence mesoscale meander in the boundary layer has on regional scale transport.

Tropospheric Flow

The major new consideration in regional scale transport, as opposed to mesoscale transport, is that the longer travel distances may allow the pollutants to be mixed or carried through the full depth of the troposphere. This brings us to the question of which is the most realistic manner to treat the transport of pollutants. There are three basic alternatives.

The first alternative is to consider the flow to be horizontally layered, with material in each layer being carried at an average velocity which applies through the thickness of the layer. Transfer of material from one layer to the next is carried out by diffusion only.

The second alternative is to consider the velocity to have a vertical as well as a horizontal component. The major deviation from the first alternative is that the flow throughout all the layers must satisfy the condition of continuity. This means material may be transferred between layers by a vertical velocity component as well as by diffusion.

The third alternative is to consider the flow to be adiabatic and confined to isentropic surfaces. Danielsen, 1961,³ has demonstrated that serious discrepancies can occur between trajectories calculated from isobaric and isentropic considerations.

In the initial version of this model the first alternative will be implemented. But, possible refinements from both the second and third alternatives are being investigated.

Flow Through Frontal Storms

Another major consideration in regional scale transport is the frontal storm. In a mesoscale area the effects of a frontal storm generally last only a few hours. However, over a regional scale the influence of a frontal storm can be felt for one to several days. Therefore, the complex flow and the associated precipitation in these storms can make them a major factor in determining the fate of the pollutants over the region. Initial work in this area is to determine the frequency of occurrence, severity and duration of these storms over the region.

DISPERSION

Hypotheses Regarding the Horizontal Dispersion

Approximation of the horizontal dispersion of a continuous plume from a point source with a series of discrete overlapping puffs involves two major considerations. First, the diffusion of the puffs about their respective centers must be accomplished using parameterized σ values derived over time periods smaller than the averaging time for the winds. Second, the variation of the average wind will spread the puffs relative to each other. The combination of these two factors will approximate the total horizontal dispersive effect of the atmosphere.

When the Pasquill type σ_y curves are extrapolated to regional scale, the σ_y values corresponding to travel distances on the order of a 1000 km are small compared to the spatial resolution by which the winds can be defined. Therefore the horizontal shear effect by the defined winds on the individual puffs is neglected. This hypothesis is to be thoroughly tested before this concept will be completely accepted. The σ_y values presently used in the model are those suggested by Briggs.⁶

Hypotheses Regarding the Vertical Spread

The vertical extent of pollutants released into the atmosphere from a surface based structure has been described as a function of two independent physical processes: a) growth caused by turbulent diffusion and

b) inhibition caused by the presence of a more stable layer aloft.⁴ The lower layer is called a mixed layer with height z_i when the change of lapse rate is such that a temperature inversion aloft exists with base at height z_i .⁵ In this case z_i may be used as a dispersion modeling parameter limiting the assumed upward penetration of pollutant released from an effective stack height within the mixed layer. The depth of the layer normally increases during the daytime and decreases at night.

Such a mixing depth is a useful parameter for synoptic dispersion modeling if each of the following is true:

- 1) The effective stack height of the source is in the lower less-stable layer.
- 2) The bottom of the inversion aloft is well defined.
- 3) This definition and its change with time are known over the area of interest.
- 4) The pollutant concentrations above the lid can be neglected.

Obviously the last three criteria are less applicable to the regional scale than they are to the scale of an urban pollution model. Consider the case of material released into an unstable mixed layer capped by a stable inversion aloft during the daylight morning hours. The material is assumed to be confined to the unstable layer. As the afternoon progresses the mixing height increases, and the material is still confined to the mixed layer. However, the following morning when the mixing layer is once again shallow, the material is both above and below the lid. Therefore the base of an inversion aloft restrains the vertical penetration of the material only during the first day of travel. For this reason the concept of a diurnally changing mixing depth does not appear to be useful for modeling the dispersion of pollutant material beyond the first day of travel.

On the other hand, the much lower diffusivity in the stratosphere compared to the troposphere strongly suggests that any realistic model of

the vertical dispersion include an asymptotic limit not greater than the tropopause height. Since we have found no observations of vertical dispersion on a regional scale, any mathematical representation of the process is necessarily of a very tentative nature. Even though the assumption of Gaussian distribution in the vertical is not tenable, the assumption is made initially to facilitate ground level concentration estimation by means of the familiar Gaussian models. The expressions chosen for σ_z follow the scheme of Briggs,⁶ utilizing one formula for each of the Pasquill stability categories. In fact the formulae are altered from the form given by Briggs only so as to build an asymptotic limit for σ_z into each formula. Since this property is found in the original formulations by Briggs for the E and F stabilities, the alterations are confined to the other four stabilities.

The six suggested expressions for σ_z are given as functions of travel distance x and apply to x and σ_z expressed in meters. They are given in Table 1.

TABLE 1. Suggested Expressions for σ_z as Functions of Distance (x) in Meters.

Stability	σ_z
A	$0.20x/(1 + 2 \cdot 10^{-5}x)$
B	$0.12x/(1 + 2 \cdot 10^{-5}x)$
C	$0.08x/(1 + 2 \cdot 10^{-4}x + 4 \cdot 10^{-10}x^2)^{1/2}$
D	$0.06x/(1 + 1.5 \cdot 10^{-3}x + 4 \cdot 10^{-10}x^2)^{1/2}$
E	$0.03x/(1 + 3 \cdot 10^{-4}x)$
F	$0.02x/(1 + 3 \cdot 10^{-4}x)$

REMOVAL PROCESSES

Dry Deposition

In the initial model this phenomenon will be incorporated simply as source depletion as a function of distance. The possibility of incorporating Horst's⁷ surface depletion mechanism in the puff model is under investigation.

Wet Deposition

Precipitation scavenging of atmospheric contaminants, an extremely complex process, is the subject of intensive investigation at this laboratory. The results of these investigations should show how the process may be approximated in the initial assessment model. The preliminary effort to incorporate scavenging into the model will be to approximate the time history of cloudiness and precipitation over the region of concern. This should provide a more realistic appraisal of the effect of the spatial variability of this removal process.

Transformation

The radiological and chemical transformation processes occurring in the contaminants as they move away from the source is another very complex and very important part of the overall problem. The incorporation of these processes will have to be done through parameterization in the initial stages of this work.

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A MODEL TO COMPUTE LOWER LEVEL
MONTGOMERY STREAM FUNCTIONS - PART II

W. E. Davis

Changes are reported in a model to compute low level Montgomery stream functions. An error analysis was performed on the improved model. The errors in stream function were compared to the change in potential temperature from the potential temperature of the surface. This comparison revealed that the improved model produced errors in stream functions within $\pm 0.05 \times 10^7 \text{ cm}^2 \text{ sec}^{-2}$, for $\sim 5^\circ - 6^\circ \text{ A}$ greater than the surface potential temperature at 0700 PST, and for $\sim 3^\circ - 5^\circ \text{ A}$ at 1300 PST.

INTRODUCTION

Past work with the Davis model for computing ψ , the Montgomery stream function,¹ was limited because of absolute errors greater than $0.05 \times 10^7 \text{ cm}^2 \text{ sec}^{-2}$ in the model's calculations. The present work is directed toward improvements in the model which will reduce the total error. With the reduction of the error, as well as delineation of data with high error potential, the model results may be filtered. This filtered stream function data may then be used as input to trajectory calculations.

The 1973 results indicated that errors of $\pm 0.05 \times 10^7 \text{ cm}^2 \text{ sec}^{-2}$ in the model's computation of ψ could be expected at a pressure 100 mb less than the surface pressure. However, one of the problems with the model was the nonallowance of superadiabatic lapse rates. In order to incorporate these lapse rates, the model was changed.

Another problem existed in computing the change, due to the effects of water vapor from the surface, in the height of the first potential temperature surface above the surface potential temperature. Again the model was altered to include the reported surface dew point into the calculations.

MODEL

The model has been previously reported.¹ Basically, inputs from four radiosonde sites are utilized with bilinear interpolation of the radiosonde data of the hourly reporting stations within a grid. The radiosonde data together with surface reports are then used to calculate ψ values. One of the options in the model is the use of surface data at Stampede Pass, Washington (SMP) to adjust the model and recalculate values of ψ .

One of the improvements in this model was to include a provision for calculating a superadiabatic layer near the ground. This was accomplished by putting in the temperature, pressure, and mixing ratio of the minimum potential temperature from one to four of the radiosonde sites used in the model. A delta potential temperature, a delta mixing ratio, as well as delta pressure were computed. These values were used as input for a bilinear interpolation over the grid of hourly reporting stations. A linear time interpolation was made with suppression of the superadiabatic calculation until 1000 PST.

A second improvement was using the observed dew point at the hourly reporting stations. This altered the previously calculated stream function

on the first potential temperature surface. With these two improvements, the model was rerun using August and November 1972 data. A comparison was made with soundings taken at Seattle, Washington (SEA); Portland, Oregon (PDX); and Hanford, Washington (HMS).

RESULTS

The results of the calculations are shown in Table 1. The 1974 results can be compared with the 1973 results by comparing Table 1 with Table 2. A different procedure was

TABLE 1. Errors in ψ ($\times 10^{-7} \text{ cm}^{-2} \text{ sec}^2$).

$\Delta\theta$	November				August		November				August	
	A 0700 PST						A 0700 SMP added					
	PDX		SEA		SEA		PDX		SEA		SEA	
	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ
1	.006	.011	.004	.011	.004	.012	.004	.011	.005	.024	.010	.016
2	.006	.013	.007	.014	.021	.011	.004	.015	.002	.019	.013	.015
3	.009	.018	.002	.024	.041	.014	.002	.015	.01	.026	.022	.010
4	-.016	.024	.009	.030	.061	.022	.004	.023	-.01	.031	.026	.014
5	.019	.031	.015	.036	.073	.025	.006	.031	-.013	.038	.036	.016
6	.031	.042	.024	.043	.086	.047	.008	.040	-.013	.043	.042	.023
7	.04	.048	.018	.048	.110	.044	.008	.047	-.012	.041	.049	.030
8	.043	.051	.031	.045	.130	.048	.006	.048	-.011	.047	.058	.036
9	.046	.055	.034	.053	.14	.059	.003	.053	-.022	.051	.066	.042
10			.032	.061	.15	.049	.009	.053	-.022	.070	.060	.049
$\Delta\theta$	November				August		November				August	
	B 1300 PST						B 1300 SMP added					
	PDX		SEA		SEA		PDX		SEA		SEA	
	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ
1	.006	.027	.016	.028	0	.022	.015	.027	.013	.035	.0	.021
2	.007	.030	.024	.031	.015	.033	.013	.033	.005	.019	.010	.030
3	.011	.032	.030	.035	.019	.027	.014	.036	.005	.032	.018	.035
4	.015	.038	.039	.037	.017	.036	.016	.035	.003	.042	.021	.021
5	.015	.042	.044	.038	.016	.048	.008	.040	-.004	.050	.016	.033
6	.017	.040	.044	.044	.009	.054	.004	.045	0	.048	.006	.046
7	.016	.048	.047	.047	.020	.072	-.009	.049	-.001	.050	.020	.070
8	-.01	.029	.034	.050			-.022	.055	-.03	.040		
9	0	.03	.033	.069								
10												
$\Delta\theta$	November											
	B 1300 PST						1300 SMP added					
	HMS		HMS									
	$\bar{x}$	σ	$\bar{x}$	σ								
1	-.01	.02	-.01	.024								
2	0	.02	-.01	.029								
3	.01	.03	-.02	.039								
4	.016	.031	-.035	.043								
5	.022	.046	-.045	.060								
6	.022	.056	-.068	.073								
7	.022	.072	-.086	.085								
8	.012	.082	-.13	.092								
9	.001	.11	-.17	.11								
10	.00	.12	-.24	.12								

used in 1974 with the error being plotted versus change in potential temperature ($\Delta\theta$). The $\Delta\theta$ plotted is potential temperature of model minus the surface potential temperature.

As can be seen in Table 1 and Table 2, the results for PDX are approximately the same. The same is true with the SEA with decrease in accuracy in $\Delta\theta$ of 8, 9, and 10. The best improvement was in the mean error in the HMS data with an increase in the ability of the model to estimate the observed sounding. The improved model did not show any increase in ability to reduce the standard deviation.

Another calculation was made using the 1973 result of keeping the error in ψ within $\pm 0.05 \times 10^7 \text{ cm}^2 \text{sec}^{-2}$ for a pressure range within 100 mb of the surface pressure. It was hoped that this technique could be used to filter data with a high error potential. Thus a reliable set of data could be arrived at for trajectory calculations from the stream functions.

The result of this second calculation is compared with the inclusion of all data as shown in Table 3. For November data, SEA and PDX generally show a decrease in the mean and in the standard deviation for the data.

For $\Delta\theta = 5^\circ \text{ A}$, HMS shows a worsening of the data in the mean and a possible worsening in the standard deviation. In the August data, the mean value is definitely higher and the standard deviation nearer zero in the data within 100 mb of the surface, as opposed to total data. However, applying the same procedure to the model with SMP added, the mean is closer to zero and the standard deviation reduced. Thus, it appears that using SMP, as well as restraining the pressure range to surface pressure to surface pressure - 100 mb, results in ψ whose error is $< 0.05 \times 10^7 \text{ cm}^2 \text{sec}^{-2}$, for a $\Delta\theta$ range of 6° A during November for SEA and PDX. In the August data, the range for stream functions $< 0.05 \times 10^7 \text{ cm}^2 \text{sec}^{-2}$ remained $< 3^\circ \text{ A}$ for HMS. When both SMP and pressure restraint were added, the $\Delta\theta$ was increased to $4^\circ - 5^\circ \text{ A}$.

It can therefore be concluded that the model's calculations, by filtering stream function calculations only over a pressure range of surface pressure to surface pressure - 100 mb, and including Stampede Pass into the model, result in Montgomery stream functions within the error limits of $\pm 0.05 \times 10^7 \text{ cm}^2 \text{sec}^{-2}$ for $\Delta\theta$ of $4^\circ - 5^\circ \text{ A}$.

TABLE 2. Errors in ψ ($\times 10^{-7} \text{ cm}^2 \text{sec}^2$) 1973 Model.

$\Delta\theta$	November				November					
	A 0700 PST				B 1300 PST					
	PDX		SEA		PDX		SEA		HMS	
	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ
1	.00	.02	.01	.01	.011	.03	.02	.04	.01	.02
2	.00	.02	.01	.01	.01	.01	.03	.04	.02	.02
3	.00	.02	0	.02	.02	.02	.04	.04	.03	.03
4	.01	.02	.01	.03	.02	.03	.05	.04	.03	.04
5	.02	.02	.02	.04	.02	.04	.05	.04	.05	.05
6	.03	.04	.03	.04	.02	.04	.06	.05	.06	.06
7	.04	.05	.02	.05	.02	.04	.07	.05	.06	.07
8	.04	.05	.01	.05	.02	.04	.06	.05	.06	.08
9	.04	.05	.01	.06	.01	.04	.08	.04	.03	.06
10	.05	.06	.01	.08	.02	.06	.09	.05	.04	.07

TABLE 3. Errors in ψ ($\times 10^{-7}$ cm⁻²sec²) for Combined Times (0700 and 1300 PST).

November							August			
A Within 100 MB of Surface							SMP added			
$\Delta\theta$	SEA		PDX		HMS		SEA		SEA	
	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ
1	.004	.011	0	.01	-.01	.02	.002	.016	.006	.018
2	.010	.014	.010	.020	0	.02	.016	.017	.012	.022
3	.009	.022	.006	.015	.010	.025	.038	.016	.019	.013
4	.018	.020	.010	.020	.025	.033	.058	.021	.027	.014
5	.023	.029	.020	.030	.038	.052	.071	.024	.037	.016
6	.025	.025	.010	.023			.088	.035	.040	.022
7	.021	.030	.				.10	.04	.047	.028
8	.030	.038					.11	.04		
9							.126	.051		
10							.120	.049		

November							August			
B Total							SMP added			
$\Delta\theta$	SEA		PDX		HMS		SEA		SEA	
	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ
1	.01	.02	.006	.020	-.010	.020	.002	.016	.006	.018
2	.017	.025	.006	.021	0	.020	.019	.022	.012	.022
3	.018	.033	.010	.024	.010	.030	.034	.021	.020	.022
4	.026	.038	.015	.030	.016	.031	.047	.033	.025	.018
5	.031	.040	.017	.036	.022	.046	.055	.043	.032	.020
6	.037	.046	.023	.042	.022	.056	.060	.061	.030	.036
7	.039	.047	.030	.048	.022	.072	.086	.064	.041	.042
8	.037	.048	.026	.050	.012	.082	.100	.077		
9	.041	.061	.026	.052	.001	.110	.114	.091		
10	.043	.065	.022	.061	00	.120	.127	.080		

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REGIONAL TRANSPORT AND DIFFUSION STUDIES
OVER THE COLUMBIA BASIN

W. F. Sandusky and D. L. Elliott

A case study of wind fields and transport over the Columbia Basin has been made. The analysis, using techniques employed in mesoscale research, reveals spatial and temporal variability in computed wind fields and hypothetical particle trajectories. Much of the variability in the flow can be attributed to the topographical influence on the regional flow pattern.

INTRODUCTION

With an increasing interest in Hanford as a site for a nuclear park and waste storage facility, radiological dose assessment on the regional scale (>100 km) becomes important. Previous studies¹ on the mesoscale (10 km-100 km) have shown that dose assessments based on the single station wind rose approach for the accidental or extended release case could be in serious error. This is due to the fact that terrain variation has an effect on the low level wind field not accounted for by a wind rose scheme. With these thoughts in mind, selected case studies of transport and diffusion on the regional scale were made as an extension to mesoscale transport and diffusion studies underway at Hanford.

WIND FIELD AND TRAJECTORY STUDIES

The data base for the study included wind speed and direction data from 22 National Weather Service reporting stations located in eastern Washington, northeastern Oregon and northwestern Idaho. This data, whether reporting hourly or once every 3 hours, was put into the same model as used in mesoscale research.² However, the study area was expanded to include a region contained by the Columbia Basin and the various topographic features surrounding the basin, since a major objective here is to investigate the influence of topography on the regional wind flow pattern. Figure 1 is an example of computer plotted wind fields using September 15, 1973, as a test case.

This day was chosen since relatively high winds occurred in the Hanford area. The temporal changes in the wind field can easily be seen along with a rather large scale eddy in east-central Washington. In the last two map sequences (2000, 2300), the diurnal effects on the wind field are evident. During the nighttime, drainage flow from the higher elevations down into the basin area is established.

In conjunction with the wind field analysis, trajectory plots (Figure 2) of serially released hypothetical "particles" were produced. This type of computer plot allows some indication of a plume configuration for an accident analysis.

Trajectories, based on single station wind data, were also constructed. In this case, the Hanford tower winds were used as if they applied over the whole region. Comparison of the wind field and single station trajectory plots reveals pronounced differences in the regional transport pattern, resulting from the two methods. Much of this difference can be attributed to topographical influences and drainage winds during the night and early morning.

No attempt was made to validate the trajectory plots. However, from SF₆ tracer studies in the mesoscale transport research program,³ some degree of confidence in the trajectory plots has been established under certain atmospheric conditions for distances up to 60 km.

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FIGURE 1. Examples of Wind Field Plots over a Grid Covering Eastern Washington, Northeastern Oregon and Northwestern Idaho. The dotted figure in south central Washington represents the Hanford Area. The wind data from each station are plotted in standard form, and the interpolated winds are plotted as vectors at the grid points.

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FIGURE 2. Examples of Trajectory Plots Showing the Paths of Hypothetical Particles Released Every 3 Hours and Transported by Time Series of Objectively Interpolated Wind Fields. The numbers at the ends of the trajectories indicate the order of release. The letters along the trajectories represent position every 3 hours. Initial particle starting time and date is given at the top of each plot. WF and SS represent wind field and single station analysis.

DISCUSSION AND RECOMMENDATIONS

From this limited study, it is seen that spatial variability of the winds and resulting transport patterns in the near surface layer are present. Another interesting feature, which is not presented here, is the generated wind fields at various levels of the troposphere. These plots, which were generated from rawinsonde and pibal (pilot balloon) data, provide some indication of the vertical shear of the wind field with height and allow

for an improved estimate of the transport pattern under various atmospheric stability conditions.

As Hanford grows in terms of nuclear facilities, the needs for regional assessment techniques also grows. The model described here in terms of graphical display techniques could be a vital tool for Hanford meteorologists and radiologists in routine release or accident case analysis.

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A SYSTEMATIC ERROR IN THE MEASUREMENT
OF PLUME CROSSWIND CONCENTRATION
DISTRIBUTIONS WITH MOVING SAMPLERS

J. V. Ramsdell and P. Michael*

A systematic error in measuring the variance of the crosswind concentration distribution of real plumes with mobile samplers is examined mathematically. The error arises when the movement of the plume during a sampler traverse is neglected. On an individual traverse the error may result in either over- or underestimates of the true concentration variance. However, when a number of traverses are made there is a systematic overestimate of variance, the magnitude of which is a function of the relative velocities of the plume and sampler.

INTRODUCTION

The increased interest and emphasis on long range diffusion measurements has necessarily led to the use of mobile samplers in diffusion measurement programs. These measurements are fundamentally different from the measurements which have been made with fixed samplers arrayed in stationary grids. The moving sampler measurements essentially describe an instantaneous cross-section of a plume, while the latter describe a cross-section of the time integrated plume. The variances of the concentration distribution obtained in the two experimental techniques can be related to one another using the model of Gifford.¹ Experimental data on the relationship has been given in past reports in this series.²⁻⁵

It has been established that to a reasonably high degree the two variances are related by

$$\sigma_y^2 = \sigma_{yi}^2 + M_i^2 \quad (1)$$

where σ_y^2 is the crosswind variance of the time integrated plume concentration distribution, σ_{yi}^2 is the mean variance of the instantaneous plume crosswind concentration distribution, and M_i^2 is the variance of

the positions of the center of mass of the instantaneous plume. Examination of the estimation of σ_{yi}^2 and σ_{yi}^2 follows.

Basic Formulation of the Variance

The concentration distribution variance of a real plume using a moving sensor can be rigorously formulated as

$$\sigma_{yi}^2 = \frac{\int_{y_1}^{y_2} \left\{ \int_{t_0}^t (v_s - v_p) dt \right\}^2 C(y) dy}{\int_{y_1}^{y_2} C(y) dy} - \left[\frac{\int_{y_1}^{y_2} \int_{t_0}^t (v_s - v_p) dt C(y) dy}{\int_{y_1}^{y_2} C(y) dy} \right]^2 \quad (2)$$

where V_s and V_p are the crosswind components of the velocities of the sampler and plume respectively, $|V_p| < V_s$, $C(y)$ is the concentration at position y , y_1 and y_2 are positions on either side of the plume,

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and t_0 and t are the time of the start of the traverse and the time at position y respectively.

The form of Equation (2) can be simplified considerably if the sampler and plume velocities are considered to be constant during any single traverse. In this case taking $t_0 = 0$,

$$\sigma_{yi}^2 = (V_s - V_p)^2 \left[\frac{\int t^2 C(y) dy}{\int C(y) dy} - \left(\frac{\int t C(y) dy}{\int C(y) dy} \right)^2 \right] \quad (3)$$

For practical problems it is usually convenient to replace $C(y) dy$ in Equation (3) by $C(t) dt$.

The assumption of a constant velocity for the sampler follows directly from the usual experimental technique; the assumption of a constant plume velocity is made to simplify the analysis. In analysis of diffusion data obtained with a moving sampler a more restrictive assumption with respect to plume velocity is generally made. That assumption is that the plume velocity is not only constant, but that it is identically zero. If the concentration variance estimated under this assumption is denoted S_{yi}^2 , the relationship between it and σ_{yi}^2 is given by

$$\frac{S_{yi}^2}{\sigma_{yi}^2} = \frac{V_s^2}{(V_s - V_p)^2} = \left(1 - \frac{V_p}{V_s} \right)^{-2} \quad (4)$$

Thus, if on a particular traverse the plume and sampler are moving in the same direction, the variance will be over-estimated. Conversely if they are moving in opposite directions the variance will be underestimated. This then identifies the nature of the error to be examined.

Convergence of the Estimate of the Mean Variance

The relationship between the mean and instantaneous plume crosswind

concentration variances in Equation (1) requires an estimate of the mean crosswind concentration variance in the instantaneous plume, not an estimate of the variance on an individual traverse. Since Equation (4) indicates that the error in the estimate of the variance on individual traverses may be either positive or negative, the estimate of the mean variance of the instantaneous plume concentration distribution might be assumed to converge to the true value as the number of traverses increases. However, that is not the case. The estimated mean variance converges to an over-estimate of the true variance except when the plume velocity is identically zero.

If the plume crosswind velocity is assumed to be a random variable, Equation (4) may be averaged according to

$$\left(\frac{S_{yi}^2}{\sigma_{yi}^2} \right) = \int_{V_{p1}}^{V_{p2}} \left(1 - \frac{V_p}{V_s} \right)^{-2} P(V_p) dV_p \quad (5)$$

where $P(V_p)$ is the probability distribution associated with the values of V_p between V_{p1} and V_{p2} . This integral has been evaluated for three cases: a plume moving at a constant velocity, a uniform distribution of plume velocities, and a plume velocity distribution given by $V_p = bV_s \cos \theta$, where θ has a uniform distribution between 0 and π , and b is a constant between 0 and 1.

Briefly, the first case corresponds to traversing through the portion of a plume which was released during a gradual change in wind direction or for a single traverse through the plume. In this case the probability density function for plume velocity is

$$P(V_p) = \begin{cases} 1/2 V_p = bV_s \\ 1/2 V_p = -bV_s \\ 0 \text{ otherwise} \end{cases}$$

where b is a positive fraction less than 1. When these values are substituted into Equation (5) the result is

$$\left(\frac{S_{yi}}{\sigma_{yi}}\right)^2 = \frac{1 + b^2}{(1 - b^2)^2} \quad (6)$$

The second case probability density function is

$$P(V_p) = \begin{cases} \frac{1}{2bV_s} & -bV_s < V_p < bV_s \\ 0 & \text{otherwise} \end{cases}$$

where again b is less than 1. Introducing this function into Equation (5) results in

$$\left(\frac{S_{yi}}{\sigma_{yi}}\right)^2 = \sum_{n=0}^{\infty} b^{2n} = \frac{1}{1-b^2} \quad (7)$$

Finally in the third case the probability density function is given in terms of a randomly distributed angle θ rather than a probability density function for V_p . The two density functions are related by

$$P(V_p) dV_p = P(\theta) d\theta \quad (8)$$

The hypothesized density function for $P(\theta)$ is

$$P(\theta) = \begin{cases} \frac{1}{\pi} & 0 < \theta < \pi \\ 0 & \text{otherwise} \end{cases}$$

When this density function and the relationship $V_p = bV_s \cos \theta$ are substituted into Equation (5) and the resulting expression integrated the result is

$$\left(\frac{S_{yi}}{\sigma_{yi}}\right)^2 = \frac{1}{(1 - b^2)^{3/2}} \quad (9)$$

In all cases the concentration variance estimated by assuming a stationary plume is larger than the true variance. The seriousness of this bias is seen to be a function of the absolute value of the ratio between the assumed plume and sensor velocity components which has been indicated by the parameter b . The

expressions given in Equations (6), (7) and (9) have been evaluated for three values of b , and the results are presented in Table 1. This table clearly shows that when b is large the bias is significant and that as b approaches zero the bias becomes negligible.

TABLE 1. Ratio Between Crosswind Concentration Variance for Stationary and Moving Plumes for Three Assumed Plume Velocity Distributions and Three Values of the Parameter b . All Values Indicate an Overestimate of the Variance.

Case	1/2	1/4	1/10
1	2.222	1.209	1.031
2	1.333	1.067	1.010
3	1.540	1.102	1.015

Discussion of Results

The results given above have two significant implications in the measurement of plumes using moving samplers. The first is that to minimize the systematic overestimate of the mean instantaneous plume crosswind concentration variance, the sampling traverse should be made as rapidly as possible. Of course, instrument response and resolution of the recorder used effectively limit the maximum traverse speed. The effects of slow traverse speeds are more likely to be significant when samplers are carried on surface vehicles than when they are carried on aircraft. The second implication of the results is that plumes should be traversed a sufficient number of times to avoid the situation represented in case 1. If these precautions are taken, the bias in the estimate of the concentration distribution crosswind variance will be small compared to the random variations of the variance. If not, the bias may be significant.

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TRANSPORT AND DIFFUSION MEASUREMENTS
WITH AN AIRBORNE GAS CHROMATOGRAPH

R. N. Lee, M. M. Orgill,
L. L. Wendell and P. W. Nickola

The physical and chemical properties of sulfur hexafluoride (SF_6) have prompted its frequent application as an atmospheric tracer. A real-time SF_6 detection system constructed for aircraft operation has undergone extensive testing during the winter and spring of 1974. Data are reported which confirm its utility and identify its limitations.

INTRODUCTION

Sulfur hexafluoride (SF_6) and halocarbons of the freon type have been found to be of great value as tracers in atmospheric transport and diffusion studies over the mesoscale range.^{1,2} These materials are well suited to the requirements of these studies by virtue of their ready detection at sub-ppb levels, nondepositing nature, nontoxicity and chemical inertness. An aircraft mounted system for the real-time detection of SF_6 has recently undergone several tests in the vicinity of the Hanford Reservation. These flights have permitted an appraisal of its operational characteristics, testing of a plume tracking procedure and generated data useful in evaluating recent mesoscale modeling efforts.

SULFUR HEXAFLUORIDE DETECTION SYSTEM

A schematic of the SF_6 detection system appears in Figure 1. This system is nearly identical to that described in an earlier report.³ The only modification has been the change in location of solenoid valve B. Operation of the system, based on frontal elution chromatography, remains unchanged. Sensitivity of the electron capture detector to oxygen limits the sampling time to 10-15% of the flight time, and necessitates a periodic backflush to purge oxygen from the system.

Concentration data recorded in this paper are based on a laboratory

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FIGURE 1. Schematic of SF_6 Detection System in Backflush Mode. Valve positions during sampling mode of operation are indicated by dashed lines.

calibration of the system. This was done by preparing a concentrated SF_6 -air mixture. Syringe addition of this mixture to a stream of bottled air using a syringe pump and Matheson Dyna-Blender dilution system permitted the preparation of SF_6 -air sample in the appropriate concentration range.

PLUME TRACKING

A total of 12 experimental flights have been conducted to date. Release of SF_6 has been from the top of

the Hanford Meteorological Tower (121 m) except during very windy conditions, when the source was located at ground level. Table 1 lists the dates and times for seven of these flights along with the general meteorological conditions. Difficulties in locating the plume, meteorological conditions and instrumentation problems limited the quality of data acquired during the remaining flights.

Constant volume balloons were used to track SF₆ plumes during five of the flights listed in Table 1. These were launched a few minutes after initiation of SF₆ gas release to provide a visual indication of plume location. Although the observed SF₆ plume segment paths were nearly identical to the constant volume balloon trajectories (Figure 2), this method of plume tracking was not totally satisfactory. The need to maintain visual contact with the balloon prevented adequate vertical and lateral sampling. Concentration data accumulated during these tests have generally been near source height (120 to 150 m above ground). Specific values referred to in the next section are the maxima observed for a single traverse of the plume.

A second disadvantage of this technique relates to the distance over which SF₆ may be followed. Since the problem of locating the plume has been overcome with the use of a constant volume balloon, sampling has been restricted to the air parcel surrounding the balloon. Consequently, few measurements have been made at large distances from the source. The maximum distance thus far achieved (~65 km) does not represent the limiting range of the system.

In addition to the aforementioned tracking problems, the aircraft ground speed (~63 m/sec) and the restricted sampling time (~50 sec) place a limit of approximately 3 km on the width of the sampling window. At distances out to 10 km the plume was generally well defined so that a complete cross-sectional display of tracer concentration could be seen. Beyond this distance, the plumes were more diffuse and a complete profile was generally not possible. Although limitations of the sampling system and tracking technique must be recognized, the potential value of this tracer system to atmospheric studies is significant as illustrated in the following section.

TABLE 1. SF₆ Experimental Dates and Conditions.

FLIGHT NUMBER	DATE	TYPE OF RELEASE	TIME OF RELEASE (PST)	AVERAGE SOURCE RATE (kg/hr)	SURFACE STABILITY	PASQUILL STABILITY CATEGORY	WIND AT SOURCE (mps)	CONSTANT- VOLUME BALLOON
3	12/26/73	TOWER (121 m)	14:00 - 14:10	6.5 (36 cfh)	SLIGHTLY STABLE	E	NW/5	NO
4	1/3/74	TOWER (121 m)	10:29 - 11:32	6.5	NEUTRAL TO SLIGHTLY STABLE	D TO E	NW-ESE 1	YES
6	1/22/74	SURFACE	11:36 - 12:36	13	SLIGHTLY STABLE	E	SW/11	YES
7	1/23/74	TOWER (121 m)	12:37 - 14:00	13 (72 cfh)	NEUTRAL TO SLIGHTLY STABLE	D TO E	WSW/8	NO
9	1/30/74	TOWER (121 m)	11:17 - 12:32	13	SLIGHTLY UNSTABLE TO NEUTRAL	C TO D	NNW-ESE 2	YES
11	5/29/74	TOWER (121 m)	05:25 - 06:42	8-17 (42-96 cfh)	SLIGHTLY STABLE TO EXTREMELY UNSTABLE	E TO A	WNW-NW 8	YES
12	6/3/74	TOWER (121 m)	05:29 - 07:10	20.6 (114 cfh)	SLIGHTLY STABLE TO MODERATELY UNSTABLE	E TO B	WNW-W 4.5	YES

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FIGURE 2. Hanford Area with Wind Measurement Locations, Numerical Analysis Grid, SF₆ Detection Locations and Constant-Volume Balloon Trajectories. 1 - January 30, 1974; 2 - January 22, 1974; 3 - January 30, 1974.

COMPARISON BETWEEN TRANSPORT AND DIFFUSION MODELING AND SF₆ SAMPLING RESULTS

These test results have provided valuable information for testing and validation of some current transport and diffusion modeling efforts. Data from a network of wind towers over the Hanford Reservation were used to generate wind fields and trajectories from a previously developed computational and plotting technique.⁴ Figure 3 shows trajectories computed for periods when SF₆ sampling was in progress as compared to actual CVB trajectories, and locations where SF₆ was detected. Flights 4 (January 3) and 9 (January 30) showed the largest differences between observed and calculated trajectories. On January 3, due to a failure of the wind telemetry system, the computed trajectory was determined using wind

data from the source only. Close agreement with the observed trajectory is not to be expected. During the January 30 flight, a wind shift developed in the upper region of the boundary layer and transported the CVB and SF₆ plume to the west. The lower telemetering winds (15-30 m) failed to reflect this change in the wind direction until somewhat later resulting in a large discrepancy in the computed trajectory. In the case of flight 6 (January 22), the trajectory of the SF₆ plume segment around the CVB and the computed trajectory were approximately the same.

The downwind centerline concentration at the most frequent sampling height was estimated with the standard continuous plume formula and parameters by assuming $Z \sim H$ (effective emission height). The modified expression which was used for this calculation is

$$x = \frac{Q}{\sqrt{2\pi} \sigma_y L \bar{u}} \quad (1)$$

where Q is the source strength, $\bar{u}$ mean wind speed affecting the plume, σ_y the lateral standard deviation in the plume concentration distribution and L the mixing depth. Substitution of the appropriate values for Q , $\bar{u}$, σ_y , and L in Equation (1) permitted centerline concentrations to be computed for the distances where SF₆ was measured.

The computed and observed concentrations as a function of distance for the May 29, 1974, flight are presented in Figure 4. The similar slopes and calculated concentration curves are indicative of similar dilution rates. This was achieved with the calculated curve by varying the mixing depth (L) and the mean wind velocity ($\bar{u}$) with time or distance.

The discrepancy between observed and calculated concentration values probably results from the parameters σ_y , L and $\bar{u}$, used for obtaining calculated concentrations, being average values while the observed concentrations were derived from instantaneous measurement.

One striking feature of the observed centerline concentration was the abrupt change in dilution rate

*TRAJECTORY COMPUTED USING SOURCE WIND ONLY

**SF₆ SOURCE WAS AT GROUND LEVEL

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FIGURE 3. Computed and Observed Trajectories of Constant-Volume Balloons from SF₆ Source and Selected SF₆ Detection Locations.

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FIGURE 4. SF_6 Centerline Concentration at 121 m above Ground as a Function of Distance for Flight #11.

at about 35 km. This was apparently the result of a change in atmospheric stability, or mixing depth, follow-

ing sunrise on this clear warm spring morning.

Centerline concentrations have been calculated for four additional tests in the series using a similar approach. As with the May 29 flight, calculated and observed dilution rates were similar and the observed instantaneous concentrations were consistently higher than the calculated values.

CONCLUSIONS

The utility of SF_6 as an atmospheric tracer has been confirmed by a series of experiments over the Hanford Reservation. Like Dietz, we have found the value of this material to be enhanced by the use of an aircraft mounted real-time direction system. Some of the limitations which have been identified during this study are associated with plume tracking and may be at least partially eliminated by modification of the CVB tracking technique.

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FREQUENCY DISTRIBUTION OF ATMOSPHERIC TRACER
CONCENTRATION DURING PERIODS OF LOW WINDS

P. W. Nickola, G. H. Clark*
and J. D. Ludwick

Tracer releases into stable and unstable atmospheres are examined with respect to the frequency distribution of short-period (10-sec) concentrations. Wind speed was about 1.5 m/s during each experiment. Although unstable conditions resulted in a greater number of excursions of concentration above the mean plume centerline concentration than did stable atmospheric conditions, stable conditions had a greater proportion of concentrations above four times that mean.

In the two low wind speed experiments examined, the observed exposures were considerably lower than those predicted by a Gaussian plume model with Pasquill's frequently employed dispersion parameters.

INTRODUCTION

Most computational formulae and models for diffusion in the atmosphere deal with relatively long-period averages of concentration--periods on the order of tens of minutes and longer. However the turbulent nature of the atmosphere causes many short-period deviations from the mean concentration. It is not only the tolerance of a receptor to the mean concentration that should be of concern, but also the tolerance to the short term excursions from this mean. Two recent tracer releases on the Hanford diffusion grid afforded an opportunity to examine the short-period fluctuations of concentrations in considerable detail.

THE FIELD GRID AND EXPERIMENTS

The two experiments examined entailed release of the tracer krypton-85 for periods of about 30 min. Release was at a constant rate from an elevation of about 0.3 m. Sampling,

primarily at a height of 1.5 m, extended to a distance of 300 m from the release point. Concentrations were recorded as a series of consecutive 10-sec means at each of 127 field locations.

The experiments were carried out during a series of "ground-level" tracer releases in ostensibly calm winds. Although there were brief periods during both experiments when the winds were below anemometer threshold values, wind speeds averaged about 1.5 m/s during each test.

Meteorological and stability data pertinent to the tests are presented in Table 1. The meteorology was measured on towers about 500 m from the tracer release point.

Terrain in the area is quite flat. Vegetation is primarily sagebrush and steppe grasses.

Although the period of tracer release was about 30 min in each experiment, the near real time mode of the tracer data recording system permitted selection of a 22-min period from the "middle" (with respect to time)

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TABLE 1. Winds, Temperatures and Stability during Experiments C6 and C7.

METEOROLOGY

Elevation	Speed (m/s)		σ_u (m/s)		σ_θ (deg)		Elevation	Temp ($^{\circ}$ F)	
	C6	C7	C6	C7	C6	C7		C6	C7
0.76 m	1.1	0.8	0.44	0.39	18.0	19.0	0.9 m	49.5	39.2
1.5	1.5	1.4	0.56	0.36	16.6	15.1	6.1	49.2	41.2
3.0	1.7	1.8	0.66	0.40	15.0	13.6	15.2	48.5	41.9
6.1	1.9	2.3	0.70	0.45	16.3	11.6	30.5	47.9	42.6
12.2	1.9	3.0	0.71	0.50	14.4	7.8			
24.4	2.1	-	0.76	-	15.7	6.0			

STABILITY

Test	Richardson No. (2.1 to 50 m)	pasquill Class by Regulatory Guide 1.23 ⁽¹⁾				Pasquill Class by Weather Conditions*
		ΔT		σ_θ		
		30.5 to 0.9 m	15.2 to 0.9 m	1.5 m	24.4 m	
C6	-1.40	A	A	C	C	D (Overcast)
C7	0.16	G	G	C	E	E/F ("Night", 0.4 Cloud Cover)

* From Table 3-1 of Turner Workbook⁽²⁾

of each plume. Thus equilibrium portions of the plumes (portions without leading or trailing edge effects of diffusion) were selected for analysis.

THE MEAN PLUME

Figures 1 and 2 depict mean concentrations or exposures obtained by integrating over the selected 22 min periods. Figure 3 compares the mean peak concentrations (normalized to the low but measurable wind speeds) with similar concentrations generated from a Gaussian plume model with Pasquill's familiar stability curves. The computed curves are those given in Figure A-1 of Meteorology and Atomic Energy (1968).³ It is clear that despite the latitude in method of selection of stability class (see Table 1), use of the Pasquill curves results in overestimation of the mean peak concentration ($\bar{x}_{pu}/Q'$) in both cases. Although these experiments were not carried out during completely calm winds, it is comforting to find that the extremely high concentration sometimes expected with very low wind speeds was not experienced during release into either an unstable or a stable atmosphere.

FIGURE 1. Test C6. Mean Concentration ($\bar{x}$) Normalized to Unit Emission Rate (Q'). Units of Plotted and Isopleth Data are $10^{-5} \text{ sec m}^{-3}$.

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FIGURE 2. Test C7. Mean Concentration ($\bar{x}$) Normalized to Unit Emission Rate (Q'). Units of Plotted and Isopleth Data are $10^{-5} \text{ sec m}^{-3}$.

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FIGURE 3. Mean Concentration Normalized to Unit Wind Speed ($\bar{u}$) and Emission Rate Versus Distance from Source. Test C6, Unstable (Pasquill A to D); Test C7, Stable (Pasquill E to G).

CONCENTRATIONS DURING SHORT INCREMENTS OF TIME

The distribution of concentrations contributing to the mean is obtainable from records generated by the Hanford inert gas tracer monitoring system.⁴ In the subject experiments (C6 and C7), concentrations were recorded as a series of end-to-end 10-sec mean concentrations at each of 127 field locations. The pattern of the sampling field grid at the 1.5 m elevation is suggested by the sampler positions indicated in Figures 1 and 2.

All short-period (10-sec) concentrations (x_{10}) were normalized to a fraction of the mean plume centerline concentration ($\bar{x}_p$). This $\bar{x}_p$ is the parameter most generally of interest, and most frequently estimated in hazards appraisals.

Figure 4 gives least squares curves relating the frequency of occurrence of various normalized short-period concentrations ($x_{10}/\bar{x}_p$) to the location in the plumes. The location is specified with respect to the plume centerline ($\bar{x}_{\text{Local}}/\bar{x}_p$) at the same distance from the source. The points from which the curve for $x_{10}/\bar{x}_p \geq 2$ were generated are plotted to give an idea of the scatter of data from which curves on Figure 4 were generated.

There appeared little reason to present separate curves for the data generated at 100 m versus that generated at 300 m. However, except for the $x_{10}/\bar{x}_p \geq 2$ curve, there did appear to be a stratification by field test. This difference was presumed due to the difference in stability.

An example conclusion from the curve $x_{10}/\bar{x}_p \geq 2$ is that regardless of stability, the mean plume centerline location experiences short-period (10 sec) concentrations at least twice the mean centerline concentration about 20% of the time. At a location where the mean concentration is only half the mean plume centerline concentration, the short-period concentration exceeds the mean centerline concentration about 9% of the time.

Note that unstable test C6 showed a higher frequency of $x_{10}/\bar{x}_p \geq 1.0$ than did the stable test. Conversely,

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FIGURE 4. Frequency of Occurrence of Various Normalized Short-Period Concentrations Versus Location with Respect to Plume Centerline.

stable experiment C7 showed a higher frequency of $x_{10}/\bar{x}_p \geq 4.0$ than did the unstable experiment. As mentioned earlier, no stability effect was discernable at $x_{10}/\bar{x}_p \geq 2.0$. Thus the mean concentration during unstable conditions evolved from a greater number of short-period concentrations relatively close to the mean concentration; the mean during stable conditions evolved from a smaller number of relatively high concentration excursions from the mean.

Also displayed in Figure 4 are curves for the stable and unstable tests depicting the minimal percent of time (i.e., the percent of 10 sec "puffs," not necessarily consecutive) required for one half of the tracer mass to pass. That is, the 10-sec concentrations for a given sampler were ordered by magnitude, and an integration was carried out starting with the highest concentration. The number of time periods necessary to sum to half the mass or exposure passing the sampler determined the fraction

of time plotted on the half-mass curve. Again the effects of fewer but larger short-period concentrations are in evidence for the stable case.

The abscissa along the top of Figure 4 defines position in the plume as a function of σ_y rather than as a fraction of centerline concentration.

Ramsdell⁵ reported "peak-to-mean" concentration ratios of about 4 for near plume centerline locations. This ratio of 4 was for T/t ratios of about 20 but appeared unlikely to increase significantly with increasing T/t . (T/t is the ratio of times used in computing the mean and the short-period concentrations.) Wind speeds during Ramsdell's experiments averaged 3.8 mps. Although the data are not shown here, for the current low wind speed cases, with a T/t of 132, the peak-to-mean ratio near plume centerline seems to level off at an average near 8 for both stable and unstable conditions.

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MICROSCALE LOW WIND SPEED TRANSPORT AND DISPERSION CALCULATIONS

G. H. Clark* and L. L. Wendell

A mesoscale atmospheric transport and dispersion model has been modified for a microscale application to distances of 300 m downwind of a pollution source. Preliminary results from three light wind dispersion tests have shown significant inhomogeneity in the prevailing wind fields and encouraging trends in the computation of σ_y and σ_z from wind turbulence intensities.

A study has been initiated to directly compare the results from a microscale application of a transport and diffusion model with the simultaneously measured krypton gas exposures during experiments conducted under light wind or stagnation conditions.

As described by Nickola and Clark,¹ a concentric array of Geiger-Müller samplers has been installed 1.5 m above ground level at distances of 25, 100, 200, and 300 m from the source. The sampler separations on the respective arcs are 90, 15, 20, and 10 degrees of azimuth. In order to define the prevailing wind field and turbulence conditions across the sampler array, three component Gill UVW anemometers at 3.7 m above ground level have been positioned near the source and on the 300 m arc north, south, east, and west of the array center. The wind data were recorded on a Metrodata 1/4-in. digital magnetic tape recorder with a sampling interval of 0.41 sec per scan of input channels. A supporting analogue magnetic tape recorder was also operated simultaneously. In addition, an independent wind speed and direction instrument with similar response characteristics and strip chart output was located near the source Gill anemometer for a direct comparison of the wind time histories.

To date, three light wind tests have been conducted, two under near neutral to unstable daytime conditions, and a third with a more stable early morning atmosphere. In Figure 1, an example is presented of the wind fields from the unstable-neutral test, designated C6. The winds have been averaged over about 52.5 sec, and it is obvious there can be great inhomogeneity across the sampler array, especially under

FIGURE 1. Wind Fields (Averaged over 52.5 sec) on the Light Wind "Stagnation" Grid During Test C6.

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the light wind conditions. This will be an important consideration for the atmospheric transport aspects of the model.

In the application of the meso-scale transport and diffusion model of Start and Wendell² in the micro-scale, both in space and time, it is inappropriate to use the Pasquill-Gifford diffusion parameters, which implicitly apply to diffusion times on the order of 20 minutes. Csanady³ and others have suggested that close to the source both the vertical and horizontal diffusion parameters can be given by Equation (1).

$$\sigma_z = \frac{\sigma_w}{\bar{u}} \cdot x \quad (1)$$

$$\sigma_y = \frac{\sigma_v}{\bar{u}} \cdot x$$

where σ_w , σ_v are the standard deviations of vertical and lateral wind speeds, respectively; $\bar{u}$ is the mean wind speed and x the distance downwind.

As a first attempt at the problem, we have calculated the turbulence intensities ($\sigma/\bar{u}$) from an average of the 52.5 second data from the five G-11 anemometers. Variations of σ_y and σ_z , with downwind distance x measured and computed using techniques from Start and Wendell,² are plotted in Figure 2. For test C7, the stability classification was anywhere between B and G, depending on the criteria used (e.g., USAEC Safety Guide 23; σ_θ ; Pasquill by weather conditions).⁴ A comparison of the computed curves with those of

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FIGURE 2. A Comparison of the Measured and Computed Diffusion Parameters, σ_y and σ_z , with the Pasquill-Gifford Curves for Test C7.

Pasquill-Gifford for the appropriate stability classes shows good agreement in slope.

In future, a direct comparison will be made between the observed krypton exposures at each sampler location and the similar transport and diffusion model values computed at the same position. For this to be successful, it is essential to have excellent time agreement between the different data acquisition systems. Experimental refinements are currently being implemented towards this end.

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COMPARATIVE ANALYSIS OF ATMOSPHERIC BOUNDARY LAYER TURBULENCE THROUGH A 60-M LAYER

D. C. Powell and T. W. Horst

Power spectral analysis is made of Hanford site wind component data taken by sonic anemometers at 7.5, 15, and 60 m. The analysis indicates that extension of the constant flux layer concept to 60 m is justified for quasi-neutral conditions but not for stable conditions. It is also shown that considerable latitude must be allowed when applying published power spectral models to power spectral analysis of similar data from another homogeneous site by different authors.

INTRODUCTION

Many semiempirical expressions for modeling dimensionless power spectra may be found in literature on atmospheric boundary layer turbulence. In this article sample power spectra from analysis of wind components at the Hanford site are used, not to propose another set of model curves, but to indicate what departures may be found when published modeling curves are compared with spectral analysis of data from another homogeneous site by different authors. The modeling expressions were previously published by the Air Force Cambridge Research Laboratory (AFCRL).^{1,2}

Some departure may be expected when comparing such analyses, since different investigators use different analysis methods and data from different quasi-homogeneous sites.

The Hanford data analyzed are sonic anemometer readings taken at heights of 7.5, 15, and 60 m on the same tower on the Hanford site. Since the AFCRL turbulence measurements were taken at heights not exceeding 23 m, there is particular interest in the fit of the analysis of the Hanford 60-m data into the constant-flux-layer parameterization determined by measurements at the lower levels.

HANFORD DATA AND AFCRL MODELS USED

The following comparisons are based on analysis of four quasi-neutral tests and two stable tests. Specifics regarding the tests are as follows.

In Table 1, $\bar{U}$ is the mean wind speed at 15 m, k is Von Karman's constant, and L is the Monin-Obukhov stability length given by

TABLE 1. Test Specifics.

Test Name	Time-date	$\bar{U}$ (mps)	kL (m)
EB4	2213-2253 July 21, 1970	5.5	112
EB5	0106-0146 July 22, 1970	6.3	110
EB6	2143-2237 July 23, 1970	8.6	1300
EB8	1910-2004 July 28, 1970	6.9	-760
EB11a	2358-0052 July 30-31, 1970	3.7	3.8
EB11b	0122-0216 July 31, 1970	3.6	7.3

$$kL = -u_*^3 / [(g/\theta)\overline{w'\theta'}] \quad (1)$$

where u_* is the friction velocity, g is the earth's gravitational constant, θ is the mean absolute temperature, and $\overline{w'\theta'}$ is the temperature flux. The first four tests listed are quasi-neutral, and the last two are quite stable.

The AFCRL power spectral models for the quasi-neutral tests are given¹ by Equations (2), (3), and (4).

$$\frac{nS_u(n)}{u_*^2} = \frac{105f}{(1 + 33f)^{5/3}} \quad (2)$$

$$\frac{nS_v(n)}{u_*^2} = \frac{17f}{(1 + 9.5f)^{5/3}} \quad (3)$$

$$\frac{nS_w(n)}{u_*^2} = \frac{2f}{1 + 5.3f^{5/3}} \quad (4)$$

In the above equations n is frequency, and $nS_\alpha(n)$ is the logarithmic power spectrum (hereafter called the power spectrum) for $\alpha = u, v, w$, the longitudinal, lateral and vertical wind components, respectively. The dimensionless frequency, f , is given by

$$f = n(z - d)/\bar{U} \quad (5)$$

where z is the height of measurement, and d is the displacement length (given as 1 m for the Hanford site).

For the stable cases the modeling equation is²

$$\frac{nS_\alpha(n)}{\sigma_\alpha^2} = \frac{0.164[f/(f_0)_\alpha]}{1 + 0.164[f/(f_0)_\alpha]^{5/3}}, \quad (6)$$

$\alpha = u, v, w$, where σ_α^2 is a turbulence variance and $(f_0)_\alpha$ is the value of f at which the $-2/3$ slope of the high frequency range extrapolates to a dimensionless spectral value of unity. According to Kaimal² the f_0 are empirical functions of the Richardson number, $Ri(z)$, given by

$$(f_0)_u = 2.8 Ri(z), \quad (7)$$

$$(f_0)_v = 1.5 Ri(z), \quad (8)$$

$$(f_0)_w = 0.5 Ri(z), \quad (9)$$

for $0.05 \leq Ri(z) \leq 0.2$.

CONSTANCY OF MOMENTUM FLUXES WITH HEIGHT

The ratios of the momentum flux at 60 m to that at 7.5 m for the six tests are given in Table 2. For the four quasi-neutral tests (EB4 through EB8) the mean ratio is 1.02, and the standard deviation is 0.43. For near-neutral stability, Haugen, et al.³ found an average ratio of about 1.0 and a standard deviation of about 0.3 for $\tau_{22.6}/\tau_{5.6}$. Since the mean ratio is about the same for the two sets of data and the standard deviation for the Hanford set involving the 60-m height is only slightly greater than the standard deviation of the AFCRL

TABLE 2. Ratios of Momentum Flux Between 60 m and 7.5 m, $\tau_{60}/\tau_{7.5}$.

Test Name	$\tau_{60}/\tau_{7.5}$
EB4	1.50
EB5	1.25
EB6	0.55
EB8	0.79
EB11a	negative
EB11b	0.23

near-neutral data, the extension of the constant flux layer concept to include the 60 m height for near-neutral conditions seems reasonable.

For the two stable tests (EB11) there is obviously no constant flux layer extension to 60 m. Such a result is expected since $Ri(z)$ was supercritical at $z = 60$ m.

POWER SPECTRAL CHARACTERISTICS - QUASI-NEUTRAL TESTS

The principal characteristics of the power spectral analysis may be summarized as follows:

1. For all tests the vertical component power spectrum, $nS_w(n)$, exceeds the model curve, Equation (4), by about 30%. This is not surprising since different investigators have found σ_w^2/u_*^2 to vary anywhere from 1.3⁴ to 1.8.³ The AFCRL model, Equation (4), integrates to 1.45, while the values of σ_w^2/u_*^2 obtained from the Hanford quasi-neutral data are about 1.8 at 7.5 m and 15 m. Values of $\sigma_w^2/(-\overline{u'w'})$ are given for the three heights over the four tests in Table 3.

The sample $nS_v(n)$ did not exceed the model curve, Equation (3), as conspicuously as the sample $nS_w(n)$ exceeded the model curve, Equation (4). According to theory, the relationships of the lateral and vertical component sample spectra to their respective model curves should be the same in the

TABLE 3. Values of $\sigma_w^2/(-\overline{u'w'})$ for Four Quasi-Neutral Tests.

z(m)	Test Name	EB4	EB5	EB6	EB8
60		2.4	2.2	1.9	2.1
15		1.9	1.8	1.9	2.0
7.5		1.8	1.8	1.7	1.7

inertial subrange if the model curves are of the same level, which is expected to be 4/3 the level of the longitudinal component spectrum. However, the ratios of $nS_v(n)$ and $nS_w(n)$ to $nS_u(n)$ as given by Equations (2), (3), and (4) are 1.29 to 1 and 1.22 to 1 respectively. In other words the lateral component spectrum model curve is higher than the vertical in the inertial subrange by about 6%. Some of the sample lateral component spectra from the Hanford data sample approximated the level of the concurrent longitudinal spectrum rather than the level of the vertical spectrum.

2. At low frequencies, there is considerable unparameterized departure of the spectra from the model curves, Equations (2), (3), (4), see Figures 1 and 2. Considerable low frequency energy above the model curves is shown for all three components for tests EB4 and EB5. On the other hand most of the spectra from test EB6 do not show excess low frequency energy compared to the model curves, and, in fact, the spectra at 60 m show a decided deficiency of low frequency energy. The low frequency part of the spectra for EB8 approximates the model more closely than do those from the other tests.
3. The 60-m power spectra do not depart from the constant flux layer model in the mean in any consistent way. However, the mean square departure of the 60-m spectra from the model is somewhat greater than is true for the lower levels. When the dimensionless spectra

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FIGURE 1. Dimensionless Power Spectra, Test EB4.

stratify in the inertial subrange rather than collapsing, the order is not necessarily according to

height. In other words, the 60-m spectrum may be the middle curve in such a stratification.

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FIGURE 2. Dimensionless Power Spectra, Test EB6.

POWER SPECTRAL ANALYSIS - STABLE CASE

Powell and Horst,⁵ found that the dimensionless frequency, $(f_0)_\alpha$ from the two EB11 tests averaged about 60%

of the values predicted by Kaimal.² Subsequently the power spectra from Test EB11b have been replotted with the high frequency portions corrected for averaging and aliasing. The

variances have also been corrected accordingly. The results continue to show $(f_o)_\alpha$ values consistently lower than those predicted by Kaimal. The ratios of the measured $(f_o)_\alpha$ to the predicted $(f_o)_\alpha$ are given for each component at the 7.5 m and 15 m levels in Table 4.

The measured values of f_o depend on the calculated variances. Because of the high frequency slope of $-2/3$, the measured value of $(f_o)_\alpha$ varies inversely with σ_α^3 . Therefore, if the calculated variance from the 15 m data were 0.83 of the values actually calculated, the $(f_o)_\alpha$ values at 15 m would have been in agreement with those given by Kaimal.²

f_o values were not calculated for the 60-m data since $Ri(z)$ was supercritical (about 0.65) at this height. The turbulence variances at 60 m were about half as great as those at lower levels. Furthermore the slope of the 60-m spectra at high frequencies was somewhat less than $-2/3$, possibly because of the low turbulence level associated with the supercritical Richardson number.

SUMMARY

The following three observations are made from the above analysis:

1. The Hanford vertical component power spectra are found to exceed the AFCRL modeling expression by about 30% in the mean in the in-

ertial subrange, the lateral component power spectra exceed the corresponding AFCRL modeling expression by a lesser amount, and the mean fit of the longitudinal power spectra to the corresponding AFCRL model is quite good in the inertial subrange. The stratification width within the inertial subrange of different sample power spectra modeled by the same curve is usually within a factor of 1.5. On the other hand the departure at low frequencies is much greater; in some cases the mean spectral level may depart from the model curve by an order of magnitude. Such departures are greater for the horizontal component spectra than for the vertical component. These results indicate the allowance that must be made in individual cases for uncertainty in predicting diffusion concentrations from empirically or spectrally derived dispersion parameters.

2. The extension of constant flux layer modeling up to 60 m follows from analysis of the Hanford data since no systematic departure of the 60-m data suggesting the need for additional parameterization is observed.
3. The dimensionless frequency determinations, $(f_o)_\alpha$, from the Hanford data average out to about $2/3$ the values predicted by AFCRL literature on the power spectra for stable conditions.

TABLE 4. Measured and Predicted Values of the Dimensionless Frequency $(f_o)_\alpha$.

z = 7.5 m	measured	predicted	ratio
u	0.035	0.063	0.56
v	0.11	0.19	0.57
w	0.21	0.35	0.60
z = 15 m			
u	0.06	0.08	0.75
v	0.18	0.24	0.75
w	0.35	0.45	0.78

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ATMOSPHERIC TRANSFORMATION PROCESSES

During recent years the PNL study of the transformation of atmospheric trace constituents has concentrated primarily on developing and using an airborne measurement facility. Progress in the past year has included developing a precision calibrations laboratory for the analytical instrumentation, acquiring an aircraft large enough to carry the equipment needed to measure aerosols plus precursor gases, and conducting field experiments in St. Louis and elsewhere to measure transformation processes.

The program has also progressed in two additional areas. The first of these is in the incorporation of solar radiation aspects, regarding both the effects of pollutants on the radiation, and the influence of radiation on the pollutants. The second additional area is that of transformation modeling. In the past year new diagnostic numerical models have been developed to help interpret data acquired in this program. These aspects will be described in the following contributions.

- **ATMOSPHERIC AEROSOLS AND TRACE GASES**
- **TRACER STUDIES IN THE METROMEX EXPERIMENT**
- **BATTELLE MEMORIAL INSTITUTE PHYSICAL SCIENCES PROGRAM**

EVOLUTION OF THE ST. LOUIS AEROSOL SIZE DISTRIBUTION

A. J. Alkezweeny

Time changes of the particle size distribution in a parcel of air, moving over the St. Louis area, were measured. The air parcel trajectory has traced with a tetroon. A pronounced change in the particle size distribution was observed. Gas to particle transformation involving existing nuclei probably contributed greatly to the observed changes.

INTRODUCTION

In this age of expanding industry, it is becoming increasingly important to understand the behavior and implications of released airborne material. The modifications which occur in the atmosphere between the point of release and the receptor can markedly influence the impact on man. These modifications occur through chemical reaction of trace gases, reaction of gases with particulates, and/or coagulation of aerosol particles. The importance of these processes depends on the physical and chemical characteristics of the material emitted from the sources, and the meteorological conditions. These processes also play an important role in the wet or dry removal of material from the atmosphere. Therefore, any complete modeling effort aimed at predicting the contribution of a source to areas downwind of it, should be cognizant of these processes.

Adverse effects of emitted material are often linked to atmospheric transformation processes. For example, the sulfur dioxide levels usually characteristic of polluted atmospheres provide only a mild respiratory irritant; however, sulfuric acid mist is more irritating to the respiratory system.^{1,2} Reduction of solar radiation caused by suspended

aerosol particles may be enhanced as a result of particle coagulation and chemical reaction. Gas to particle transformation plays an important role in the removal of trace gas in the atmosphere. This transformation is believed to be a principal step in scavenging of sulfur and nitrogen compounds.³

The objective of this experiment is to provide an input to a mathematical air pollution model which is currently under development,^{4,5} and to identify the significant physical and chemical processes that occur in urban plumes. The first phase of this project was conducted in St. Louis as part of the 1974 METROMEX program. In this report, a brief description of the experiment and a summary of the results are presented and discussed. Details of the aerosol and trace gases program will be reported later.⁶

EXPERIMENT

This experiment has involved the measurements of aerosol particle size distributions, in the range of 0.003 μm to 5 μm particle diameter, as a function of time in a parcel of air over the St. Louis area, using an instrumented Cessna 411 aircraft.⁷ Size distribution of particles up to 1.0 μm in diameter were measured with

an airborne Whitby Electrical Aerosol Analyzer and those from $0.3\text{ }\mu\text{m}$ to $5\text{ }\mu\text{m}$ particle diameter were obtained with a Royco Optical Sensor Model 220, a pulse converter, pulse height analyzer, and a printer. A General Electric particle counter was also used to measure the concentration of Aitken nuclei. Since the Whitby Electric Aerosol Analyzer requires about two minutes to scan through the total particle-size distribution, an air sample was captured into a 2-ft^3 plastic bag, and subsequently sampled by the instrument.

In order to follow the time evolution of the particle size distribution in a given air particle, a tetroon was released to a constant altitude, and its path was intersected by the aircraft at different times, while the measurements were performed simultaneously. A total of three successful flights were made during July and August, 1974. The meteorological conditions during the three flight days are as follows:

July 17: High pressure centered over Pennsylvania at 0600 CST gave light southeasterly flow and clear skies over St. Louis (STL). The nearest weather and frontal action was some 350 miles to the northwest, though there was a weak stationary front 250 miles to the south. Visibility at St. Louis was restricted by haze during the day indicating a rather stagnant air mass. At 850 mb, the high pressure was centered over Virginia and North Carolina. At 3000 ft, wind over STL at 0600 was estimated to be southerly, becoming more southwesterly with height. By 1800 CST, the 850 mb high was centered over STL.

August 21: Clear skies, associated with a large high pressure cell centered off the coast of Maine, extended over east central U.S. from the Gulf of Mexico to the Great Lakes. At 0600 CST, St. Louis reported clear skies, light southeasterly winds and ground fog. No frontal activity or weather was nearer than a weak frontal system and squall line 270 miles northwest at 0600 CST. Visibility restriction due to haze was reported during the day after the ground fog dissipated. High clouds appeared in the afternoon as the aforementioned front drifted southeasterly. The 850

mb chart for 0600 CST indicates light southerly flow over the STL region. This flow had become more westerly by 1800 CST.

RESULTS AND DISCUSSION

The first flight was made on July 17, at 1330. A tetroon was released from Granite City to an altitude of 3000 ft msl. The aircraft tracked the tetroon for about 1 hr until it lost altitude and was abandoned. Two measurements were made during the course of the tetroon tracking, one above the release site, and the second $1/2$ hr later. The tetroon velocity was estimated at 11.5 miles/hr. Using data from a series of pilot balloons and from the tetroon trajectory, a continuing path for the air parcel was predicted, and the aircraft continued sampling along the predicted path. Figure 1 shows the resulting particle surface distributions measured at different times. The large increase in the particle distribution between the 1500 and 1700 hr was caused by the entrainment of material when the air parcel passed over the Alton area, which contains several oil refineries and other industry. Beyond this point, no visible source is present, therefore, the change in the particle distribution reflects the result of material interaction within the parcel. Figure 2 shows the same data plotted in terms of particle number distribution.

The August 20 flight was concentrated on determining the particle distribution as a function of altitude over St. Louis up to 5000 ft msl, at 1400 hr local time. The temperature inversion height was located between 4000 and 5000 ft msl. The total particle count, as measured by the Aitken Particle Counter dropped from about 2000 particle/cm³ to less than 200 particle/cm³. The change is also reflected in the particle distribution measurements (Figure 3). The reduction of particle concentration across the temperature inversion was also observed by the author over Seattle, Washington.⁸

The last flight was made on August 21 about noon. The sky was clear and the wind approximately from the south-southeast at about 10 mph at 3000 ft msl. A tetroon was released from Alton Civic Airport to this altitude.

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FIGURE 1. Time Changes of the Particle Surface Distribution for July 17 Flight.

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FIGURE 3. Particle Surface Distributions over St. Louis Measured on August 20.

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FIGURE 2. Time Changes of the Particle Number Distribution for July 17 Flight.

The top of the plume was visible from the aircraft, and estimated to be between 4000 to 5000 ft msl. Four measurements were made every 1/2 hr starting over the airport. The results of this flight are plotted in terms of particle surface distributions and number distributions in Figure 4 and Figure 5 respectively. The figures clearly demonstrate the growth of aerosol particles. The Aitken nuclei concentrations during this period were 2×10^4 , 1.5×10^4 , 10^4 , and 8×10^3 /cm³ for the time 1200, 1230, 1300, and 1330 hr respectively. The concentrations of particles below about 0.06 μm decreased in time and the concentrations of larger particles increased. The mass concentration of particles below 1 μm seems to increase in time, which indicates that particle growth by chemical condensation is larger than growth by coagulation.

CONCLUSION

Particle size distribution was measured in the range of 0.003 μm to 5 μm particle diameter, as a function of time in a parcel of air over St. Louis, using an instrumented Cessna 411 aircraft and a tetron to race the air

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FIGURE 4. Time Changes of the Particle Surface Distribution for August 21 Flight.

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FIGURE 5. Time Changes of the Particle Number Distribution for August 21 Flight.

parcel trajectory. Pronounced changes in the particle size distribution were observed. The concentration of particles below about $0.06 \mu\text{m}$ decreased in time, and the concentrations of larger particles increased. Furthermore, the mass concentration increased with time. These findings suggest that gas to particle transformation involving existing nuclei plays an important role in the growth of particles in the St. Louis plume. Further experiments are planned which include the time changes in the concentrations of reactive trace gases, and to identify mechanisms for the

decay and growth of atmospheric particles.

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CONCENTRATIONS AND RATES OF REMOVAL OF CONTAMINANTS
FROM THE ATMOSPHERE IN AND DOWNWIND OF ST. LOUIS

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The Metropolitan Meteorological Experiment (METROMEX) is a study by several organizations of inadvertent weather modification, particularly precipitation changes, caused by urban-industrial effects in the St. Louis metropolitan area. Two main experiments are being conducted by the Radiological Chemistry Section during METROMEX. One of these is the measurement of trace elements, sulfates, SO_2 , Freons, and CO upwind, in, and downwind of St. Louis to determine the decrease in the atmospheric concentrations downwind of the city that result from atmospheric mixing, dry deposition, and, in the case of SO_2 , chemical reactions. The concentrations of anthropogenic trace elements such as lead and bromine generally decreased rapidly between city center and 20 miles downwind of the city because of vertical mixing, but then decreased slowly with distance beyond that. The concentrations of SO_2 , however, decreased much more rapidly downwind of the city due to conversion to sulfates and deposition on vegetation and the earth's surface. The half-residence time of SO_2 in the atmosphere was calculated to be 4.5 hr. The sulfate concentrations increased downwind of the city due to the conversion of SO_2 to sulfates, but the increasing sulfate concentrations accounted for only about 10% of the decrease in the SO_2 concentrations.

INTRODUCTION

The St. Louis metropolitan area generates large quantities of pollutant materials, some of them toxic, into the atmosphere. These contaminants are carried large distances by the prevailing winds and may affect regions considerably downwind of St. Louis. The downwind decreases in the atmospheric concentrations of these contaminants with time and distance depend upon atmospheric mixing processes, wet and dry deposition on the earth's surface, and in some cases, chemical conversion processes. The relative importance of mixing processes versus removal processes in decreasing the contaminant concentrations downwind of St. Louis may be

determined by comparing the concentrations of nonconservative contaminants with the concentrations of conservative contaminants. Freon-11, which is released into the atmosphere primarily by aerosol spray cans, is essentially conservative, having a residence time in the atmosphere of several years.

EXPERIMENT

The concentrations of trace elements, sulfates, SO_2 , CO, and Freons are being measured in air samples collected upwind, in, and downwind of St. Louis during July of 1973 and 1974 to determine the atmospheric concentrations of these contaminants produced by St. Louis, and to determine

the rates at which the concentrations are decreased downwind by atmospheric mixing processes, dry deposition on the earth's surface, and (in the case of SO_2), chemical reactions.

In 1973 the aerosol samples were collected using HiVol pumps to draw air through 5 cm diameter IPC filter papers at flow rates of about $1.7 \text{ m}^3/\text{min}$. These samples underwent x-ray fluorescence and neutron activation analysis for trace elements. In 1974 11 cm diameter Whatman-41 filter paper was employed at a flow rate of about $0.6 \text{ m}^3/\text{min}$. Samples were collected for SO_2 analysis by bubbling air through sodium tetrochloromercurate solutions at rates of $1 \text{ l}/\text{min}$. Evacuated 460 cm^3 capacity gas cylinders were used to collect whole air samples for analysis for CO and Freons in 1973. However, the results of the Freon and CO analyses were fairly erratic, probably because of the very short sampling period, so in 1974 a sampler was used which collects a whole air sample in 1 or 2 hr by slowly drawing a piston up a cylinder.

In 1973, air samples were collected on six afternoons upwind, in, and on three arcs downwind of St. Louis. These were nonconvective days in which the mixing layer was capped by an inversion. The three sampling arcs were set up 20, 40, and 60 miles downwind of St. Louis on four of the six sampling days; 20, 40, and 80 miles downwind on one day; and 20, 50 and 80 miles downwind on the other day. The sampling arcs were set up by five people equipped with a completely mobile sampling apparatus. On each day, each arc consisted of five more or less equidistant sampling locations. These were arranged on a 30-to 50-mile line perpendicular to the predicted wind 20 miles downwind of the St. Louis arch. On three of the days vertical profiles were taken using the Battelle Cessna-411 aircraft. Contaminant concentrations as functions of particle size are being determined from Lundgren impactor samples collected upwind of St. Louis, on the roof of the downtown Federal Building, and at two other sites in the St. Louis area.

In 1974 the sampling procedure was changed somewhat. On each sampling day at least one set of air samples was collected upwind of St. Louis,

sequential samples were collected on the roof of the Federal Building, and two to four sequential samples were collected at stationary sites on arcs 20 to 100 miles downwind of St. Louis.

RESULTS

On July 16 and 17, 1973, the wind was nearly easterly and the atmospheric contaminant concentrations were very similar. Therefore, the atmospheric concentrations on these 2 days were plotted together on a coordinate system using the average wind direction measured on each day at Lambert Field, St. Louis as the X axis. Isopleths of concentration were then drawn by computer. These isopleths were then superimposed on a map of the St. Louis area with the X axis aligned in the east-west direction. On July 22, 27, and 28, 1973, the wind was nearly westerly and the measured atmospheric concentrations were similar, so the same procedure was followed to obtain isopleths of concentration for these days.

A. Lead and Bromine

Lead and bromine are released into the atmosphere in urban areas primarily by automobile exhaust. The lead concentrations for July 16 and 17 and for July 22, 27, and 28 are plotted in Figures 1 and 2, respectively. The bromine concentration isopleths are similar to those of lead and are not shown. The lead and bromine concentrations at the Federal Building in downtown St. Louis averaged 20 times the upwind concentrations. The concentrations decreased very rapidly between downtown St. Louis and the 20-mile arc. This large decrease probably resulted from dilution caused by vertical mixing through the mixing layer, since particle size analysis using Lundgren impactors indicates that only a small fraction of the lead and bromine would be expected to settle out in this distance, and because the decrease in concentration farther downwind of St. Louis was much smaller. Seventy percent of the lead and bromine was present on particles smaller than 0.3μ and only 10% was present on particles larger than 3μ . The lead and bromine concentrations at the outer arc averaged 68% and 74%, respectively, of the concentrations at the 20 mile arc.

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FIGURE 1. Lead Concentration in 10^{-7} g M^{-3} on July 16 and 17, 1973.

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FIGURE 2. Lead Concentration in 10^{-7} g M^{-3} on July 22, July 27 and 28, 1973.

On July 16 and 17, 1973, samples were collected by aircraft on approximately 30-mile north-south flight tracks above the 20- and 40-mile arcs. At the 20-mile arc the lead concentrations averaged $2.1 \times 10^{-7} \text{ g/m}^3$ in the ground level samples collected below the flight track, $4.1 \times 10^{-7} \text{ g/m}^3$ at 1000 ft, $2.7 \times 10^{-7} \text{ g/m}^3$ at 2000 ft, and $1.4 \times 10^{-7} \text{ g/m}^3$ at 3000 ft. At the 40-mile arc the lead con-

centration averaged $1.9 \times 10^{-7} \text{ g/m}^3$ at ground level and $1.8 \times 10^{-7} \text{ g/m}^3$ at 2000 ft. The bromine concentrations at the 20-mile arc averaged $3.0 \times 10^{-8} \text{ g/m}^3$ at ground level, $8.7 \times 10^{-8} \text{ g/m}^3$ at 1000 ft, $2.4 \times 10^{-8} \text{ g/m}^3$ at 2000 ft, and $4.4 \times 10^{-8} \text{ g/m}^3$ at 3000 ft. On July 22, 1973, the lead concentrations at the 20-mile arc east of St. Louis averaged $1.6 \times 10^{-7} \text{ g/m}^3$ at ground level below the aircraft flight track, $2.2 \times 10^{-7} \text{ g/m}^3$ at 1000 ft, and only $0.6 \times 10^{-7} \text{ g/m}^3$ at 1500 ft. However, the 1500-ft concentration increased to $2.4 \times 10^{-7} \text{ g/m}^3$ at the 40-mile arc and then decreased to $1.6 \times 10^{-7} \text{ g/m}^3$ at the 60-mile arc. The ground level concentration below the flight track averaged $1.8 \times 10^{-7} \text{ g/m}^3$ and $1.3 \times 10^{-7} \text{ g/m}^3$ at the 40- and 60-mile arcs, respectively.

The 1974 ground level lead and bromine measurements show the same pattern of a rapid decrease in concentration between downtown St. Louis and a much slower decrease thereafter. On July 23, 1974, the lead concentration decreased only 9% between the 20-mile arc and the 90-mile arc. The average standard deviation between sequential samples collected at a given location downwind of St. Louis was 35%.

B. Sulfur Dioxide and Selenium

The SO_2 concentrations on July 16 and 17 and on July 22, 27, and 28 are plotted in Figures 3 and 4, respectively. The major SO_2 emission sources are also shown in the figures. Nearby sources have been combined to give their total emission. The downtown SO_2 concentrations averaged 20 times the upwind concentrations. However, the maximum concentrations were observed at the 20-mile arc because the major SO_2 sources were mostly north of the downtown sampling site. It is very probable that the maximum concentrations actually occurred closer to St. Louis than the 20-mile arc. However, no samples were collected between the downtown site and the 20-mile arc. Therefore, the actual shape of the computer-drawn contours between the downtown site and the 20-mile arc should not be taken seriously. The SO_2 concentrations decreased much more rapidly between the 20-mile arc and the outer arcs than did the concentrations of the other contaminants measured. The SO_2 concentration at the outer arc averaged only 27% of the 20-mile arc

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FIGURE 3. Sulfur Dioxide Concentrations in 10^{-5} g M^{-3} on July 16 and 17, 1973.

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FIGURE 4. Sulfur Dioxide Concentrations in 10^{-5} g M^{-3} on July 22, 27 and 28, 1973.

concentration. This relatively large decrease in SO_2 concentration was probably due to the conversion of SO_2 to sulfates in the atmosphere and to the reaction of SO_2 with vegetation. The half-residence time for SO_2 in the atmosphere was calculated to be 4.5 hr.

Selenium is generally emitted into the atmosphere by the same sources that emit SO_2 , so the comparison of the SO_2 and selenium concentration decreases between the 20-mile arc and the outer arcs is especially useful in demonstrating that SO_2 is removed from the atmosphere more rapidly than particulate materials. The selenium concentrations on July 16 and 17, 1973, are shown in Figure 5. The selenium concentrations decreased very slightly between the 20-mile arc and the outer arc.

The SO_2 concentrations in 1974 varied with distance downwind of St. Louis in much the same manner as in 1973. The average standard deviation between sequential samples collected at a given location downwind of St. Louis was 70%, which is double that of lead, probably because of the discrete nature of the SO_2 sources. The SO_2 concentrations in downtown St. Louis were considerably higher in

1974, although it is difficult to compare the 2 years because of the differing wind directions.

C. Sulfates

The sulfate concentrations in downtown St. Louis averaged only a few percent higher than the upwind concentrations, probably because it takes an appreciable period of time for the SO_2 produced by the St. Louis metropolitan area to be converted to sulfate (Figures 6 and 7). Eighty-five percent of the sulfate in downtown St. Louis was present on particles smaller than 0.3μ and only 2% was present on particles larger than 3μ . The sulfate concentrations increased with distance, with maximum concentrations being observed at the 60 mile arc, indicating a continual conversion of SO_2 to sulfates. On July 27 and 28 the sulfate concentrations decreased between the 60- and 80-mile arcs. The increase in sulfate concentration between the 20-mile arcs and the 60-mile arcs was only about 10% of that necessary to account for the decrease in the SO_2 concentration, even if it is assumed that the sulfate concentration was decreased by 30% by dilution and deposition, as was the case for lead and bromine. Therefore, the large decrease in the

FIGURE 5. Selenium Concentrations in 10^{-9} g M^{-3} on July 16 and 17, 1973.

SO₂ concentration with distance downwind of St. Louis was probably due primarily to the reaction of SO₂ with vegetation.

D. Freon-11

The Freon-11 concentration at the 20-mile arc averaged 60% of the down-

town concentration in 1973. Freon-11 is released primarily in the suburbs, while lead and bromine are produced in large amounts by downtown traffic. This explains why the decrease in Freon-11 concentration between downtown St. Louis and the 20-mile arc was considerably less than the decrease in the lead and bromine concentrations, although deposition of lead and bromine on the earth's surface by gravitational settling and turbulent diffusion certainly contributed to the decreases in their concentrations. The Freon-11 concentrations at the outer arc (60 or 80 miles) averaged 90% of concentrations at the 20-mile arc. Comparison of the decreases in lead, bromine, and Freon-11 concentrations between the 20-mile arc and the outer arc indicate that dilution due to atmospheric mixing was responsible for approximately a 10% concentration decrease, while deposition on the earth's surface caused approximately a 20% decrease in the lead and bromine concentrations.

The measured Freon-11 concentrations were much more consistent in 1974 than in 1973, probably because the samples were collected over periods of an hour or two, rather than a few seconds. However, the standard deviation between sequential samples still averaged 60%.

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FIGURE 6. Sulfur Concentration in 10^{-6} g M^{-3} on July 16 and 17, 1973.

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FIGURE 7. Sulfur Concentration in 10^{-6} g M^{-3} on July 22, 27 and 28, 1973.

E. Other Trace Elements

Arsenic, bromine, chlorine, copper, mercury, lead, sulfur, antimony, selenium, zinc, and possibly magnesium are the trace elements which were greatly enriched relative to iron in St. Louis air, indicating that they were primarily of anthropogenic origin. Elements of anthropogenic origin can also often be identified by their particle size distribution, since they are generally present on smaller particles. Calcium and iron are primarily of crustal origin, and 50% of the calcium and 45% of the iron was present on particles larger than $10\ \mu$, while only 7% of the calcium and 14% of the iron was present on particles smaller than $0.3\ \mu$. On the other hand, only 11% of the anthropogenic element copper was present on particles larger than $10\ \mu$, while 62% was present on particles smaller than $0.3\ \mu$. Titanium and vanadium are two elements which did not appear to be greatly enriched relative to iron in St. Louis air, but whose particle size distribution indicated a large anthropogenic contribution. Twenty-two percent of the titanium and vanadium were present on particles larger than $10\ \mu$, while 53% and 42%, respectively, were present on particles smaller than $0.3\ \mu$.

Isopleths of concentration have been drawn for all of the anthropogenic elements and for iron and lanthanum for July 16 and 17, 1973. The contours of antimony, arsenic, chlorine, chromium, mercury, and zinc concentration are very similar to those of lead and bromine, except that the decreases in the antimony and zinc concentrations between the Federal Building in downtown St. Louis and the 20-mile arc were much smaller than for the other elements. The copper concentrations at the 20-mile arc were somewhat higher than in downtown St. Louis on both July 16 and 17 and July 22, 27, and 28.

Also, the peak concentrations occurred downwind of Alton rather than St. Louis. The copper concentrations decreased with distance beyond the 20 mile arc at about the same rate as did the other trace elements. However, the copper concentrations should be viewed with suspicion since the samples could have been contaminated by copper given off by the electric motors of the HiVol samplers, even though the filters were separated from the HiVols by 10 ft of pipe. The concentrations of lanthanum, which should be primarily of crustal origin, on July 16 and 17 are shown in Figure 8. The lanthanum concentration varied much more randomly than did the concentrations of the other elements, although the lanthanum concentration did appear to be somewhat higher in the St. Louis plume.

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FIGURE 8. Lanthanum Concentrations in 10^{-9} g M^{-3} on July 16 and 17, 1973.

A USER-ORIENTED COMPUTER CODE FOR SOLUTION
OF THE GENERAL EQUATIONS OF CONTINUITY FOR
TRACE SUBSTANCES IN THE ATMOSPHERE

J. M. Hales

This report describes a new general-purpose computer code for obtaining finite difference approximations to solutions of the continuity equations for trace atmospheric constituents. This code is intended for use in diagnosis of field data, particularly in the Atmospheric Aerosols and Trace Gases program.

The advent of rapid, large computers has been accompanied by a host of special purpose algorithms for approximating solutions to the equations of continuity for trace substances in the atmosphere. In general, however, these algorithms have been composed to fit the specific requirements of individual research efforts, or have been dedicated to computer systems too large or specialized to permit widespread usage throughout the scientific community. At the Battelle-Northwest Laboratories there has been a need for such a general purpose code for use as a diagnostic tool to interpret data from a variety of field experiments. This report describes an algorithm that has been developed during the past year to fulfill this need.

Named PROTEUS, this code obtains finite-difference approximations (FDA's) to solutions of continuity equations, for trace substances mixing and interacting within the atmosphere. The system is written in standard FORTRAN IV language in a highly user-oriented layout, and allows FDA's to be obtained on computers that are generally available to today's researchers. The important limitations of the code are:

- turbulent mixing processes are treated in terms of effective diffusion parameters; i.e., the "K-theory" assumption is made,

- the system is assumed to be in a steady state (or at least in a quasisteady state),
- longitudinal diffusion is assumed to be unimportant.

The first assumption is necessary because of the lack of a reasonable alternative--at least with the present state of development of fluid-mechanics theory and computational hardware. The two other assumptions were made because they each allow an order-of-magnitude reduction in computation-hardware demands. These two stipulations could be removed fairly easily, rendering the code more general than it exists in its present state. Such an action, however, would necessitate the use of computers sufficiently large to detract severely from the utility of the code for its intended purposes. In view of these assumptions it is emphasized that the code is of little use where the second or third assumptions become invalid. These assumptions are expected to be fulfilled for many potential applications of the code; good meteorological judgement, however, should be exercised with each application to ensure that such conditions do indeed exist.

Aside from the above features PROTEUS is extremely general and flexible. The governing equations and boundary conditions are given as follows:

$$\begin{aligned}
& V_x \frac{\partial C_A}{\partial x} + V_y \frac{\partial C_A}{\partial y} + V_z \frac{\partial C_A}{\partial z} \\
& = \frac{\partial K_y}{\partial y} \frac{\partial C_A}{\partial y} + \frac{\partial K_z}{\partial z} \frac{\partial C_A}{\partial z} + K_y \frac{\partial^2 C_A}{\partial y^2} \\
& + K_z \frac{\partial^2 C_A}{\partial z^2} + \text{Gen (A)} - V_s \frac{\partial C_A}{\partial y} \\
& - C_A \frac{\partial V_s}{\partial z} \quad (1)
\end{aligned}$$

$$\begin{aligned}
C_A &= C_A \Big|_{\text{background}} \quad \text{at } y = \pm \infty \\
&\quad \text{and at } z = \infty \quad (2)
\end{aligned}$$

$$\begin{aligned}
f(C_A) &= V_s(0) C_A(0) \\
&- K_z(x, y, 0) \frac{\partial C}{\partial z} \Big|_0 \quad \text{at } z = 0 \quad (3)
\end{aligned}$$

for component A. Similar equations apply for components B, C, ..., N, where N is the total number of components in the system. Nomenclature in equations 1 through 3 is consistent with that used elsewhere in the literature, the C's denoting concentration and the V's representing components of the wind field (except for V_s , which is a settling velocity for the trace component in question). All volumetric processes are combined in the generation term, Gen. This term represents the generation rate of the trace material by chemical reaction, source terms, gas-aerosol or aerosol-aerosol interactions, or precipitation scavenging. The term f in equation 3 is simply a generalized deposition flux, which may be varied at the user's demand.

From the above equations it may be seen that the code provides for the following situations:

- a general wind field in all directions
- a general diffusivity field
- generalized chemical reaction kinetics for all reactant species
- generalized aerosol coagulation, condensation and settling
- generalized treatment of precipitation scavenging
- generalized dry-deposition--resuspension behavior.

These generalized features are realized in the code by the interrogation of user-written subroutines which describe the desired transport and rate properties. To enhance its user orientation, the code incorporates the feature of default-mode operation. In total default mode simplified properties are supplied within the main program, and the executed code generates a FDA corresponding to the Pasquill-Gifford bivariate-normal equation. Starting from this point the user can proceed to elaborate his model by adding subroutines in a "building-block" fashion.

PROTEUS is currently being tested by comparing a variety of solutions for special cases where analytical solutions are available. The first intended application of the code is in conjunction with the Aerosols and Trace Gases program for interpretation of aircraft data on aerosol-transformation processes. It is intended that the code be fully documented and published in the near future in a BNWL report.

AN ALGORITHM FOR COMPUTING AEROSOL GENESIS
AND TRANSFORMATION IN THE ATMOSPHERE

R. L. Drake

This report describes research progress in developing a submodel to describe aerosol transformations for incorporation with the overall solution of the system of atmospheric plume equations. This submodel is expected to be used extensively in the Aerosols and Trace Gases Program.

INTRODUCTION

For the requirements of the Aerosols and Trace Gases Program, the solutions of the continuity equations described in the preceding report must involve valid microphysical description of aerosol formation and transformation processes. The research described in this report is devoted to development of an appropriate aerosol transformations module filling this need. The aerosol genesis and transformation processes are incorporated in the PROTEUS program via the generation terms, GEN in the trace-component continuity equations. As currently given in the PROTEUS program, the general equation of continuity for trace substances in the atmosphere can be interpreted either as a three-dimensional, steady-state model in an Eulerian frame of reference, or as a time-varying, two-dimensional model in a Lagrangian frame of reference. For the latter interpretation, the frame of reference is being advected downstream with the mean flow. In the following discussion, we will initially assume the system is three-dimensional and time-varying.

GENERAL CONTINUITY EQUATIONS

In general, an urban plume or a plume from a large industrial plant or power plant will contain many interacting species of aerosols and trace gases. The conservation of mass equations governing this complex set of reactions may take the following forms:

Conservation of the i^{th} aerosol specie

$$\begin{aligned} \frac{\partial n_i}{\partial t} + \vec{v} \cdot \vec{v} n_i - \frac{\partial V_{si} n_i}{\partial z} = & T(n_i) \\ & + \left\{ - \frac{\partial G_i n_i}{\partial v} + C(n_i) + B(n_i) \right. \\ & + \sum_{\substack{j=1 \\ i \neq j}}^I R_{ij}(n_i, n_j) \\ & \left. + S_{Hi} + S_{pi} - S_i \right\}, \end{aligned} \quad (1)$$

Conservation of the j^{th} gaseous specie undergoing heterogeneous nucleation

$$\begin{aligned} \frac{\partial C_j}{\partial t} + \vec{v} \cdot \vec{v} C_j = & T_{Hj} + \left\{ \sum_r \dot{s}_{jr} \right. \\ & \left. + \sum_p \dot{s}_{jp} - \sum_r \dot{s}_{jr}^1 - R_j \right\}, \end{aligned} \quad (2)$$

Conservation of the j^{th} gaseous specie undergoing homogeneous nucleation

$$\begin{aligned} \frac{\partial h_j}{\partial t} + \vec{v} \cdot \vec{v} h_j = & T_{Nj} + \left\{ \sum_q \dot{H}_{jq} + \sum_p \dot{H}_{jp} \right. \\ & \left. - \sum_q \dot{H}_{jq}^1 - R_{Nj} \right\}. \end{aligned} \quad (3)$$

The independent variables in this system are assumed to be the space variables (x, y, z) , where z is the

vertical coordinate, the volume (or mass) of the particle v , and the time t . The various dependent variables in the system are the $i = 1, 2, \dots, I$ different aerosol species, and the gaseous species undergoing heterogeneous, homogeneous nucleation, or no nucleation at all. The quantity $n_i = n_i(v, x, y, z, t)$ is the number density of the i th aerosol species, $C_j = C_j(x, y, z, t)$ is the mass concentration of the j th chemical species undergoing heterogeneous nucleation, and $h_j = h_j(x, y, z, t)$ is the j th chemical species undergoing homogeneous nucleation

Currently, the PROTEUS program assumes that the ambient flow field $V = (V_x, V_y, V_z)$ is determined from meteorological measurements or is postulated by the user. The settling velocities V_{si} in Equation (1) are specified and depend upon the volume or mass of the particles, type and shape of the particles and the vertical location. The turbulent flux terms, $T(n_i)$, $T(h_j)$ and $T(N_j)$, are based on some type of K-theory formulation.

The $\{-\}$ bracketed terms in Equation (1), (2) and (3) are analogous to the GEN term in the governing equation in the PROTEUS program. The terms in these brackets are defined as follows:

- G_i = Condensational growth rate,
- $C(n_i)$ = Coagulation mechanism,
- $B(n_i)$ = Breakup mechanism,
- $R_{ij}(n_i, n_j)$ = Coagulation processes between different aerosol species,
- S_{Hi} = Rate of production of particles by homogeneous nucleation of the chemical species,
- S_{pi} = Rate of input of particles from primary sources,
- S_i = Other sink processes,
- $\sum_r \dot{s}_{jr}$ = Rate of production of species C_j by the r chemical reactions,
- $\sum_p \dot{s}_{jp}$ = Rate of production of species C_j by the p primary sources,

$\sum_r \dot{s}_{jr}^1$ = Rate of removal of species C_j by the r^1 chemical reactions,

R_j = Rate of removal of species C_j by condensation onto the existing aerosol,

$\sum_q \dot{H}_{jq}$ = Rate of production of species h_j by the q chemical reactions,

$\sum_p \dot{H}_{jp}$ = Rate of production of species h_j by the p primary sources,

$\sum_q^1 \dot{H}_{jq}^1$ = Rate of removal of species h_j by the q^1 chemical reactions,

R_{Nj} = Rate of removal of species h_j by homogeneous nucleation (see the S_{Hi} term in Equation 1).

ELIMINATION OF INDEPENDENT VARIABLES

In Equation (1), we will eliminate the v -dependency by assuming that the aerosol spectra vary in a prescribed manner with respect to v . For example, if the spectra are always log-normally distributed in v , then $n_i(v, x, y, z, t)$ can be replaced by three integral power moments. The moments are defined by

$$M_{ij}(x, y, z, t) = \int_0^{\infty} v^j n_i(v, x, y, z, t) dv,$$

where $j = 0, 1, 2$. (4)

In Equations (1) to (4), another independent variable is eliminated by assuming a steady-state condition in the Eulerian framework or by neglecting the x -dependency in the Lagrangian framework. Hence, the PROTEUS program consists of three independent variables; namely, the variables (x, y, z) for the Eulerian framework, and (y, z, t) for the Lagrangian framework.

A more complete discussion of the work considered here will appear in a forthcoming BNW report.

SIMILARITY SOLUTIONS OF AN AEROSOL BALANCE
EQUATION FOR A HOMOGENEOUS MEDIUM

R. L. Drake

This paper is concerned with the search for similarity solutions of an aerosol balance equation in a homogeneous medium. The processes which contribute to the time evolution of the aerosol spectrum are coagulation, the condensation of vapors and gases on particles, and external sources and sinks of particles. Since the details of this work will occur in a forthcoming Battelle report, we only include the transformed equations and a short discussion of how they are solved; we include no solutions in this paper.

INTRODUCTION

In this work we are concerned with the time evaluation of an aerosol spectrum taking place in a homogeneous medium. The processes contributing to the time-variation of the spectrum are a particle growth mechanism due to gases or vapors condensing on the particles, and the coagulation of particles by various physical mechanisms and particle source and sink mechanisms. Thus, the system involves two independent variables, the particle volume u or v , and the time t . There is only one dependent variable, the number density or the aerosol spectrum $n(v,t)$. This last quantity is defined such that $n(v,t)dv$ is the total number of particles, whose volumes are in the interval $v \pm 1/2 dv$ at time t , per unit volume of the medium in which they are embedded. The parameters which govern the system are the coagulation parameter $\beta(u,v,t)$, the particle condensational growth rate $I(v,t)$ and the source or sink of particles $S(v,t)$. The physical units of these quantities are as follows:

$$[n] = L^{-6}, [I] = [\beta] = L^3 T^{-1}, \text{ and} \\ [S] = L^{-6} T^{-1},$$

where $[A]$ is the dimension of A , L is a characteristic length and T is a characteristic time.

Our work consists of a search for the forms of the system parameters and the initial number density $n(v,0)$ that will allow $n(v,t)$ to evolve in such a way that a similarity solution will exist. That is, we are searching for the conditions that will allow $n(v,t)$ to be replaced by a single spectrum $\psi(\eta)$, (see Figure 1). Thus, the two independent variables v and t will be "coalesced" into a single independent variable η ; and the dependent variable $n(v,t)$ will be replaced by the similarity spectrum $\psi(\eta)$.

Some of the applications and advantages of the transformed system are given in the following list.

1. The governing equation for $\psi(\eta)$ is an ordinary integrodifferential equation as opposed to a partial integrodifferential for $n(v,t)$. Therefore, if the transformed system represents a physical system of interest and numerical methods are required for obtaining solutions, the system for $\psi(\eta)$ requires considerably less computer time.
2. It is a fact that similarity solutions, or "self-preserving" spectra, have been observed both in the laboratory and in the field.^{1,3,4}

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FIGURE 1. A Schematic of a Similarity Transformation Being Applied to an Evolving Aerosol Spectrum.

3. The system for $\psi(\eta)$ is usually easier to analyze qualitatively and these qualitative results can be used in the analysis of the original spectrum $n(v, t)$.
4. Even though the transformed system may not be exactly equal to the physical system of interest, it may be sufficiently close to it that the $\psi(\eta)$'s will give meaningful upper and lower bounds for the $n(v, t)$ in question.
5. Any exact solutions of the transformed system can be used to check numerical techniques which have been devised to solve for the $n(v, t)$ in a more complex physical system.
6. Similarity solution can be used to approximate nonsimilar systems if the reason for the nonsimilarity is not too crucial.

7. From a set of similarity solutions we may be able to devise a series or asymptotic expansion for $n(v, t)$ that will be useful in solving more complex systems involving $n(v, t)$.

THE DEFINING EQUATION

The defining equation for a distribution of particles evolving in a homogeneous medium under the influence of sources and sinks, growth by condensation, and coagulation is given by

$$\frac{\partial}{\partial t} n(v, t) + \frac{\partial}{\partial v} [I(v, t) n(v, t)] = S(v, t) + \frac{1}{2} \int_0^v \tilde{F}(u, v-u, t) du - \int_0^\infty \tilde{F}(u, v, t) du \quad (1)$$

where

$$\begin{aligned} \tilde{F}(u, v, t) &= \beta(u, v, t) n(u, t) n(v, t) \\ &= \beta(v, u, t) n(v, t) n(u, t) \\ &= \tilde{F}(v, u, t) \geq 0 \end{aligned} \quad (2)$$

The second term on the left-hand side of Equation (1) is the rate of growth by condensation. The first integral on the right-hand side of Equation (1) gives the addition of particles due to smaller particles coagulating and the second integral gives the loss of v -particles due to coagulation with other particles. We have a pure coagulation process if $S \equiv I \equiv 0$; a pure condensation process if $\beta \equiv S \equiv 0$; and if $S \neq 0$, the density function $n(v, t)$ is reinforced by a source or deleted by a sink.

For both mathematical and physical reasons certain integral power moments of $n(v, t)$ are important. They are

$$N(t) = \int_0^\infty n(v, t) dv, \quad (3)$$

$$\sigma(t) = \sqrt[3]{36\pi} \int_0^\infty v^{2/3} n(v, t) dv, \quad (4)$$

$$\phi(t) = \int_0^\infty v n(v, t) dv, \quad (5)$$

where $N(t)$ is the total number concentration, $\sigma(t)$ the total surface area concentration, and $\phi(t)$ the total volume concentration, all at time t .

THE TRANSFORMED SYSTEM

The equations of transformation to be employed in this analysis were obtained from Friedlander (1964). They are

$$\eta = \frac{N(t)}{\phi(t)} v, \quad \xi = \frac{N(t)}{\phi(t)} u, \quad (6)$$

$$\psi(\eta) = \frac{\phi(t)}{N^2(t)} n(v, t). \quad (7)$$

In addition, the parameters are assumed to satisfy the following:

$$\beta(u, v, t) = \alpha(t) \beta(\xi, \eta) = \sigma(t) \beta(\eta, \xi), \quad (8)$$

$$I(v, t) = a(t) I(\eta), \quad (9)$$

$$S(v, t) = b(t) S(\eta). \quad (10)$$

The time-varying quantities $\alpha(t)$, $a(t)$ and $b(t)$ are further restricted by

$$\left\{ \begin{array}{l} A_1 = \lim_{\eta \rightarrow +0} [I(\eta) \psi(\eta)] = \text{finite}, \\ A_2 = \frac{\alpha(t)}{\alpha(t) \phi(t)} = \text{constant}, \\ A_3 = \frac{b(t) \phi(t)}{\alpha(t) N^3(t)} = \text{constant}, \\ \int_0^\infty I(\eta) \psi(\eta) d\eta = 1, \\ S_1 = 1, \\ \int_0^\infty \int_0^\infty F(\xi, \eta) d\xi d\eta = 1, \end{array} \right. \quad (11)$$

where

$$\left\{ \begin{array}{l} F(\xi, \eta) = \beta(\xi, \eta) \psi(\xi) \psi(\eta) = F(\eta, \xi), \\ S_v(\eta) = \int_\eta^\infty \xi^v S(\xi) d\xi, \quad S_v(0) = S_v. \end{array} \right. \quad (12)$$

If we make the following simplifications and notation changes

$$\left\{ \begin{array}{l} I(\eta) = (1 - A_1)\eta + f(\eta), \\ S_0(\eta) = [(1 - S_0)\eta + g(\eta)] \psi(\eta), \\ h(\eta) = f(\eta) + A_5 g(\eta), \\ N(\xi) = \int_0^\xi F(\theta, \xi - \theta) d\theta \\ \quad - 2 \int_0^\infty F(\theta, \xi) d\theta, \\ M_v(\eta) = \int_\eta^\infty \xi^v \psi(\xi) d\xi, \quad M_v(0) = M_v, \\ A_5 = A_3/A_2, \quad A_6 = A_1 + A_5 S_0, \end{array} \right. \quad (13)$$

then the governing equation for $\psi(\eta)$ is given by

$$\begin{aligned} & 2 A_2 [h(\eta) \psi(\eta) - A_6 M_0(\eta)] \\ & = \eta \psi(\eta) - M_0(\eta) - \int_\eta^\infty N(\xi) d\xi. \end{aligned} \quad (14)$$

For the pure coagulation system, Equation (14) becomes

$$\eta \psi(\eta) - M_0(\eta) = \int_\eta^\infty N(\xi) d\xi. \quad (15)$$

If there is no coagulation, the governing equation is

$$h(\eta) \psi(\eta) - A_6 M_0(\eta) = 0. \quad (16)$$

NOTES CONCERNING THE SOLUTIONS OF EQUATIONS (14), (15), (16)

A complete discussion of the properties of the solutions of Equations (14), (15) and (16), and the actual solutions themselves will be in a forthcoming BNW report and also in a paper published in the open literature. We have obtained some solutions of Equations (14) and (15) for specified values of $h(\eta)$ and $\beta(\xi, \eta)$ by the direct application of Laplace transforms. However, a more fruitful approach is to specify

$F(\xi, \eta)$ and the parameter $h(\eta)$, solve Equations (14) and (15) for $\psi(\eta)$, and then compute $\beta(\xi, \eta)$ from Equation (12). By taking different classes of functions for $F(\xi, \eta)$, a catalogue of β 's and the corresponding ψ 's can be constructed. The use and interpretation of this catalogue is given in the introduction of this report.

Equation (16) can be easily solved for $\psi(\eta)$ given the parameter $h(\eta)$.

Although they are not given in this report, we can write down the differential equations for $N(t)$ and $\phi(t)$. Given N and ϕ , we can calculate the values for $\sigma(t)$. Also, from the values of $N(t)$, $\phi(t)$ and $\psi(\eta)$, we can reconstruct the evolving spectrum $n(v, t)$ and the physical parameters $S(v, t)$, $I(v, t)$ and $\beta(u, v, t)$.

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ATMOSPHERIC AEROSOL PARTICLES IN SURFACE-LEVEL AIR

W. G. N. Slinn

An indication of some of the "structure" in aerosol particle size "spectra" is shown by plotting log area distributions of available data and drawing inferences therefrom.

Because of increased interest in air pollution and its control, in the earth's radiation budget, and in the fate of noxious trace materials released into the atmosphere, much new information has recently been obtained about atmospheric aerosol particle size distributions. These are defined and interrelations among them are shown in Table 1 where, for example, $(\Delta N/\Delta D)$ ΔD is the number of particles per unit volume whose length scale is between D to $D + \Delta D$.

The purpose of this note, following the suggestions in the important

paper by Whitby et al.,¹ is to point out some of the "structure" of the size "spectra" which can be seen when the data are plotted as log area distributions. This is illustrated in Figure 1, which was inferred from data given in the References.¹⁻⁵ Although attempts were made to accurately reflect the data, they possess such dramatic variability as a function of the variables shown that the curves should be considered only qualitative until more data is obtained. This holds especially for the "combustion" and "forest fire" curves.

TABLE 1. Interrelations Among Various Particle "Size" Distributions.

"Size" Distributions	Usual Symbols For Cumulative Distribution Functions	Usual Symbols For Distribution Functions*	Convenient Units	Alternative Symbols For Distribution Functions*	Derivation From The Number Distribution
number	N	$\Delta N/\Delta D$ **	number $\text{cm}^{-3} \mu\text{m}^{-1}$	$n(D)$	$n(D)$
log number	-	$\Delta N/\Delta \ln D$	number cm^{-3}	$n(\ln D)$	$D n(D)$
length	L	$\Delta L/\Delta D$	$\mu\text{m cm}^{-3} \mu\text{m}^{-1}$	$l(D)$	$D n(D)$
log length	-	$\Delta L/\Delta \ln D$	$\mu\text{m cm}^{-3}$	$l(\ln D)$	$D^2 n(D)$
area	S, A	$\Delta S/\Delta D$	$\mu\text{m}^2 \text{cm}^{-3} \mu\text{m}^{-1}$	$s(D)$	$\pi D^2 n(D)$
log area	-	$\Delta S/\Delta \ln D$	$\mu\text{m}^2 \text{cm}^{-3}$	$s(\ln D)$	$\pi D^3 n(D)$
volume	V	$\Delta V/\Delta D$	$\mu\text{m}^3 \text{cm}^{-3} \mu\text{m}^{-1}$	$v(D)$	$(\pi/6) D^3 n(D)$
log volume	-	$\Delta V/\Delta \ln D$	$\mu\text{m}^3 \text{cm}^{-3}$	$v(\ln D)$	$(\pi/6) D^4 n(D)$
mass	M	$\Delta M/\Delta D$	$\mu\text{g cm}^{-3} \mu\text{m}^{-1}$	$m(D)$	$\rho v(D)^\dagger$
log mass	-	$\Delta M/\Delta \ln D$	$\mu\text{g cm}^{-3}$	$m(\ln D)$	$\rho v(\ln D)$

*To be consistent with mathematical statistics these would be called density functions.

**The characteristic size of an individual particle, D , is not easily identified especially when different measurement techniques are used (e.g., light scattering, aerodynamic behavior of particles, their mobility, etc.). In theoretical studies the particles are usually treated as spheres and D is taken to be the particle diameter.

ρ is the average mass density for an individual aerosol particle.

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FIGURE 1. A Qualitative Indication of Some of the "Structure" of the Particle Size "Spectra" which is Apparent When They are Plotted as Log Area Distributions.

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WET REMOVAL PROCESSES

As a consequence of experimental and theoretical findings during the past few years, the precipitation scavenging program has concentrated on the following three areas, where additional research is now most critically needed: 1) development of a valid and reliable technique to determine rates and mechanisms for the below-cloud scavenging of submicron aerosols; 2) measurement and modeling of entrainment-scavenging processes in frontal storms; and 3) measurement and modeling of entrainment-scavenging processes in convective storms.

Progress in the first area has occurred primarily through additional modeling studies, and the design of a tentative technique for in situ washout measurement. Additional experimental data for the last two areas has been obtained during the past year from tracer-release studies conducted at Quillayute and St. Louis. The vehicle for tracer release was a light, twin-engine aircraft (Cessna 411), and owing to its load-carrying capability the possible types of investigations were substantially restricted. The recent acquisition of a larger (DC-3) aircraft, however, will allow study of a much larger spectrum of storm types, and the simultaneous measurement of cloud-physics parameters. Progress during the last year in each of the above three categories is described in the following contributions.

- **PRECIPITATION SCAVENGING**
- **RADIOACTIVE FALLOUT RATES AND MECHANISMS**
- **COOLING TOWER AND COOLING POND ATMOSPHERIC IMPACT**
- **TRACER STUDIES IN METROMEX EXPERIMENT**
- **ENVIRONMENTAL PROTECTION AGENCY SCAVENGING STUDIES**
- **BATTELLE MEMORIAL INSTITUTE PHYSICAL SCIENCES PROGRAM**

BELOW CLOUD SCAVENGING OF PARTICLES

W. G. N. Slinn

Predictive formulae for below-cloud, rain and snow scavenging of particles are presented and compared with available data.

INTRODUCTION

It is commonly accepted that precipitation can efficiently cleanse the atmosphere of suspended particles. Considerable research has been directed toward obtaining formulae to quantify this observation. This abstract outlines recent results obtained by the author dealing with the removal of aerosol particles from beneath clouds. Elsewhere in this volume the author focuses on the in-cloud scavenging problem. Details of these studies can be found in the original publications.^{1,2}

PROBLEM FORMULATION

If the number of particles per unit volume, $n(\vec{r}, t)$, at position $\vec{r}$ and time t is treated as a continuous field then n satisfies a continuity equation

$$\frac{\partial n}{\partial t} + \vec{v} \cdot \nabla n = -\nabla \cdot \vec{F} + P - R \quad (1)$$

where $\vec{v}$ is the (incompressible) air velocity; $\vec{F}$ is the (molecular) diffusional flux, which is usually negligible; P is the volume production rate and R is the rate at which particles are removed per unit volume. A time average of (1) and an approximation for the average concentration/velocity fluctuations can lead to a formulation for the mean concentration, $\bar{n}$, in terms of an effective (eddy) diffusivity. If gas-to-particle conversions and inter-particle coagulation are ignored, then $\bar{P}$ can be taken as zero. If precipita-

tion scavenging is the dominant removal process then it is easy to verify that $\bar{R} = \Lambda \bar{n}$ where

$$\Lambda(a) = \epsilon \int_0^\infty d\ell N(\ell) A E(a, \ell) v_t(\ell) \quad (2)$$

Here $N(\ell)d\ell$ is the number of hydrometeors per unit volume which are characterized by some size scale ℓ (usually radius, R or mass, m) between ℓ to $\ell + d\ell$; $A E(a, \ell)$ is the effective collision cross section between a hydrometeor of size ℓ and a particle of radius a ; $v_t(\ell)$ is the hydrometeor's terminal velocity and ϵ is the retention efficiency, the fraction of collisions that lead to particle capture.

RAIN SCAVENGING

For rain scavenging of particles then according to (2), we need the number distribution of raindrops. Figure 1 illustrates that many empirical formulae are available; indeed there are so many that the adequacy of any one formula is open to doubt. Further, experiments have shown that raindrop size distributions vary substantially even during a single rain storm. Therefore it is suggested that an approximation to (2) is not only adequate but consistent with the accuracy with which rain scavenging prediction can be made. Using the expression for the rainfall rate

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FIGURE 1. A Comparison of Some Empirical Formulae Proposed by Various Authors for Raindrop Size Distribution Functions, All Evaluated at the Same Mean-Volume Drop Diameter and Rainfall Rate.

$$J_0 = \int_0^\infty dR N(R) \frac{4}{3} \pi R^3 v_t(R) \quad (3)$$

then the rain scavenging or rainout rate, (2), is approximated by

$$\Lambda(a) = c_1 \epsilon J_0 E(a, R_m) / R_m \quad (4)$$

where c_1 is a constant $O(1)$ (the value $c_1 = 1/2$ is suggested) and R_m is the mean raindrop radius. For R_m one may wish to use Mason's expression³ for steady rains

$$R_m = 0.35 \text{ mm } (J_0 / 1 \text{ mm hr}^{-1})^{1/4}. \quad (5)$$

During the reporting period a semi-empirical expression has been suggested for the collision efficiency

for the case when the dominant physical processes contributing to collisions between particles and drops are Brownian diffusion, interception and inertial impaction. The result is shown in Figure 2. Sufficient data exists (not shown in Figure 2) to consider the diffusion and impaction expressions reliable to within about 20%. Major uncertainties remain for particles of size $0.1 \leq a \leq 1 \mu\text{m}$ as is indicated by the scatter of the laboratory data shown. This scatter is probably caused by inadequate laboratory simulation of the atmospheric problem. In the complete report¹ discussion is made of the influence during the laboratory experiments of the kinetics of induced drop circulation, particle wetting and thermophoresis. Recent comments by Beard⁷ have suggested to the author that it may be appropriate to replace the number 4 in the interception term shown in Figure 2, by the number 1. It is suggested that for most atmospheric aerosol particles the retention efficiency, ϵ , can be taken as unity.

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FIGURE 2. A Proposed Semiempirical Expression for the Particle-Drop Collision Efficiency. ν is the kinematic viscosity of air; μ_w and μ_a are the dynamic viscosity coefficients for water and air, respectively, and τ is the particle relaxation time.⁴⁻⁶

SNOW SCAVENGING

There is little data and less theory available to estimate snow scavenging. In the report,¹ approximations even more crude than the above suggest a snow scavenging rate

$$\Lambda = \Sigma = c_2 g \frac{\rho_w}{\rho_a} \frac{J_0}{v_t} \mathcal{E}(a, \ell) \quad (6)$$

where g is the acceleration of gravity; ρ_w is the density of water and ρ_a that of air; J_0 is the snowfall rate in rainwater equivalent; v_t is the terminal velocity of the snowflake type and $\mathcal{E}$ is the collection efficiency. The resulting dependence of the snow scavenging (snowout) rate on snowfall rate for $\mathcal{E} = 1$ is shown in Figure 3. Some data obtained by Engelmann et al.¹⁰ are also shown. The line for the removal of process-plant iodine by needles is obtained from (6) with $\mathcal{E} = 1.3 \times 10^{-2}$; for AgI particles, (6) is used with $\mathcal{E} = 6 \times 10^{-4}$. These $\mathcal{E}$ values as well as others and a first suggestion for $\mathcal{E}$ as a function of particle size and crystal type are shown in Figure 4.

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FIGURE 3. The Snow Scavenging Rate as a Function of Precipitation Intensity. The top curves, with $\mathcal{E} = 1$, show the influence of crystal type.¹⁰

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FIGURE 4. A Suggestion for the Particle-Snowflake Collision Efficiency. The Stokes number, S , and critical Stokes number S_* are defined in Figure 2.¹⁰⁻¹³

CONCLUSIONS

Predictive formulae for rain and snow scavenging of particles have been presented. They fairly adequately reflect available data but more experimental data is needed to test them, especially for rainout of

0.1 to 1.0 μm particles and for snowout of all particles. Future theoretical studies will concentrate on the important problems associated with variations of particle characteristics, especially for particles within industrial plumes.

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BELOW CLOUD PRECIPITATION SCAVENGING OF POLLUTANT PLUMES:
EFFECT OF NONVERTICAL PRECIPITATION ON DEPOSITION PATTERNS

M. Terry Dana

The traditional treatment of below cloud scavenging assumes that precipitation particles fall vertically, and thus the deposition reflects plume conditions directly above the deposition point. The actual deposition patterns, because the hydrometeors are wind blown, reflect plume conditions upwind; the effect is complicated by the fact that hydrometeors of different sizes see different plumes enroute to a given deposition point. Generally the deposition "plume" is narrower and more peaked than the air concentration plume. At great distances from the plume source, the cross plume integrated scavenging flux is always less than that expected from vertical precipitation.

INTRODUCTION

Coinciding with a projected increase in the use of domestic fossil-fuels for energy production is an enhanced awareness of the health and general quality-of-life impact of atmospheric emissions from fossil-and other fuel processing plants and generating stations. Of considerable concern--and an important component of the Precipitation Scavenging Program--is the effect of the interactions between pollutant plumes and precipitation, and resultant deposition to the ground and biota. This report examines the effect of non-vertical (or "wind-blown") precipitation on precipitation scavenging of material from plumes, and assesses potential errors arising from the traditional assumption of vertically falling precipitation.

A useful way to express the rate at which pollutant material is removed from a plume is through the use of a scavenging rate.* The basic definition of scavenging rate for mass of pollutant Λ_m may be stated:

$$\Lambda_m \equiv \frac{k_m}{\chi_m} \quad (1)$$

where k_m is the rate of transfer of material from the plume to precipitation ($\text{g/cm}^3 \text{ sec}$), and χ_m is the local air concentration of material (g/cm^3).* Being in units, sec^{-1} , this term expresses the fractional removal of the mass of pollutant in a unit volume per second. If the scavenging is irreversible (once transferred to the precipitation, the pollutant cannot escape to the gas phase), and if the value of Λ_m is constant throughout the plume at a given distance x_A from the source, the mass flux of pollutant to the ground at a point (x_A, y) may be expressed as

$$F(x_A, y) = \Lambda_m \int_0^\infty \chi(x_A, y, z) dz \\ = c(x_A, y) J \quad (2)$$

* Traditionally, the "washout coefficient."

* The subscript m is applied here to distinguish the quantities so subscripted from similar terms for pollutant number, area, etc.

where y is the cross-plume dimension, c is the precipitation concentration at the ground, and J is the precipitation rate.

The cross-plume integrated flux,* (mass of pollutant removed per unit distance downwind per unit time) at the distance x_A is given by

$$W = \int_{-\infty}^{\infty} c(x_A, y) J dy$$

$$= \Lambda_m \int_{-\infty}^{\infty} \chi(x_A, y, z) dz dy. \quad (3)$$

Experimental evaluation of W generally involves measurement of concentrations at evenly spaced points on the ground under the plume, and integration by summation. Since the measured rate is the mass of pollutant removed per unit ground area per unit time, W may thus be expressed as

$$W = \frac{\Sigma_m \Delta y}{A^* T}, \quad (4)$$

where Σ_m is the total mass of pollutant recovered, Δy is the distance between collectors, A^* is the area of one collector, and T is the sampling time.

The general practice for relating W to Λ_m for irreversible scavenging (and Λ_m not a function of χ) assumes that the precipitation particles fall vertically, thus allowing the integration on the right side of (3) to be performed simply for a suitable plume. If the plume is not depleted between the source and the point of interest (prior scavenging and chemical reaction, etc., insignificant), the double integral reduces to Q_0/u , where Q_0 is the source strength (g/sec) and u is the mean wind speed. Thus, the usual experimental expression:

$$\Lambda_m = \frac{u}{Q_0} W = \frac{u \Sigma_m \Delta y}{Q_0 A^*}. \quad (5)$$

* Traditionally, the "washout rate."

The assumption of vertical precipitation results in the conclusion that the mass of pollutant removed is everywhere proportional to the mass directly above the collection point, and thus that the mass deposition or concentration "plume" has the same characteristics as the plume aloft.

Because they have finite fall velocities, however, hydrometeors never fall vertically as long as there is a nonzero wind speed. A more exact determination of the relationship between the scavenging flux or concentration and the plume as encountered by the hydrometeor requires that the integration be along the trajectory of the hydrometeor.

PLUME INTEGRATION ALONG A RAINDROP PATH

The geometrical argument used for specifying a hydrometeor trajectory is illustrated by Figure 1. If it is assumed that there is no turbulence and a constant wind speed u is acting, clearly

$$\frac{V_t}{u} = \frac{z}{x_A - x}, \quad (6)$$

where V_t is the terminal velocity of fall of the hydrometeor.

The plume model used in this analysis is the Pasquill-Gifford bivariate normal plume with reflection:¹

$$\chi(x, y, z) = \frac{Q_0}{2\pi \sigma_y \sigma_z u} \exp \left(-\frac{y^2}{2\sigma_y^2} \right)$$

$$\left[\exp \left(-\frac{(z - h)^2}{2\sigma_z^2} \right) + \exp \left(-\frac{(z + h)^2}{2\sigma_z^2} \right) \right], \quad (7)$$

where σ_y and σ_z are the standard deviations in the cross-plume and vertical, and h is the source height. If the standard deviations are assumed proportional to x , ($\sigma_y = ax$, $\sigma_z = bx$) and the vertical position of the hydrometeor is related to x as in (6), the resulting form is

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FIGURE 1. Geometry of Hydrometeor Trajectory Analysis.

$$x(x, y) = \frac{Q_0}{2\pi abu} \frac{1}{x^2} \exp\left(-\frac{y^2}{2ax^2}\right)$$

$$\left\{ \exp\left(-\frac{1}{2}\left[\frac{(x_A - x)\alpha - h}{bx}\right]^2\right) + \exp\left(-\frac{1}{2}\left[\frac{(x_A - x)\alpha + h}{bx}\right]^2\right) \right\} \quad (8)$$

where $\alpha = v_t/u$.

Integration of (7) over z from $z = 0$ to $z = \infty$ implies integration of (8) over x from $x = x_A$ to $x = -\infty$, but since there is no plume where $x < 0$, the lower limit may be taken as zero. Through suitable changes of the integration variable, the hydrometeor trajectory integrated concentration x_w takes the form

$$x_w(y) = \frac{Q_0 \alpha}{2\pi abu} \int_{x_A}^{\infty} \left\{ \exp\left[-\left[A v^2 + 2Bv + Y\right]\right] + \exp\left[-\left[A' v^2 + 2B'v + Y\right]\right] \right\} dv, \quad (9)$$

where $v = x^{-1}$, and A, A', B, B' , and Y are not functions of v . The integration may be performed exactly, with the solution given by

$$x_w(y) = \frac{Q_0 \alpha}{4\sqrt{\pi} abu x_A} \left[\frac{1}{\sqrt{Y D^2 + Y}} \exp\left(-\frac{Y Y}{Y D^2 + Y}\right) \operatorname{erfc}\left(\sqrt{Y D^2 + Y}\right) - \frac{Y D}{\sqrt{Y D^2 + Y}} + \frac{1}{\sqrt{Y D'^2 + Y}} \exp\left(-\frac{Y Y}{Y D'^2 + Y}\right) \operatorname{erfc}\left(\sqrt{Y D'^2 + Y}\right) - \frac{Y D'}{\sqrt{Y D'^2 + Y}} \right] \quad (10)$$

$$\text{where } Y = \frac{\alpha^2}{2b^2}, \quad Y = \frac{y^2}{2a^2 x_A^2}, \quad D = 1 - \frac{\beta}{\alpha}$$

$$D' = 1 + \frac{\beta}{\alpha}, \quad \text{and } \beta = \frac{h}{x_A}.$$

The scavenging implications of (10) can be examined by comparing the tra-

jectory-integrated concentration with the vertically integrated concentration. The latter, for the bivariate-normal plume, is given by

$$x_V = \frac{Q_0 \exp(-Y)}{\sqrt{2\pi} \alpha u x_A} \quad (11)$$

The ratio of x_W to x_V , which, via (2) is equivalent to the precipitation concentration ratio c_W/c_V , is

$$R \equiv \frac{\alpha}{2\sqrt{2}} \exp(Y) \left[\quad \right], \quad (12)$$

where the contents of the square brackets are those of (10).

On the plume centerline ($y = 0$), the ratio reduces to

$$R_0 = \frac{1}{2} \left[\frac{1}{1 - \frac{\beta}{\alpha}} \left\{ 1 + \operatorname{erf} \left(\frac{\beta}{\sqrt{2}b} \right) \right\} + \frac{1}{1 + \frac{\beta}{\alpha}} \operatorname{erfc} \left(\frac{\beta}{\sqrt{2}b} \right) \right]. \quad (13)$$

It must be noted at this point that because the plume model employed in this analysis contains a source point singularity [$x(0,0,h) = \infty$], x_W and thus R will approach infinity for $\alpha = \beta$ and $y = 0$, that is, for hydrometeor trajectories which pass through the source point. Obviously the plume model is also not trustworthy for the near-source-point region, as unreasonably high concentrations are predicted. Thus, (10) cannot be used for hydrometeor trajectories which, enroute to the "sampling distance" x_A , pass near the source of the pollutant. There are limiting conditions, however, where the scavenging impact of nonvertical precipitation can be effectively evaluated.

LIMITING CASES

1. Trajectories Above Source Point ($\alpha > \beta$) and β Large.

In this case, x_A is relatively near the source, and the precipitation is all passing over the source point. For certain values of β and b (those for which the value of the error func-

tions in (13) are sufficiently near unity) the centerline concentration reduces to

$$R_0 \approx \frac{1}{1 - \frac{\beta}{\alpha}}. \quad (14)$$

This assumption is accurate to within approximately 1% when $\beta \geq 2.6b$. The variable b is usually interpreted as σ_y , the standard deviation of the elevation angle of the wind, and is rarely larger than 0.5. Results of calculations using (13) and $b = 0.1$ (shown in Figure 2) do not deviate significantly from the ratios predicted by (14) for all values of $\beta \gtrsim 0.1$.

2. Trajectories Under the Source Point ($\beta >> \alpha$).

At this limit, $R_0 \rightarrow 0$, except for $\beta \lesssim 0.2$. Significant deposition resulting from this plume "undercutting" only occurs at distances relatively far from the source and for β nearly equal to α , as Figure 2 shows.

3. Sampling at Great Distances from the Source ($\beta \rightarrow 0$).

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FIGURE 2. Nonvertical to Vertical Centerline Rainwater Concentration Ratio: $b = 0.1$.

This is perhaps the most important situation, as the source-point difficulties are obviated, and the predominant scavenging hydrometeors pass above the source point. The $\beta \rightarrow 0$ limit, with X_A very large compared to h , or $h = 0$ (ground-level release) to (12) is

$$R(\beta \rightarrow 0) = \frac{\alpha}{\sqrt{2}b} \exp \left(Y \left[1 - \frac{Y}{Y + Y} \right] \right) \frac{1}{\sqrt{Y + Y}} \operatorname{erfc} \left(\sqrt{Y + Y} - \frac{Y}{\sqrt{Y + Y}} \right). \quad (15)$$

Examination of (15) reveals that the concentration due to nonvertical hydrometeors is always less than or equal to that resulting from vertical precipitation. Equality ($R = 1$) occurs only at $y = 0$. This is the result of the hydrometeors "seeing" a narrower plume than that directly above the sampling point. In illustration, Table 1 lists ratios R calculated from (15) for raindrops of various sizes, and for various values of y (expressed as azimuth angle from the centerline). The terminal velocities V_t of raindrops used in these calculations were obtained from an expression² fitted to the experimental data of Gunn and Kinzer.³ Since raindrop terminal velocities approach an asymptotic limit as the

diameter increases, it is possible to determine maximum values of R . These are listed in the column marked "limit," and were calculated assuming a maximum V_t of 1000 cm/sec.

CROSS-PLUME INTEGRATED FLUXES

Several values of W , as defined by (3), were calculated by simple summation of concentrations calculated at values of y corresponding to 5° azimuth spacings. For this purpose, in addition to monodisperse raindrops, a raindrop size spectrum was used. The spectrum, divided into 20 discrete diameter intervals, was chosen to represent the prefrontal, nonconvective rainfall of the Pacific Coast. The spectrum is essentially lognormal, with geometric mean diameter of 0.04 cm and geometric standard deviation of 1.86. Scavenging calculations for the spectrum involved use of (10) to gain concentrations for each size class (see Table 1), integrating by summation over the spectrum to arrive at average concentrations for the spectrum, and then summing over y . The resulting ratios of nonvertical to vertical cross-plume integrated fluxes are listed in Table 2; similar ratios for monodisperse spectra and $\beta = 0$ are listed in Table 3.

TABLE 1. The Ratio of Nonvertical to Vertical Rain Concentration for $\beta = 0$, $\alpha = 0.15$, $b = 0.10$, $u = 500$ cm/sec.

Azimuth, ¹ Degrees	Raindrop Diameter, cm					Limit ²	Spectrum ³
	0.016	0.035	.064	.100	.185		
0	1	1	1	1	1	1	1
+ 5	0.72	0.89	0.95	0.96	0.98	0.99	0.94
+ 10	0.39	0.67	0.81	0.87	0.92	0.93	0.81
+ 15	0.22	0.46	0.64	0.74	0.82	0.85	0.67
+ 20	0.14	0.32	0.49	0.61	0.72	0.76	0.53
+ 25	0.09	0.22	0.38	0.49	0.62	0.66	0.43
+ 30	0.06	0.16	0.29	0.40	0.52	0.57	0.34

¹Measured from plume centerline.

²Ratio for maximum raindrop terminal velocity $V_t \approx 10$ m/sec.

³Lognormal, parameters in text.

TABLE 2. Ratios of Nonvertical to Vertical Cross-Plume Integrated Fluxes. $a = 0.15$, $b = 0.10$, $u = 500$ cm/sec, and a lognormal pre-frontal rain spectrum.

β	0	0.05	0.10	0.25
W_w/W_v	0.88	0.90	0.97	1.01

A wind speed of 500 cm/sec was used for the flux ratio calculations, which results in some values of β/α approaching unity for some raindrop sizes and some values of β . As noted above, (10) is not reliable for $\alpha \approx \beta$, and is unstable due to the discrete nature of the spectrum. Thus, consideration was limited to small values of β , where the raindrops considered passed well above the source point.

The cross-plume integrated scavenging flux ratios indicate that for very small β , W_w is really somewhat less than that which one would expect from vertical rainfall. And in fact, despite what size the raindrops may be, the cross-plume integrated flux ratio will always be below unity because of the limiting terminal velocities of raindrops. As β increases, however, the cross-plume integrated flux, and thus Λ_m as derived from measurement, becomes close in magnitude to the vertical expectation. The centerline concentration ratio becomes greater than unity* while the off-centerline ratios remain less. The nonvertical trajectories of raindrops (at least) thus narrow the scavenging deposition "plume" as compared to the air concentration plume above.

For α such that $\beta \approx \alpha$ for a significant portion of the raindrops

which are effective in scavenging, the peak concentration is apt to be very high; unfortunately these cannot be computed with certainty using a source-point singularity model. It is likely, however, that quite large concentrations, or "hot spots" in deposition may result at regions where depositing raindrops have passed near the pollutant source.

It must be emphasized that the hydrometeor trajectory geometry used in this analysis is a very simple one, representing a first order approximation to the real turbulent atmosphere.** It is clear as well that the bivariate normal plume model used in this analysis is inadequate to describe the air concentration near the source point.

CONCLUSIONS

An examination of the scavenging concentrations and fluxes resulting from the nonvertical trajectories of raindrops passing through pollutant plumes reveals that the actual deposition patterns due to scavenging may be significantly different from those expected on the basis of vertical rainfall. While the cross-plume integrated flux of pollutant does not appear to vary much from the vertical case, these are regions downwind of pollution sources where "hot spots," or quite large rain concentrations of

* By how much we can't say, because of the inadequacy of the plume model.

** And as such is probably a poorer approximation for the case of snowflakes, which are more susceptible to small-scale turbulent motions.

TABLE 3. Ratios of Nonvertical to Vertical Cross-Plume Integrated Fluxes. $a = 0.15$, $b = 0.10$, $u = 500$ cm/sec.

Drop Diameter cm	0.016	0.035	0.064	0.100	0.185	Limit ¹
W_w/W_v	0.59	0.80	0.88	0.92	0.95	0.96

¹For maximum $V_t \approx 1000$ cm/sec.

the pollutant may occur. In the limit of large relative sampling-distances, the deposition patterns

approach the vertical case, except that off the plume centerline, the concentrations are always reduced.

NOMENCLATURE

m: mass; ℓ : length; t: time

$\Lambda, \Lambda', B, B', D, D', Y,$	Derived forms as constants in integrations; defined in text.
Λ^*	Area of precipitation collector, ℓ^2 .
a, b	Proportionality constants in plume parameters.
c_v, c_w	Precipitation concentrations of pollutant, vertical and nonvertical trajectories respectively, m/ℓ^3 .
c	Precipitation pollutant concentration, m/ℓ^3 .
F	Precipitation pollutant flux, $m/\ell^2 t$.
h	Source height, ℓ .
J	Precipitation rate, ℓ/t .
k_m	Rate of transfer of pollutant from plume to precipitation, $m/\ell^3 t$.
Q_0	Rate of release of pollutant from source, m/t .
R	Ratio of nonvertical to vertical precipitation concentration or flux.
R_0	Plume centerline ($y = 0$) concentration ratio.
T	Time of sampling, t.
u	Mean wind speed, ℓ/t .
V_t	Terminal velocity of precipitation particle, ℓ/t .
V	Integration variable, ℓ^{-1} .
W	Cross plume integrated pollutant flux, $m/\ell t$.
x, y, z	Coordinates of system ℓ .
x_A	Downwind distance of interest for sampling, ℓ .
α	Ratio of terminal velocity of precipitation particle to wind speed.
β	Ratio of source height to sampling distance.
Δy	Precipitation collector separation, ℓ .
Λ_m	Scavenging rate ("washout coefficient"), t^{-1} .
Σm	Sum of the masses of pollutant collected cross-plume, m.
σ_y, σ_z	Standard deviations of Pasquill-Gifford plume, in y and z directions, respectively, ℓ .
x_m	Gas-phase pollutant concentration, m/ℓ^3 .
x_v, x_w	Raindrop trajectory-integrated pollutant concentration, vertical and nonvertical trajectories, respectively, m/ℓ^2 .

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PRECIPITATION SCAVENGING OF POWER PLANT EFFLUENTS:
RAINWATER CONCENTRATIONS OF SULFUR AND NITROGEN
COMPOUNDS AND EVALUATION OF RAIN SAMPLER
DESORPTION OF SO₂

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A scavenging experiment was performed at a coal-fired power plant; the primary objective was to evaluate the extent to which SO₂ may desorb from rainwater on standard rain sampler surfaces prior to chemical fixing in the collection bottles. Through a comparison of the results obtained from standard samplers and accompanying no-desorption samplers, it was found that about 20% of the SO₂ was lost from the former. This result cannot be considered applicable to all situations, but it appears from calculations and qualitative meteorological information that the 20% figure is probably near a maximum for power plant plume conditions. Sulfates and nitrogen compound concentrations were also measured in the rain; while unusually high sulfate concentrations were found near the stack, NO₂⁻, NO₃⁻, and NH₄⁺ concentrations there were near low detection limits. Mobile sequential sampling was used for determining short-term concentration fluctuations, and also for locating the plume through real-time pH measurement.

An expected increase in the number of fossil-fueled energy facilities raises concern regarding the impact of various atmospheric emissions on human health and quality of life. Of no small import is the effect of the deposition of sulfur and nitrogen compounds to the ground surface and biota via precipitation scavenging. An experiment under the auspices of the Precipitation Scavenging Program was conducted at the Centralia Steam Plant in western Washington State; this experiment was something of a follow-up to an earlier series at the same site, sponsored by the Environmental Protection Agency,¹ but several additional research objectives were pursued. These objectives, which are of importance for scavenging experimentation in general in addition to the single source scavenging problem, are listed below in order of their priority:

1. An evaluation of the potential for desorption of SO_2 from standard precipitation sampler surfaces;
2. Measurement of rainwater concentrations of nitrogen compounds -- NO_2^- , NO_3^- , NH_4^+ -- in addition to the sulfur oxides SO_2 and SO_4^{--} ;
3. A demonstration of the usefulness of sequential rain sampling to evaluate the effect of short-plume fluctuations on rainwater concentrations; and
4. An examination of the effect of effluent-stream SO_3 injection on sulfur compound washout near the source.

BACKGROUND

An earlier series of field experiments at the Centralia plant,¹ was primarily aimed at determination of scavenging concentrations and rates for sulfur compounds from the plume of a coal-fired power plant. This research was one phase of a three-year study to develop a comprehensive model for the prediction of the removal of sulfur dioxide from such plumes.^{1,2,3} Rain samples were collected on arcs of samplers at distances from 0.4 to 11 km downwind of the power plant stack. Because of

the spatial extent of the sampling array, and a limited number of field personnel available, rain samples collected over periods of less than 1-3 hours were not practical. It had been previously recognized that short-term plume fluctuations were of some significance to SO_2 washout,² so it was desirable that shorter sampling times be considered in the present study.

Computer calculations, using the SO_2 washout model EPAEC, were compared with the experimental results and found to agree quite well, with the exception that measured rain concentrations at the closest sampling line (0.4 km from the stack) were significantly lower (by factors of from 3 to 10) than those predicted. There were two proposed explanations for this discrepancy. The first of these is that the Gaussian point-source plume model employed by EPAEC is inadequate to describe the plume near the source. Since EPAEC considers the nonvertical nature of raindrops' trajectories (some of which pass very near to the stack on their way to the rain collectors) as the source point singularity of the Gaussian model, this could contribute to overestimates of rain concentrations. In addition, values of the Gaussian plume parameters are not well known for near-source conditions.

The second proposed explanation was that the predicted concentrations were reasonably accurate, but that the measurements were low because of desorption of SO_2 from raindrops on the collector surface. The standard rain collector consists of a funnel (of area 325 cm^2) attached to a plastic collector bottle. When the rainwater reaches the bottle, the SO_2 is fixed--either chemically (the bottle doped with a tetrachloromercurate solution) or by freezing (the bottle packed in dry ice)--but prior to that, the SO_2 -laden rainwater is subject to desorption if the ground-level air concentration of SO_2 is sufficiently low. The latter condition is generally met near the source of an elevated plume, and assuming that the EPAEC predictions are correct, the rainwater concentrations should be high. At greater distance from the

source, the rain concentration approaches a value which is in equilibrium with the ground-level air concentration, so the presumed sampling error has decreasing significance with distance downwind.

Since the time of the earlier experiment series at Centralia, routine operation of the plant has included injection of sulfur trioxide into the effluent gas streams just ahead of their entry into the electrostatic precipitators. This has helped overcome a tendency for inefficiency in precipitator operation when low-sulfur coal is burned. The amount of SO_3 added, about 16 ppm of the effluent stream, may have a significant effect on the chemistry of the plume, and therefore the washout properties of the compounds of interest in this study. Thus, one objective of the present study was to examine any such changes which may be attributable to the SO_3 injection procedure.

EXPERIMENT DESIGN

The collector-surface desorption effect was evaluated in the present experiment by employing, in addition to the standard (dry ice freezing type) sampler,⁴ a special sampler which would fix the SO_2 by freezing the rainfall immediately on contact. This sampler consisted of a plastic bottle, cut to form a cylindrical container. This container was housed in an insulating shell, and was charged with about one liter of liquid nitrogen (LN). In operation, most of the raindrops collected fell directly into the LN and were immediately frozen (generally forming a solid pellet, but sometimes shattering into an ice "dust"); some raindrops impacted with the wall of the container, but these also froze immediately because of the low temperature which the entire inner container reached within a short time after being charged.

A line of standard collectors was deployed at a distance of about 440 m in the normal downwind direction. (This sampling line was essentially the same as Line A of the earlier experiment series.¹ In fact, the sampling position numbers are the same, but because the generating unit operating was the unit south of that operating in the earlier series, the distance from the source was about 50 m greater.) Six of the special LN samplers were

prepared to stand alongside the standard collectors at sites estimated to be near the plume centerline. As long as the two samplers at each site sampled the same rain, the resulting rain concentration measurements could be compared to evaluate the effect of desorption from the standard collector.

Short-time rain samples were to be collected by vehicle-mounted sequential samplers. The collection area of these samplers, about 1 m^2 , was sufficient so that adequate samples for chemical analysis could be obtained in about 5 minutes in a moderate rain. It was hoped that the duration of the experiment would be such that short-time samples could be collected at a number of points along roads under the plume, and to greater distances downwind than previously attempted.

EXPERIMENT

Suitable conditions for the experiment (designated C-A) occurred on March 1, 1974. During the AM hours, a rather continuous rain fell, but the wind direction was opposite to that desired, and the speed averaged less than 100 cm/sec. After a short letup in the rainfall, the wind at stack top level shifted to the normal southerly direction and the rain resumed at a very steady rate, lasting through the sampling period 1315-1515 PST. The wind speed remained very low (averaging 141 cm/sec at stack top level for the above period) and the surface wind direction remained from the north. The plume appeared quite broad, and meandered noticeably during the run; considerable lofting of the plume was also apparent. As a result of the unusually broad plume, the deployed sampling line was not sufficient to contain the plume in terms of washout, but the sample comparison pairs were well located. The low wind speed and plume loft was fortuitous in that it ensured that the ground-level air concentration of SO_2 remained very low; this condition was helpful for the desorption effect evaluation, with one possible complication discussed below. As a result of the short rainfall duration, only limited short-time samples were collected.

At the end of the sampling period, the LN samples were carefully transferred to plastic bottles and sealed. All the rain samples were kept frozen and chemical analysis was performed

subsequently at the Richland laboratory. With the exception of H^+ , the concentrations of which were determined from standard pH measurement, all of the pollutant analyses were conducted by wet-chemical methods using Technicon Autoanalyzers. The species analyzed, the techniques used, and their sensitivity limits are listed in Table 1.

Relevant emissions data for the time of Run C-A are listed in Table 2. Unfortunately, the Centralia Steam Plant was not operating in a normal

manner on that date. Because of a coal miners' strike, stockpiled coal was being burned; the irregular burning, ostensibly because of wetness, required the use of some fuel oil. Hourly output and firing rate data, and visual observation indicated, however, that the operation was fairly uniform during the sampling period. Another fortuitous development was that the average SO_2 output and power generation for Run C-A were very nearly the same as for runs of the earlier series.¹

RESULTS AND DISCUSSION

The SO_2 washout concentrations measured for the sampling line are shown as radial bars on the sampling line map Figure 1. The dry ice freezing sample (C) concentrations are solid bars, and the liquid nitrogen freezing samples (N) concentrations are dashed bars. Figure 2 similarly shows concentrations measured for SO_4^{--} and pH. The N sample concentrations are not shown on Figure 2 because they were not significantly different from those of the C samples, with exceptions noted below. Cross-plume integrated washout fluxes,^{1,4} for Run C-A are listed in Table 3, along with those for runs of the earlier series, for comparison. Some of the former are indicated as lower limits, because the sampling line did not adequately contain the

TABLE 1. Analytical Procedures for Rain Chemistry.

Species	Analytical Procedure
H^+ (pH)	Glass electrode
SO_2 (dissolved)	Modified West & Gaeke method ⁵
SO_4^{--}	Methylthymol blue method ⁶
NO_3^-	Phenoldisulfonic acid method ⁶
	Chromotropic acid method ^{7,8}
NH_4^+	Phenoldisulfonic acid method ⁶
NO_2^-	Saltzman method ⁹

TABLE 2. Source Information:^a Centralia Steam Plant, Run C-A.

Time	Firing Rates		Power Output Mw	Stack Output			
	Coal, ^b kg/hr x 10^{-5}	Oil, ^c l/hr x 10^{-3}		SO_2 ^d ppm	SO_2 mo/sec	SO_3 ^e ppm	SO_3 mo/sec
1300 - 1400	2.54	4.94	334				
1400 - 1500	2.50	4.75	326				
1500 - 1600	2.35	5.32	292				
1128 - 1328				445	9.8	3.2	0.071

^aFiring Rates and power output from plant log sheets, stack output from Washington State University stack sample data.

^bAverage sulfur content of supply coals is 0.6%.

^cNumber 2 Diesel fuel, sulfur content 0.7%.

^dAverage of two simultaneous samples.

^eThe SO_3 injection rate was 16 ppm throughout the day.

SOLID BARS: OBSERVED (DRY ICE SAMPLERS)
 DASHED BARS: OBSERVED (LIQUID N₂ SAMPLERS)
 x---x---x EPAEC ESTIMATE, GAS PHASE
 o---o---o EPAEC ESTIMATE, STAGNANT DROP
 CONCENTRATION SCALE: $\sim 10 \mu\text{mol/L}$
 DISTANCE SCALE: $\sim 160 \text{ m}$

•
 SOURCE
 (SOUTH STACK)

Neg 66340-10

FIGURE 1. SO₂ Rain Concentrations, Run C-A.

SOLID BARS: OBSERVED SULFATE CONCENTRATIONS
 DASHED BARS: OBSERVED H⁺ CONCENTRATIONS

CONCENTRATION SCALE: $\sim 100 \mu\text{mol/L}$
 DISTANCE SCALE: $\sim 160 \text{ m}$

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 SOURCE
 (SOUTH STACK)

Neg 66340-9

FIGURE 2. Sulfate and pH Results,

washout plume, as inspection of the figures shows. However, the SO₂ washout plume appears to be about 95% contained.

Surface Desorption Evaluation

As was expected, the SO₂ concentrations in the N samplers were some-

TABLE 3. Cross-Plume Integrated Fluxes for Run C-A and for Line A of the Earlier Series.¹

Pollutant	W, (gram moles/cm sec) x 10 ⁶			
	Run C-A	Run C-A	Run C-A	Run C-A
SO ₂	0.045	0.026	0.00014	0.00113
SO ₄ ⁻	0.49+ ¹	0.030	²	0.006
NO ₃ ⁻	0.012+ ¹	²	²	²
NH ₄ ⁺	~0	²	²	²
NO ₂ ⁻	~0.0002	²	²	²

¹Plume not contained

²No measurement

what higher than those for the accompanying C samplers; the ratio of C to N concentration varied from 0.63 to 0.89, averaging 0.82. Thus, one may deduce that about 20% of the SO₂ in rainwater incident on the C funnel surface desorbed from the drops before they were frozen in the sampler bottle. Since this tendency to desorb is a function of the residence time on the funnel surface (generally proportional to rainfall rate), and the gas- and liquid-phase SO₂

concentrations of SO_2 , this result cannot be applied generally to other situations involving these variables. However, the apparent very low ground-level air concentration during this run--due to low wind speed and plume rise--strengthens the conclusion that the desorption effect should rarely be severe. If there was a significant plume of SO_2 at ground level (with a peak concentration within the range of the six sampler pairs), one would expect that the concentration ratio (C to N) would be increased at the peak. This was not the case; there was no systematic variation of the C to N ratios across the six sampler positions.

The meteorological and source conditions of Run C-A, which are summarized in Table 4, were used as input data for EPAEC, and sample calculations made. Unfortunately, this required making some estimates of input parameters, particularly the wind speed (the only data available were 15-min average data from the plant-operated meteorology tower), temperature (tower temperature sensors were inoperative that day), raindrop size spectra, plume loft, and Gaussian plume parameters. Plume rise was estimated using the approach of

Briggs;¹⁰ it was assumed that the plume centerline lofted at a constant velocity of 192 cm/sec (calculated from an estimate of stack heat output). At ten stack heights downwind, the plume was assumed to level off at a height of 602 m. Raindrop sizes were measured during the present experiment, but for convenience (and because errors in the raindrop spectrum would have less effect on the EPAEC calculations than other estimates used) a typical, frontal rain spectrum was used. The parameters of the Pasquill-Gifford plume employed by EPAEC were estimated by using the Smith and Singer¹¹ sigma values for a neutral atmosphere.

EPAEC estimates were made for the sampling line for the limiting cases of reversible washout behavior (the stagnant drop and gas-phase controlled cases,¹ and for the special case of irreversible (perfectly soluble gas) washout. The concentration estimates are shown on Figure 1 as solid line "profiles" of washout concentration, and are listed in Table 5. It is true that the estimated values of input parameters could be varied considerably to achieve various shapes and magnitudes of the washout concentration profiles, but this would be pointless, in view of the limited data available on which to base the estimates. Only the initial, best estimate, calculations were done for present purposes, merely to illustrate the general values which the model would predict for this situation.

The gas-phase-controlled concentration estimates are quite close to the observed concentrations for this set of input conditions, as compared with the factor of 3 to 10 high estimates for the earlier series. Presumably, this results from the particularly low wind speed and thus high plume and relatively vertical raindrop trajectories for this experiment. The raindrops collected on the sampling line thus traversed the plume farther from the source than in the earlier experiments, and thus any near-source difficulties with EPAEC were lessened. The stagnant drop concentrations are very much higher, as they usually are close to the source, because poorly mixed drops will tend to lose less SO_2 by desorption in the lower portions of

TABLE 4. Input Parameters for EPAEC: Run C-A.

Background pH	4.8
Temperature	278 K ^a
Sampling Distance	443 m
Plume Parameters: ^a	
σ_y (11)	67.8 m
σ_z (11)	62.5 m
Wind Speed	1.41 m/sec
Stack Height	143 m
SO_2 Source Strength	9.8 gram-moles/sec
Plume Rise Velocity ^b	1.92 m/sec
Rainfall Rate	3.0 mm/hr

^aEstimated.

^bSee reference 10.

TABLE 5. Observed and Calculated SO₂ Concentrations on Line A, Run C-A.

Sample Position	Concentration, (gram-moles/l) x 10 ⁶				
	Calculated			Observed	
	Gas Phase ^a	Stagnant ^b	Irreversible ^c	C ^d	N ^e
10				4.9	
11				6.4	
12				8.2	
13	0.15			4.7	
14	2.22	0.64		11.6	
15	7.98	1.41	11.3	10.4	
16	12.3	13.0	153.	15.2	18.6
17	14.3	49.2	1324.	17.6	20.8
18	15.2	104.	7344.	17.7	20.0
19	15.2	134.	25480.	18.9	21.6
20	15.2	104.	7344.	13.4	21.4
21	14.3	49.2	1324.	14.9	17.5
22	12.3	13.0	153.	10.7	
23	7.98	1.41	11.3	8.4	
24	2.22	0.64		7.1	
25	0.15			6.4	
Cross-Plume Integ. Flux (μms/cm sec)	0.038	0.151	13.8	0.045	0.055 ^f

^aLimiting case of reversible washout, where the raindrop is completely mixed.

^bLimiting case of reversible washout, where the liquid phase mixing is slower.

^cCase of SO₂ infinitely soluble.

^dDry ice sampler.

^eLiquid N₂ sampler.

^fEstimated.

the plume. The irreversible estimates, which are essentially those of the classical, or perfectly soluble gas scavenging, are grossly high, which has been demonstrated before.^{1,2}

Despite the gas-phase-controlled estimates being close to the observed concentrations, the importance of the effect of desorption from standard samplers still hinges on whether or not the ground level air concentrations of SO₂ were sufficiently low

to promote it. All visual (and certainly olfactory) indications were that the SO₂ plume was quite elevated over the sampling line. For what it is worth, the bivariate-normal plume model predicted essentially zero ground-level air concentrations; the fact that the predicted rainwater concentrations of SO₂ were still fairly high, despite the raindrops' having essentially left the plume, supports in some measure the conclusion that collector-surface desorption is generally small.

A test of the sampler-surface desorption evaluation was provided by application of a simple gas dry deposition model developed in the earlier study.¹ The relationship between incident rain concentration C_r and final (after surface absorption or desorption) C_f can be expressed by

$$C_f - C_r = \frac{F}{J} \left[C_{eq} - \frac{1}{2} (C_f + C_r) \right], \quad (1)$$

where J is the rainfall rate, C_{eq} is the water concentration of SO_2 at equilibrium with the local gas phase concentration, and F is a factor containing a "surface effect" mass transfer coefficient and the ratio of water covered to total collector surface area. The parts of F are hard to evaluate, but several controlled-release dry deposition experiments¹ resulted in an average value of F of 10^{-5} cm/sec, a result which applies only to the particular sampler surface ("C" sampler) and general range of temperature, etc., experienced during the controlled-release experiments. The application of (1) to Run C-A, along with the assumption $C_{eq} = 0$ (essentially zero ground-level gas phase SO_2 concentration), results in a value of $C_f/C_r = 0.89$. This is quite close--probably fortuitously--to the ratio of 0.82 determined experimentally at Centralia.

Sulfate and Nitrogen Concentrations

A surprising result of the experiments was the high sulfate concentrations found on the sampling line at about three stack heights downwind. These were considerably higher than any observed before at Centralia; this is reflected in the comparisons of fluxes in Table 3. The deposition "plumes" for sulfate are generally broader than those for SO_2 ,¹ but the existence of the peak sulfate concentration some degrees to the west of SO_2 peak (which coincided with the mean wind direction at stack top level) is puzzling. The presence of sulfate in the plume near the source due to the SO_3 injection led to the expectation of some increase in sulfate concentrations in rain near the source, but even considering this, an extremely high scavenging rate for sulfate is indicated. The scavenging rate (the rate of removal of a non-reactive pollutant by irreversible

processes) assuming a steady-state nondepletion source and vertical rainfall, is given by

$$\Lambda = \frac{W u}{Q_0}, \quad (2)$$

where u is the mean wind speed, Q_0 is the rate of emission of the pollutant, and W is the measured cross-plume integration flux. Considering the lower limit W of 0.49×10^{-6} g-moles/cm sec, $u = 141$ cm/sec, and Q_0 (SO_3) = 0.071 mo/sec, Λ is then ≥ 3.5 hr⁻¹, a very high value.

Past evidence of significant sulfate rain concentrations has implied significant conversion of SO_2 to sulfate within kilometers of the source. In the earlier experiments, it was assumed that the initial sulfur in the plume was primarily in the form of SO_2 ; presence of initial sulfate (which is more likely the case in C-A) would be expected to hinder the conversion process, rather than speed it.* If we assume a small initial amount of sulfate in the plume and a reasonable sulfate scavenging rate, the only conclusion from Run C-A is that there was a rapid oxidation of SO_2 to sulfate--by whatever means--near the stack. An alternative explanation is that the time between emission and scavenging was longer than that expected by considering the wind speed alone. This may be the case, as the directional shear of the wind was severe, amounting to about 180 degrees between the ground and the stack top. It is possible that the lower portion of the plume (in which SO_2 is presumably being converted to sulfate) was captured by lower level winds moving in the opposite direction, and became subject to scavenging above the samplers at some time later than the stack top wind speed would indicate. Unfortunately, there is no better wind data than visual observation regarding this possible backwash effect.

* For the case of liquid phase oxidation, where an increase of rain-drop acidity due to sulfate scavenging would suppress SO_2 dissolution, and thus further conversion.

The measured concentrations of the nitrogen compounds were generally much lower than those of the sulfur compounds. In fact, the nitrate concentrations were close to the sensitivity limit of the analysis, and a definite plume shape was not apparent (this may follow from the same conditions which may have distorted the sulfate plume). The ammonium ion concentrations were undetectable ($< 2 \mu\text{mo}/\ell$) in the standard samplers, but significant concentrations showed up in the liquid nitrogen samplers. These are unreasonably high, however, considering the other data, and were attributed to contamination from ammonia used to develop water-sensitive paper raindrop size samples. The nitrate and nitrite concentrations measured in the N samplers were also somewhat higher. All these are probably due to the liquid nitrogen acting as a trap for NO_2 ; there was probably sufficient NO_2 around the vehicles

used on the project to account for this. The boiling point of SO_2 is also warmer than that for N_2 , but the additional SO_2 from vehicle exhaust, etc., was expected to be sufficiently low to prevent this from being a problem.

Sequential Sample Concentrations

The sequential or short-time rain samples were collected at three locations: near position 19 on the sampling line, at a point 3.9 km from the stack and at azimuth 245 degrees (wind direction), and at a point 4.3 km and 175 degrees relative to the stack. The concentrations measured at the second point were essentially zero--and in fact the spot was probably not much affected by the plume--and they will not be considered further. The times and concentrations for the other samples are listed in Table 6. It is interesting to note that the average

TABLE 6. Sequential Sample Concentrations, Run C-A.

Sample #	Location	Time ^a PST	Rainfall Rate (mm/hr)	Concentration, (gm-moles/ ℓ) $\times 10^6$			
				SO_2	SO_4^{2-}	H^+C	NO_3^-
3 ^b	A19	1103	d	0	0	0	0
4	A19	1217	1.3	16.1	153.	84.0	0
5	A19	1224	0.8	12.1	73.6	109.	0
6	A19	1229	0.8	13.5	63.6	109.	2.1
7	A19	1238	0.6	11.3	62.6	109.	4.3
8	A19	1252	1.4	10.3	15.2	47.0	3.6
9	A19	1305	d	27.8	27.6	34.0	3.6
Avg. 4-9			1.0	15.2	65.9	82.0	2.3
12	3.9 Km, 175° ^e	1349	3.0	22.2	76.3	84.	0
13	" "	1353	1.7	10.6	87.6	47.	0.7
14	" "	1400	2.5	12.0	36.5	63.	0.7
15	" "	1412	2.0	13.6	81.4	109.	1.4
16	" "	1419 ^f	1.9	3.9	51.9	84.	2.9
17	" "	1432 ^f	d	1.5	43.2	63.	2.2
Avg 12-15			2.3	14.6	70.5	76.	0.7

^aTime at start; durations were from 2 to 5 min.

^bBackground sample; up wind of plant at the time.

^cLess background of $15.8 \mu\text{mo}/\ell$.

^dNo measurement.

^eAzimuth to stack.

^fAfter completion of Line A sampling.

of samples near position 19 during part of the run was essentially the same as that measured for the entire run in the C sampler at 19.

The short-term sample concentration data did indicate that the concentration of various plume pollutants does vary with time as the plume meanders. Use of a pH probe mounted in the rain sample stream proved useful in locating the plume in areas where it could not be seen. In addition, comparison of the average concentrations with long-term sample concentrations seems to indicate a not severe desorption problem for SO_2 for the 1 m^2 area funnel. Unfortunately, time did not permit use of the sequential sampler to define the wash-out plume, or to take samples at distances greater than 4.3 km.

CONCLUSIONS

In regard to the above-noted objectives of the study, analysis of the results of the field experiment led to the following conclusions.

1. It appears that the amount of SO_2 desorbed from the standard sampler is not significant, at least in terms of being sufficient to describe the discrepancy between EPAEC model predictions and experimental measurements close to an elevated SO_2 source. The conclusion rests upon a qualitative assessment that the ground-level concentration of SO_2 at the sampling line during the present experiment was sufficiently low to permit efficient desorption from exposed, untreated rainwater. The lack of a noticeable "plume effect" on the standard-to-no-desorption sampler concentrations tends to support this conclusion, but there is a remote possibility that, as a result of the sharp directional wind shear, there could have been a significant uniform SO_2 concentration across the sampling line.

There was no quantitative data taken to confirm or deny this possibility.

2. The nitrogen compound concentration measurements were accomplished, but at least for the plant at which the experiment was conducted, the concentrations were near the sensitivity limit of the chemical analysis techniques. Plans for further such measurements at Centralia should probably include consideration of more accurate means of measurement, particularly for the ammonium ion, which is expected to be a catalyst in the SO_2 oxidation process.
3. Use of large-area, portable samplers has value in determining the fluctuations in rainfall concentration due to plume variations which are not possible with long-term fixed-site samplers. There appears to be no severe SO_2 desorption problem with the use of such a sampler. For a suitable "acid plume" (such as the case with an isolated power-plant), the plume can be located by pH measurements of real-time precipitation samples.
4. No strong conclusion can be reached concerning the effect of SO_3 injection on downwind sulfate wash-out. Though sulfate concentrations measured 0.4 km from the stack were higher than any measured before at Centralia, this may be the result of the unusual rain situation which accompanied the experiment.

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BELOW CLOUD SCAVENGING OF AEROSOLS:
SOME IDEAS FOR IMPROVED MEASUREMENTS

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A discussion on improving measurements of below cloud aerosol scavenging is presented. Careful characterization of primary and secondary aerosols by direct sampling techniques appears to be a positive step forward in understanding scavenging mechanisms. Suggested improvements to the measurement system and suitable test and field experiments are given, as a preliminary design basis for planned revision in the below-cloud scavenging program.

INTRODUCTION

A major difficulty in relating experimental observations of aerosol washout to theory arises because of the polydispersity of the aerosol system. At present, most field and laboratory techniques involve measuring an integral process such as the washout of a particle size distribution by a hydrometeor distribution, whereas particle washout theory involves monodisperse aerosols or a differential process. Dana and Hales¹ have shown that serious errors can be made in the calculation of washout coefficients and/or collection efficiencies if the characteristics of the particle and precipitation spectra are only poorly known. In this paper, some ideas for improving aerosol scavenging measurements, especially in the field, by characterizing the aerosol with respect to chemistry and size distribution as well as the hydrometeor spectrum, are presented.

REQUIREMENTS FOR SCAVENGING MEASUREMENTS

The focus of any aerosol scavenging experiment is to know what aerosol is being scavenged and how the measurements relate to actual ambient conditions and aerosol properties. In order to examine the dependence of washout on particle and raindrop size distribution, it is useful to begin with a basic definition of washout coefficient; it is the ratio of the rate of pollutant

removal from the gas phase by washout (k) to the local ambient gas phase concentration (χ)

$$\Lambda \equiv k/\chi. \quad [T^{-1}] \quad (1)$$

For monodispersed particles scavenged by a raindrop size distribution $N(R)$, a simple geometrical argument leads to the familiar form

$$\Lambda(a, R_s) = \int_0^\infty \pi R^2 E(a, R) v(R) N(R) dR, \quad (2)$$

where

a is the particle radius,

R is the hydrometeor radius,

$E(a, R)$ is the collection efficiency, and

$v(R)$ is the terminal velocity of the hydrometeor of size R .

The integral washout coefficient resulting from the polydispersity of both the raindrop and particle spectra is obtained by integrating Eq. (2) over the particle distribution:

$$\Lambda(a_s, R_s) = \int_0^\infty \Lambda(a, R_s) f(a) da. \quad (3)$$

Here, $f(a)$ is a normalized particle density (number, mass, etc.) function. The subscript s in Eqs. (2) and (3) refers to some "characteristic or representative" size of the raindrop

or particle distributions. Unambiguous determination of these "characteristic" sizes is extremely difficult, even with precise knowledge of raindrop and particle size spectra, and represents a major source of uncertainty when trying to compare experiment with theory. Indeed, one effect of aerosol polydispersity, for example, is to yield a washout coefficient whose "characteristic" aerosol size may have a value considerably larger than the intuitively reasonable geometric mean size, i.e., the presence of larger particles greatly affects the washout coefficient for the entire distribution.^{1,2}

In view of the preceding discussion, it seems that measurements of integral washout quantities should be avoided and those which give differential washout are to be preferred. Unfortunately, many technical difficulties are associated with experiments designed to measure differential washout, particularly in natural systems, such as plumes or polluted air. Basically one is required to measure changes in the gas-phase concentration, $\chi(a)$, with time and to determine the washout coefficient $\Lambda(a, R_S)$ for each particle size range from Eq. (1).

It is evident from Eq. (1) that changes in $\chi(a)$ due to effects other than scavenging will lead to incorrect washout coefficients. For field experiments, this is a serious problem because a variety of processes may be present. These include:

1. Particle growth through coagulation or accretion of water, the growth rate in this latter case being dependent on the relative humidity of the environment and on the physico-chemical nature (solubility) of the particle. The latter effect will also be of considerable importance with regard to the measuring system, particularly if the samples tend to be dehumidified with a subsequent change in size during measurement.
2. Resuspension of particulate material from the ground or other surfaces (e.g. raindrop splash, etc.), perhaps as high as ~100 m at the start of a rain.
3. Dry deposition of aerosol, particularly in the larger size classes,

will tend to mimic particle washout. For soluble particles in environments where the relative humidity is near 100%, particle growth is enhanced leading not only to increased washout but also dry removal.³

4. Advection of new aerosol into the study region changing the total aerosol loading and/or the particle size distribution.
5. Evaporation of raindrops before hitting the ground, with the effect of transporting particles of different size classes from upper to lower levels. This effect may be most pronounced at the onset of a rain episode.

Some or all of these problems become acute when particle concentrations approach background levels, i.e. relatively clean air, for then $\chi(a)$ may exhibit random fluctuations from which no information on scavenging can be obtained.

Inspection of Eq. (2) reveals that if the raindrop size distribution changes with time then incorrect washout coefficients will also be obtained. In addition to changes in rainfall intensity, size changes due to evaporation (partial or complete), coalescence, and fragmentation of the falling raindrops may have important effects. The ambient relative humidity will be a controlling factor in raindrop size changes due to evaporation. There is some evidence from frontal rainfall studies by Dana⁴ that the raindrop spectrum is invariant with rainfall rate. If this assumption is made, then washout coefficients can be normalized to unit rainfall rate and the requirement for steady rainfall during measurement is not necessary. Now the rainfall rate J depends upon the third moment of the drop size distribution, i.e.,

$$J = \int_0^\infty \frac{4}{3} \pi R^3 v(R) N(R) dR$$

$$= \frac{4}{3} \pi N_0 v_t \int_0^\infty R^3 f_0(R) dR, \quad (4)$$

where

N_0 is the total number of drops,

v_t is the terminal velocity of some "representative" drop size, and

$f_0(R)$ is a normalized number probability density function for the raindrop distribution.

Thus it is still necessary to know the mean rainfall rate and the shape of the distribution function to calculate washout. The problem of raindrop size changes is still present. Slinn³ has pointed out that the collection efficiency may not depend sensitively on the raindrop size and that Eq. (2) would become simply

$$\Lambda(a) = \frac{3}{4} E(a) J/R_m, \quad (5)$$

where R_m is a characteristic drop radius. Whether or not one can make the simplifying assumptions as made above will depend on the results of careful measurements and tests.

It should be amply clear from the foregoing discussion that precise knowledge of the particle and rain size spectra and their histories is required before one can obtain meaningful differential washout quantities. Considerable caution needs to be exercised to ensure that the measurements are representative of the particles actually being scavenged and that the measuring instruments do not alter the physical characteristics of the particles during measurement.

PREVIOUS EXPERIMENTAL APPROACHES

Up to the present time, most experimental studies of aerosol washout have been carried out with laboratory aerosols or with radioactive or chemical tracer materials.⁵ While these studies have been valuable in defining the order of magnitude for particle washout coefficients, they also have certain difficulties which make interpretation of their results ambiguous.

The laboratory experiments are generally designed to isolate the various scavenging parameters such as raindrop size, particle size, and other relevant physico-chemical properties (e.g. solubility) of the particles. The scavenging coefficient as a function of the above parameters is determined from quantitative chemical analysis of the collected raindrops. This procedure, of course,

restricts the types of aerosols which can be studied and consequently the laboratory aerosols which have been studied are not entirely representative of natural aerosols. It is also difficult to control the environmental conditions in these experiments so that duplication of actual conditions that real raindrops and aerosols experience in the atmosphere are seldom attained.

Field experiments using tracers have had only limited success. The greatest difficulty in interpreting the results of these experiments is the lack of knowledge of the tracer particle sizes and dry deposition rates. Changes in the tracer particle size distribution after release (primary aerosol to secondary aerosol) by coagulation, by attachment to other natural particles or by accretion of water (if the tracer particles are soluble) will be completely masked by the traditional measurement and analysis techniques, which generally lead to integral washout coefficients anyway.

INSTRUMENTATION AND MEASUREMENT TECHNIQUES

Recognizing that growth of the primary particles by water condensation and resuspension of ground material may represent the primary difficulties encountered in any detailed investigation of submicron particle scavenging, the problem may be turned around instead to consider these mechanisms as part of the primary objective of the experiment. Therefore, it is essential that a direct measurement technique be employed and that both the primary and secondary aerosol sizes be measured carefully and well. The first requirement ensures confidence in the measured size distributions as well as allowing for examination of individual effects, while the latter requirement is needed for distinguishing the various scavenging mechanisms.

A direct measurement technique is one where actual physical samples, e.g. filter paper samples, are analyzed according to size or any other physical characteristic with a direct measurement instrument such as a scanning electron microscope (SEM). An indirect technique, on the other hand, is where the particle classification

depends upon some theoretical relationship but must be ultimately calibrated using a direct method. Indirect techniques often have several advantages over the direct measurement techniques, including faster sampling rates, in situ capability, and ease of automation, but have the disadvantages of uncertainty in the interpretation of the measurements and masking individual effects. Certainly indirect techniques, such as optical (Royco optical particle analyzer) and electrostatic (Whitby electrostatic particle analyzer) techniques, should be used in the measurements, but only as backup to the main experiment and to compare and correlate to the results obtained with direct measurements.

Because of the importance of particle growth by water condensation, good humidity control in the sampling system is crucial. An environmental chamber, whose relative humidity can be controlled and monitored, may be used to produce dry (primary) or wet (secondary) particles from the inlet stream prior to entering the sampling system simply by regulating the relative humidity between about 60% and 100%, independent of the ambient atmospheric relative humidity. Alternatively, dry particles could be produced simply by passing the inlet stream through a dryer, without great concern about the relative humidity so long as it is low enough. To ensure that the environmental conditions are not changing during sample collection or sample classification, relative humidity sensors should be located at strategic positions in the samplers themselves.

The sampling system must be chosen with care. Ideally, the sampler should perturb the environmental conditions only minimally. This requirement is particularly important during classification of the wet particles, where a decrease in the relative humidity could cause a decrease in the aerosol sizes. For dry (primary) particles, where size changes due to evaporation are not important or even relevant, a simple filter (nucleopore or millipore) collection is adequate. Several types of particle size separators or elutriators, such as cascade impactors or centrifuge aerosol spectrometers, are currently available and may be suitable for sizing the wet, secondary particles. An inspec-

tion of the operating characteristics of the various elutriators reveals that cascade impactors tend to dry wet aerosol because of the large pressure drop typically encountered during operation, and therefore do not meet the desired requirements. The Goetz and Stoeber aerosol spectrometers appear better suited to these requirements, although the potential problem of excess water due to high relative humidity of the sample stream "washing out" the deposition pattern needs to be examined closely. A schematic diagram of a possible particle sampling system is shown in Figure 1.

For the purposes of this experiment, a simple measurement of rainfall rate and raindrop size spectra should be more than adequate. At present, an ozalid paper technique is used for raindrop sizing.

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FIGURE 1. Particle Sampling System.

APPLICATION AND EXPERIMENTS

Once a good humidity controlled system, versatile enough for both ground and aircraft measurements, has been constructed, a number of interesting and important experiments suggest themselves, such as:

1. Careful tests of the sampling system to decide which instruments will be most useful, to test the

- humidity control system, and to determine whether such a system would indeed lead to improved wash-out measurements. These tests could be carried out on laboratory generated aerosols or on natural aerosols.
2. Aerosol characterization tests on laboratory and natural aerosols, the latter under various meteorological conditions.
 3. Below cloud scavenging of natural aerosols. Here, it is important that reasonably quiet meteorological conditions exist during a rain episode, where there is little or no horizontal or vertical motion of the ambient air and no new aerosol is advected into the study region. Such conditions are presently being compiled and apparently occur with some regularity in November/December over the Hanford area. Raindrop evaporation for this time of year is probably negligible. Low particle concentrations, typical of clean air, may hamper measurements. Scavenging by snow should not present any greater difficulties than the rain scavenging experiments.
 4. Scavenging of tracer aerosols released in a plume. In principle, these measurements should be no different than those described in (3) except that artificial aerosols are introduced into the natural system. This experiment has the potential of studying several of the scavenging mechanisms at once, such as coagulation of the tracer, attachment of the tracer to natural aerosol, and tracer growth by water condensation. For successful interpretation of the results, the size distribution of the tracer must be measured at the point of release.
 5. Scavenging of aerosols in a plume, probably using an aircraft for plume sampling. This experiment is similar to Radke's plume experiments,⁶ except that the humidity will be more carefully controlled and monitored.

Because all the experiments can be carried out with optical or electrostatic particle analyzers as back-up to the direct samplers, a large amount of data will be available for assessing the reliability of these other instruments for use in scavenging measurements. Current plans are to implement a sampling system of this type during the coming year.

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IN-CLOUD SCAVENGING OF AEROSOL PARTICLES

W. G. N. Slinn

A simple model for in-cloud scavenging of aerosol particles is presented which focuses on the removal of the pollutant from the cloud.

INTRODUCTION

For some pollutants, e.g., for bomb debris from high altitude tests, the dominant mechanism of atmospheric cleansing is precipitation scavenging from within clouds. Yet considerably more is known about scavenging from below than from within clouds. Consequently it is of interest to perform experiments and develop models that will elucidate in-cloud scavenging. During the last decade more than 100 in-cloud scavenging tests have been performed.¹ In contrast there have been very few interpretive models developed. This note outlines an extension to the author's recently described model for in-cloud scavenging.^{2,3} There, attention was focused on the attachment of pollutants to condensed cloud water; here the focus is on the removal of the pol-

lutant from the cloud. A more complete description of the model can be found in the original publications.^{4,5}

GENERAL FORMULATION

Figure 1 indicates the notation and the general concept of the model. If x_f is the mean concentration of the contaminant unattached to cloud particles (f = free), it is assumed that the evolution of x_f is described by the convective diffusion equation:

$$\frac{Dx_f}{Dt} = \nabla \cdot (\tilde{K} \cdot \nabla x_f) - \alpha x_f - \psi_f x_f \quad (1)$$

where

$$Dx_f/Dt = \partial x_f / \partial t + \vec{v} \cdot \nabla x_f$$

is the total time derivative. In the

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FIGURE 1. A Summary of Notation Used in the Text.

above, $\vec{v}$ is the wind field; $\tilde{K}$ is the (second-order tensor) turbulent diffusivity;⁶ α is the rate at which the contaminant particles attach to condensed cloud water;² and ψ_f is the rate at which the particles are directly removed from the cloud by hydrometeors. Similarly the contaminant attached to condensed cloud water, x_c , evolves according to

$$\frac{D_c x_c}{Dt} = \nabla \cdot (\tilde{K}_c \cdot \nabla x_c) + \alpha x_f - \psi_c x_c \quad (2)$$

where the subscript c is added to $\vec{v}$ and $\tilde{K}$ to admit the case when the cloud particles' motion differs from the air's. If this is ignorable, then the sum of (1) and (2) gives

$$\frac{D x_t}{Dt} = \nabla \cdot (\tilde{K} \cdot \nabla x_t) - (\psi_f x_f + \psi_c x_c) \quad (3)$$

where

$$x_t = x_f + x_c$$

is the total contaminant concentration in the cloud. In (3) an overall in-cloud removal rate can be identified via

$$\psi_t = (\psi_f x_f + \psi_c x_c) / (x_f + x_c) \quad (4)$$

so that (3) can be written as

$$\frac{D x_t}{Dt} = \nabla \cdot (\tilde{K} \cdot \nabla x_t) - \psi_t x_t \quad (5)$$

ATTACHMENT

Elsewhere^{2,3} attention was focused on the attachment rate, α . For example, for attachment by Brownian diffusion of particles of radius $a(t)$ to cloud droplets of radius $r(t)$, $\alpha = 4\pi r D(a) N(\vec{r}, t)$ where D is the particle diffusivity and N is the mean number density of cloud droplets. For this case the attachment process is linear in x_f and therefore if N is fairly uniform, turbulent diffusion does not influence attachment by Brownian motion. Other physical processes that might contribute to attachment are phoretic⁷ and electrical effects.^{4,8} Other authors have considered the enhancement of attachment caused by turbulence⁹⁻¹¹ but it is not clear if this would be important for submicron-particle/cloud-droplet co-

agulation since there is little energy in the turbulent eddies whose space scale falls between the sizes of these two types of particles. "Attachment" via water vapor condensation on the particle will be considered here not as an attachment process but by accounting for variable aerosol particle size, viz., $a = a(\vec{r}, t - t_0)$. Because attention was focused on the attachment rate elsewhere,^{2,3} here major attention is on the removal rates.

REMOVAL RATES

It is clear that a mathematical formulation for the removal rates will lead to the same results as for the below cloud scavenging cases. Thus for regions in the cloud where the hydrometeors are liquid, then

$$\psi = \int_0^\infty dR N(R) \pi R^2 E(a, R) v_t(R) \quad (6)$$

where $N(R)$ is the number distribution function for the drops; $E(a, R)$ is the collision efficiency and $V_t(R)$ is the drop's terminal velocity. Recalling the difficulties⁴ with the similar expression for the below cloud case (especially the problem of specifying the drop number distribution function for predictive formulae) it is suggested that the approximation developed elsewhere^{4,5,12} can be utilized here for the in-cloud case. Thus (6) is approximated by

$$\psi = c_1 \frac{J_0(\vec{r}, t)}{R_m(\vec{r}, t)} E[a(\vec{r}, t - t_0), R_m(\vec{r}, t)] \quad (7)$$

where c_1 is a constant $O(1)$ ($c_1 = 1/2$ was used for the below cloud case); J_0 and R_m are the rainfall rate and mean drop size, respectively, at position $\vec{r}$ within the cloud at time t ; and E is the collision efficiency for collisions between particles of radius a which may vary with time $(t - t_0)$ since the particle entered the cloud at time t_0 . Subscripts f and c can be added to (7) to conform to the formalism above, e.g., see equation (4). For the case where the hydrometeors are frozen, then utilizing the approximation developed elsewhere^{4,5,12} (7) is replaced by

$$\psi = c_2 g \frac{\rho_w}{\rho_a} \frac{J_o(\vec{r}, t)}{v_t^2(\vec{r}, t)} \mathcal{E}\{a(\vec{r}, t - t_o), \ell(\vec{r}, t)\} \quad (8)$$

where c_2 is another constant $O(1)$; g is the acceleration of gravity; ρ_w and ρ_a are the densities of water and air, respectively; J_o is the precipitation rate (rainwater equivalent); v_t is the average terminal velocity of the ice crystals; and $\mathcal{E}$ is the collision efficiency for particles and ice crystals of characteristic length scale ℓ , which is not necessarily related to the crystal's size.^{4,5,12}

APPROXIMATIONS

The general formalism presented above will not be approximated for some special cases of interest.

1. Rapid Attachment

It appears that for most tracer particles used in field tests, the particles rapidly attach to the cloud water.²⁻⁵ Then in the above expressions $\psi_t = \psi_c = \psi$ and $\psi_f = 0$. Then, also, the appropriate collision efficiency is that for hydrometeor/cloud particle collisions which, to a first approximation can be taken to be unity. Consequently, under these approximations, the general formula reduces to

$$\frac{Dx}{Dt} = v \cdot (\vec{K} \cdot \nabla x) - \psi x \quad (9)$$

where the removal rate, for positions in the cloud where the hydrometeors are liquid, is

$$\psi(\vec{r}, t) = c_1 J_o(\vec{r}, t) / R_m(\vec{r}, t) \quad (10)$$

and, where they are frozen,

$$\psi(\vec{r}, t) = c_2 (g \rho_w / \rho_a) J_o(\vec{r}, t) / v_t^2(\vec{r}, t). \quad (11)$$

2. Stable, Steady-State Clouds

If in addition the cloud is horizontally homogeneous and transients can be ignored, which might be an ac-

ceptable first order approximation to reality for the removal of bomb debris from stable frontal storms, then the $\vec{r}$ -dependencies of (10) and (11) become just z -dependencies and the time variations can be ignored. Further, if diffusion is ignorable compared with transport and removal then (9) and (10) become

$$\bar{u}(z) \frac{\partial x}{\partial z} = -c_1 \frac{J_o(z)}{R_m(z)} x \quad (12)$$

where the x -axis at height z has been aligned in the direction of the wind at this height. Thus from (12) the concentration at height z (or, more reasonably, on a particular isentropic surface) falls according to

$$x(z, x) = x_o(z, x = 0)$$

$$\exp \left\{ -c_1 \frac{J_o(z)}{R_m(z)} \frac{x}{\bar{u}(z)} \right\}. \quad (13)$$

If, for elevations below the freezing level, raindrop growth does not significantly influence the rainfall rate or the mean raindrop size then as a first approximation $J_o(z)/R_m(z)$ might be taken as a constant. In this case the flux of contaminant from the cloud, if originally the contaminant was spread over a height interval ΔH , all beneath the freezing level, is obtained by integrating the removal ψx from each volume. The result for the flux under these conditions is

$$F = \Delta H x_o \psi_o \exp \{-\psi_o x / \langle \bar{u} \rangle\} \quad (14)$$

where $\psi_o = c_1 J_o / R_m$. Although this is a simple result it can lead to complicated, ground-level deposition patterns as can be seen from describing the trajectories of different size drops through typical wind shears.^{4,13}

3. Steady-State Convective Clouds

If a pollutant or tracer is continuously ingested into the updraft of a cumulonimbus cloud, and if the particles can act as cloud condensation nuclei, then it is expected that the general formalism can be approximated by

$$w \frac{\partial x}{\partial z} = \frac{-c_1 J_0(z)}{R_m(z)} x \quad (15)$$

at least for the case of liquid hydrometeors. In (15), w is the updraft speed. As an estimate of the predictions of (15) suppose that J_0/R_m and w were independent of z . Then

$$x = x_0 \exp(-c_1 J_0 z / R_m w) \quad (16)$$

which for $J_0 = 10 \text{ mm hr}^{-1}$, $R_m = 1 \text{ mm}$ and $w = 10 \text{ m sec}^{-1}$ predicts an e-fold length of 3-10 km, depending on c_1 . Thus a substantial fraction of the ingested CCN will be removed and, indeed, it is suggested that one can assume that the efficiency of the convective storm to remove particles ingested into the updraft is equal to the storm's efficiency to remove water, ϵ . The resulting removal rate is then approximately

$$\psi = c_1 \frac{J_0}{R_m} \epsilon \quad (17)$$

where for $c_1 = 1/2$ and $\epsilon = 1/3$ provides a reasonable fit to Burtsev et al.'s data.¹⁴ (See Figure 2.) Presumably this fit could be improved if account were taken of variations in the storm's efficiency to remove water. For pollutants only known initially to be in the neighborhood of a convective cell, the first task would be to estimate the amount of pollutant ingested by the storm. This is a problem in dynamics that needs further attention.

CONCLUSIONS

The above model for in-cloud scavenging should be sufficiently general to describe problems of practical interest and should provide an adequate framework for interpretation of tracer experiments. Whether or not the approximations discussed can be applied depends on specific conditions occurring in the scavenging experiments. It is encouraging that a simple version of the model appears to explain the data of Bertsev et al. satisfactorily.

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FIGURE 2. A Comparison of the Precipitation Scavenging (Washout) Rate, ψ , as Obtained Experimentally with the Approximation, (17) to the Theoretical Result. The data were obtained by releasing tracer aerosol particles into the top of the rain shaft (solid circles) or into the cloud droplet zone (open circles) of a cumulonimbus cloud.

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RESULTS OF IN-CLOUD TRACER RELEASES IN FRONTAL STORMS

W. E. Davis and J. A. Young

Six frontal storms have been injected with a variety of tracers. Measurable concentrations of the tracers have occurred in many rainwater samples examined. Maximum tracer deposition has occurred 20-30 nautical miles downwind from the area where the vertical wind profile, coupled with raindrop fall velocities of 1.5 m sec^{-1} , would indicate initial ground deposition should have occurred. Some possible explanations are presented as well as a discussion of tracers used.

INTRODUCTION

In-cloud scavenging processes appear to play a major role in cleansing the atmosphere of pollutants.¹ The actual magnitudes associated with these processes, however, are poorly known. Consequently there is a great need for field experiments to obtain knowledge of the causal relationships necessary for formulating reliable frontal-storm models. In the past, several successful field experiments have been conducted in which tracers have been injected into convective systems (see Burtsev, et al.,² Dingle, et al.,³ Semonin,⁴ Skopauskas, et al.,⁵ Summers,⁶ Warburton,⁷ and Young, et al.⁸). The work presented in this paper differs in that it deals with in-cloud scavenging in prefrontal clouds.

The present article summarizes the experimental design and results of experiments conducted in six frontal storms under our AEC/DBER-sponsored precipitation scavenging program. Final sections of the article are devoted to conclusions and a summary of plans for the ongoing program.

RESEARCH PLAN

The following is a list of research objectives for the past in-cloud scavenging experiments:

1. To field test tracers to find those that would be deposited in measurable concentrations,
2. To determine the effect of micro-physical parameters (e.g., solubility, wettability, aerosol size, cloud physics parameters) on in-cloud scavenging,
3. To assess the effect of state properties (e.g., temperature) on scavenging, and
4. To evaluate the influence of storm dynamics on removal processes.

In Table 1, which summarizes the research plans and objectives of the individual experiments, we have attempted to look at a number of different meteorological conditions in frontal storms, in order to estimate removal in storms where nucleation effects have been enhanced by silver iodide, as well as removal in storms injected with an essentially inert tracer. A discussion of the individual experiments with the results of analyses that have been performed follows.

EXPERIMENTS

Initially, part of the research plan [see (1) above] aimed to find a suitable tracer or tracers. Since Dingle² had prior success with indium,

it was selected as a tracer in the initial experiments. Lithium and silver were also selected to be released the first year.

March 11, 1971

On March 11, 1971, a test was carried out at Quillayute (UIL), Washington, with indium flares and lithium stearate released into pre-frontal clouds. The results presented by Atkinson, et al., in 1972, will be reviewed briefly here.⁹ As indicated in Table 1, the 450 g of indium were released by an aircraft at 0°C, in a stable layer at 5000 ft within the storm on two legs each 5 nautical miles. The release was made using the University of Washington's B-23 aircraft. The UIL radiosonde winds and a raindrop fall velocity of 1.5 m/sec,* were used to determine

* Fall velocity of 1.5 m/sec was selected since past UIL raindrop size measurements indicated a number mean radius of 0.02 cm.¹¹

the position for the release so that the deposition of the tracer would occur in the center of the sampling arcs. The indium deposition pattern, shown in Figure 1, did occur in a predicted area. From the pattern, a calculation showed that about 30% of the tracer deposited during about 20 min. A removal rate was then estimated to be approximately $1.5\% \text{ min}^{-1}$.

It is interesting to note that the maximum concentrations in the deposition pattern did not occur in the area where the tracer was targeted to arrive initially, but at a position downstream on the last arc. If we were to base our estimations on the assumption of pseudo below-cloud scavenging with a constant washout coefficient, the maximum deposition would be expected to occur at the initial depositing position with deposition decreasing with time. Since this doesn't appear to be the case, two possibilities have been put forth: 1) that the material injected is gradually attached to the cloud water

TABLE 1. In-Cloud Scavenging of Tracer Particles by Frontal Storms.

DATE (LOCAL)	3/11/71	4/16/71	3/21/72	3/31/72	3/31/72	2/28/73	12/5/73
RELEASE ALT (FT)	5000'	12000'	6800'	6200'	12000'	5000'	5000'
RELEASE TIME (PST)	1157-1204	1319-1327	0938-0945	1556-1615	1635-1649	1523-1541	0945-1025
RELEASE TEMP (°C)	0°	-15°	0°	0°	-6.6°	0°	2°
TRACERS RELEASED	1n 450 g (F) Li STEARATE 60 g	1n 450 g (F) AgI 500 g (F)	1n 450 g (F) AgI 2 kg (F)	RHODAMINE 600 g	1n 500 g (F) AgI 1.2 kg (F)	1n ~400 g (G) AgI (G) Li 216 g (F) HEAVY DEPOSITION ON BURNER, AMT BURNED UNK	(Re 17.4 g) (Eu 36.5 g) RELEASED RAW (G) 1n 600 g (F) Li 795 g (G) Ir 9.7 g (G) Os 7.6 g (G) (Re 17.4 g) (Eu 36.5 g) 1/2 RELEASED RAW (G)
(F) = FLARES (G) = GENERATOR							
STABILITY ₁	STABLE	UNSTABLE	STABLE	STABLE	STABLE	STABLE	STABLE
WINDS UIL (M/SEC)	180/30	182/11	173/16	210/22.5	240/23	200/13	199/22
GOALS	1. FEASIBILITY 2. WETTABLE VS NONWETTABLE	1. FEASIBILITY 2. ICE NUCLEI VS NONICE NUCLEI	1. DETECT DECREASE	1. DETECT SCAVENGING OF ORGANIC PARTICLES	1. TARGET TRACER 2. PROPAGATION OF ICE PHASE	1. PARTICLE SIZE DEPENDENCE	1. PARTICLE SIZE DEPENDENCE 2. SOLUBILITY
	100 SAMPLERS 80 KM		250 SAMPLERS 160 KM		SEQUENTIAL SAMPLERS		

¹ WITH RESPECT TO MOIST ADIABAT

Neg 720024-6cn

FIGURE 1. Indium Deposition,
March 11, 1971.

which is more easily scavenged¹⁰ and 2) that the rainfall rate increased causing the scavenging coefficient to increase which in turn caused the deposition to increase. The effect could also be a combination of the two. Figure 2 analysis shows the rainfall rate using the volume/time ratio to denote a mean rate for the time the individual samplers were out. Comparing this to the deposition, some correlation occurs between the higher rain fall rates and higher deposition rates. (This should be kept in mind when the results of the December 5, 1973, test are discussed.)

Also during this experiment, approximately 60 g of lithium was dispersed in the form of lithium stearate. None of the samples analyzed to date contained lithium concentrations significantly above background.⁹

Neg 745953-5

FIGURE 2. Rainfall Rate Grams/hr, March 11, 1971.

April 16, 1971

The second experiment was carried out on April 16, 1971, when a release of 500 g of silver iodide and 450 g of indium was made at -15°C at 12,000 ft on two legs of 5 nautical miles (see Table 1). Again, details are presented in Atkinson, et al., 1972.⁹ A photograph taken after release from the University of Washington aircraft indicated a line of convective development along the seeding path.⁹ The release, unfortunately, was made in a lighter wind speed than the previous storm. Using the same size grid for the lighter wind speed resulted in loss of detail in the deposition pattern. Figure 3 shows the deposition of indium.

Estimates based on the winds at 12,000 ft indicate the time of arrival on the first arc near UIL would be 30 min after initial deposition using a 1.5 m/sec fall velocity. The deposition path should have moved northward as indicated by the 12,000 ft winds, crossing the northern arc at 75 min after initial deposition. There is some organization to the pattern in the western edge of the arcs as seen in Figure 3. However,

Neg 720024-2cn

FIGURE 3. Indium Deposition, April 16, 1971.

there are significant measurements that indicated deposition further to the east. These measurements could not be explained from the UIL radiosonde taken at the time of release. This indicated that in future tests better definition of the wind flow pattern aloft is necessary to predict accurately the deposition pattern of the injected tracer. The wind directions computed from the 1935Z sounding, taken 1 hr and 44 min before release, provide some basis for explaining the eastern deposition. Little correlation was found to exist between measured silver and indium concentrations in the samples. Three samples contained both indium and silver, whereas 36 samples had measurable quantities of either silver or indium, but not both. It should also be noted that the maximum concentration for the two tracers occurred at different locations.

March 21, 1972

On March 21, 450 g of indium and 2 kg of silver iodide were released in prefrontal clouds over two legs of 8 nautical miles.¹² The release on this occasion was made over an expanded sampling network which is shown in Figure 4. The reason for the expanded network was the result of the previous year's analysis of a March 11 release indicating maximum deposition on the last arc.^{9,10} By increasing the downstream size of the arc, it was hoped that a falloff from a maximum deposition could be seen. Also an auxiliary radiosonde was established south of the arc system at Hoquiam, Washington, in order to better define the upper wind flow pattern. The injection of both tracers was made 6800 ft at -5°C in a stable layer by the University of Washington aircraft.

In sharp contrast to our initial predictions, virtually no scavenging occurred over the network. Only three samples showed slightly above background concentrations of indium (see Figure 4) and no samples showed concentrations above background of silver.¹³ Radar and pilot reports were examined subsequently in an attempt to evaluate this behavior. Problems existed with the RHI at Neah Bay, thus no photos or readings were available during the test. Some readings off an auxiliary scope 14 min before the test indicated returns to 10,000 ft in an area of the release. The PPI photos taken during

Neg 745953-2

FIGURE 4. March 21, 1972, In and Ag Release, 6800 ft at 0°C, 2 kg AgI and 450 g In.

the test period, however, failed to indicate any echo returns from this area.

The aircraft during the flight from UIL to the release point reported at one point breaking out of clouds into the sun at 6300 ft. A review of tape recordings taken aboard the aircraft indicated that visual ground contact occurred occasionally during the release, as well as a reported bright spot due to sun overhead that was visible at times. Information from the tape indicated that the release was made in rain although no indication of intensity was given. From this description of the clouds, the rain was probably sporadic along the release path. Also, rain gages on the ground indicated only light rain during the period when tracer deposition should have occurred. Thus the

initial conclusion reached was that although the release was made in cloud, the rate of removal from the cloud was insufficient for organized measurable surface concentrations.

March 31, 1972

Two tracer experiments were carried out during a flight conducted on March 31, 1972.¹² Rhodamine had been used previously, with success, in below cloud scavenging experiments.¹⁴ Thus it was decided that a test be made to determine the suitability of rhodamine as an in-cloud tracer. Rough calculations indicated that deposition above the detection limit of 1 ng should be possible. Based on these calculations, 600 g were released at 6200 ft at 0°C on two flight legs of 9 nautical miles¹⁰ by the University of Washington B-23.

A fluorimetric analysis of precipitation samples in the area of expected deposition revealed none above background. Samples further to the west indicated positive results plus some organization (see Figure 5).⁹ Further analysis of samples this year by a spectral fluorimeter indicated that no fingerprint for rhodamine in the samples could be detected.¹⁵ In fact, when samples were spiked with 20 ng of rhodamine, interference was still too great to obtain a fingerprint. From these results, we conclude that rhodamine is not suitable as an in-cloud tracer.

Silver iodide and indium were also released into the same storm at 12,000 ft at -15°C in a stable layer with significant liquid water. This release resulted in a "hole" in the cloud,¹² owing to the nucleation activity of the silver. The release was made during stable lapse rate conditions in a cloud containing significant amounts of liquid water, to see if nucleation by silver iodide would occur and whether deposition

of precipitation could be targeted. Again using 1.5 m/sec fall velocity and the wind profile from UIL, the targeted area was Forks, Washington (see Figure 6). Only one sample from this series has been analyzed for indium to date and it contained 2 ng.

February 28, 1973

On February 28, 1973, a second attempt was made to conduct a tracer release which would cover the extended arc system. Using the winds from radiosonde data at Quillayute, Washington, an area for release was selected for a tracer injection, as seen in Figure 7. Radar indicated echoes in that region; however, they did not appear to be precipitating. Instructions were given the personnel of the aircraft to orbit the area of release, and if they hit a precipitating cell to release. These instructions were carried out and a cell was injected with tracer by the Battelle Cessna-411. Lithium flares with 216 g of lithium and ~400 g of

Neg 740788-1

FIGURE 5. March 31, 1972, Rhodamine.

Neg 745953-10

FIGURE 6. March 31, 1972, In and Agl Release, 12,000 ft, -15°C, 500 g In, 1.2 kg Agl.

indium* from burners were released at 5000 ft at 0°C.

Rain gage measurements and 15 min sequential samples showed little rain falling (<0.01 in/45 min) on the first arc during predicted passage. No rainfall was recorded by the Lake Pleasant (A on Figure 7) rain gage during time of predicted deposition. A rain gage further to the north and west of the deposition pattern indicated 0.01 in/15 min during time of predicted deposition. The sequential samples east of Lake Pleasant indicated a minimum slightly before the time estimated for deposition, and a slight increase to 0.001 in/15 min during the time deposition should have occurred.

* 837 g were planned for release, however, due to failure in the release system, only ~1/2 was released.

The average rainfall rate during the test was calculated for each sampler from the amount collected and the time exposed. The result of this calculation indicated a maximum along Burnt Mountain Road (see Figure 8).

The initial analysis of samples for lithium is shown in Figure 7. As can be seen, the maximum deposition occurs in the area of maximum precipitation. The concentration of lithium in the water samples varied widely with values running from 0.07 ppb to 10.2 ppb. The highest deposition occurred in the samples along Burnt Mountain Road and in a sample collected at the junction of Highway 101 and the Hoh River Road (B on Figure 7). An indium analysis has not yet been performed this test.

Neg 745953-7

FIGURE 7. February 28, 1973, Lithium Deposition Pattern, ng/Bucket (0.2 m diameter) Lithium Release 216 g 5000 ft, In Cell, 0°C, Stable.

Neg 745953-6

FIGURE 8. February 28, 1973, Precipitation Rate, g/Bucket hr.

December 5, 1973

A series of tracer releases was made by the Battelle aircraft at 5000 ft in a stable layer at 2°C on December 5, 1973.¹⁶ The plan for the release of the tracer differed from past plans; previously, control on the area of burn was determined by both the rain trajectory and the wind direction at the release height with the aircraft releasing into a precipitation cell. The updated plan was formulated as a result of the March 21, 1972 test analysis, which indicated unexpectedly low deposition rates of tracer. The new plan required that a cell be visible on the PPI, an indication of precipitation occurring on the RHI, and that the cell be located in an area allowing deposition to occur over the arcs.

The tracer portion of the experiment included two tracers previously used, indium and lithium, and the addition of four new tracers for use

at UIL: europium, osmium, rhenium, and iridium. The effect of particle solubility was to be studied by the simultaneous release of a soluble tracer such as lithium and a nonwettable tracer such as iridium or indium. A water-soluble tracer should act as a much better cloud condensation nuclei than a nonwettable tracer and therefore might be scavenged more rapidly.

Other experimental features were also to be considered. A burn of a rhenium-europium mixture in the acetone generators was to be carried out to determine whether the two tracers would behave identically, showing the same deposition patterns. Also planned was a release of osmium into a precipitating cell to determine the suitability of osmium as a tracer.

On December 5, 1973, a warm front was approaching the Washington coast (Figure 9). The PPI radar at Makah AFB near Neah Bay, Washington, showed

Neg 745953-1

FIGURE 9. Battelle-Northwest Synoptic Meteorology Section, Surface Chart, Date December 5, 1973, Time 18Z.

a cell in the planned release area south of Quillayute. The decision was made to release tracers into this cell, and the Battelle Cessna-411, equipped with indium flares on one wing, and two aerosol generators on the other, was directed to this cell. The display of the transponder signal from the Cessna on the radar scope was used to position the aircraft within the precipitating cell.

When the Cessna reached the proper position, an attempt to burn the europium-rhenium solution in the aerosol generators failed because of ignition problems. The indium flares were then burned from 0950 to 0956, and a second attempt was made to light the aerosol generators. This time, because one of the generators fired and the other did not, lithium,

iridium, osmium, and the remaining rhenium-europium solution were released sequentially with only one burner firing. The burn of the last tracers was completed at 1025. The tracers were all released at 5000 ft at an ambient temperature of 2°C and in a stable layer (see Table 1). Following the release, samples of rain were collected from the 190 rain collectors over the expanded rain sampling network.

The result of the analysis of the samples to date is shown in Figures 10, 11, 12 and 13. In the case of indium, the analysis is nearly complete (Figure 10). An organized pattern from the analysis can be seen. This pattern is surprising in that the wind direction obtained from the UIL radiosonde indicated

Neg 757552-1

FIGURE 10. December 5, 1973, Indium Deposition Pattern, ng/Bucket (0.2 m diameter) Indium Release 600 g 5000 ft, In Cell, Stable, 2°C.

Neg 745953-15

FIGURE 11. Lithium Deposition Pattern, ng/Bucket (0.2 m diameter), December 5, 1973, Lithium Release 795 g 5000 ft, In Cell, Stable, 2°C.

that deposition would be oriented $\sim 200^\circ$, rather than 180° . At present, we do not have any explanation for this, except that from the drift of the aircraft during the release, a wind direction of approximately 180° was indicated. Thus the radiosonde winds at UIL do not appear to be totally representative for this release to the south. (One should note here that the previously mentioned rhodamine test on March 31, 1972, had possible tracers in an unexpected area.) The initial analysis for all tracers was based on the 200° wind direction for aligning the surface deposition pattern. This is one possible explanation why the analysis of the tracers to date has not shown an organized pattern. Future samples collected to the west will determine whether such organization exists.

When considering the estimated deposition rate for indium as a function of time (see Figure 14) we see a factor of three increase for deposition times of 3000 sec to about 3500 sec. Our analysis is not yet complete, but it appears that the maximum deposition occurs on the last arc at 3500 sec after release. Consequently, no decrease in indium deposition rate with increasing distance was observed. At this time, there has not been developed a totally satisfactory explanation for this increased deposition rate. Again, the same two possibilities for the increased deposition rate are suggested as in the March 11, 1971, case. One of these possibilities, which is being investigated presently, is that an increase in rainfall rate may have increased the deposition. The volume

Neg 745953-14

FIGURE 12. December 5, 1973, Europium Deposition Pattern, Europium ng/BUCKET (0.2 m diameter) (Half Raw, Half Burned), 73 g 5000 ft, In Cell, Stable, 2°C.

of water collected in the samplers was used to compute a mean rate of rainfall (see Figure 15), and, as during the March 11, 1971, storm, the maximum rate of rainfall did correlate with the maximum indium deposition.

From the incomplete indium analysis, the indication is that the gross deposition rate is ~ 0.17 to $0.25\% \text{ min}^{-1}$ as opposed to $\sim 1.5\% \text{ min}^{-1}$ in the March 11, 1971, case. Also it should be noted that the mean rainfall rate in the March 11, 1971, case was approximately three times higher than the December 5, 1973, case.

Neg 745953-8

FIGURE 13. December 5, 1973, Iridium Deposition Pattern, ng/Bucket (0.2 m diameter) Iridium Burner Release 9.7 g (Half Raw, Half Burned) 5000 ft, In Cell, Stable, 2°C .

Neg 745953-12

FIGURE 14. December 5, 1973 Indium Deposition Rate Versus Time.

Neg 745953-13

FIGURE 15. December 5, 1973, Precipitation Rate, g/BUCKET hr.

CONCLUSIONS AND PLANS FOR ADDITIONAL RESEARCH

Organized tracer deposition patterns have been observed in at least three of six storms injected with tracers. Maxima in deposition have occurred on the last arc on March 11, 1971, and December 5, 1973. Two possible causes of these maxima have been suggested:

1. increased deposition via increases in precipitation rate in these areas
2. the process of attachment of tracers to the cloud water may significantly increase the time necessary for the maximum deposition to occur.

The demonstration experiments summarized in this paper have succeeded in determining the feasibility of tracers and experimental methods,

and have given some initial estimates of gross rates of scavenging by frontal storms. The experiments are obviously hampered, however, by inadequate support measurements of storm dynamics and cloud microphysics. This deficiency must be removed before meaningful progress can be made in determining the causal relationships needed for modeling of the associated processes.

Plans have been formulated to remove the above experimental problems by obtaining extensive cloud physics and aerosol measurements from a tracer-release aircraft, and by employing a dual-Doppler radar system to provide detailed wind field measurements in the storm systems. With this advanced experimental program we expect to move rapidly to obtain knowledge of the causal relationships necessary for formulation of reliable frontal-storm models, which will be created in conjunction with investigators at other AEC laboratories.

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A REEVALUATION OF THE USE OF RHODAMINE
FOR IN-CLOUD SCAVENGING STUDIES

R. N. Lee

The tracer rhodamine B was released during the March 31, 1972, in-cloud scavenging experiment. Initial examination of rain samples collected during that test indicated the presence of the fluorescent dye but failed to identify the tracer unequivocally. Reexamination of these samples using a spectrofluorometer indicates that effects related to the presence of particulate matter dominate the analysis at the sub-ppb level. Data reported for the March 21, 1972 test have not been confirmed and future use of rhodamine in the in-cloud scavenging program is not recommended.

INTRODUCTION

Rhodamine, an organic dye frequently employed as an environmental tracer,^{1,2} has recently been applied to the in-cloud scavenging program.³ Rain samples collected on the day of the release have been reexamined in order to evaluate the analytical results reported for the March 31, 1972 test. The quantity of rhodamine dispensed at that time, 0.6 kg in 15 liters of methanol, represents the capacity of the dispersion system. Failure to achieve unequivocal identification of rhodamine indicates its unacceptability as a tracer for in-cloud studies.

CHEMICAL ANALYSIS

Rain samples were first analyzed with a Turner Model 110 Fluorometer. This instrument uses cylindrical cuvettes of 10 mm diameter to hold the sample solutions. Excitation quanta were provided by light from a xenon lamp, modified by passage through narrow pass filters of color specification 1-60 and 58. Rhodamine fluorescence was detected by a photomultiplier tube placed at right angles to the incident light beam. A color specification 23A sharp cut filter was placed between the photomultiplier tube and the sample to isolate light of approximately 580 nm. The close

proximity of the rhodamine absorption and fluorescent maxima (550 and 580 nm, respectively) and the inability of the fluorometer to prevent scattered radiation from reaching the photomultiplier tube introduces difficulties when examining solutions of very low concentration. Field samples are thus subject to positive interference by particulate matter as well as tracer removal via surface adsorption.¹ These errors may be so large as to demand extreme caution in the interpretation of analytical data.

The frozen rain samples were transferred from their original containers (plastic bags) to 250 ml beakers. The bags were rinsed with 10-20 ml of distilled-deionized water and the total sample evaporated under mild heating to ~5 ml. The solution was then transferred to 10 ml volumetric flasks, the beakers rinsed with water and the final solution brought to 10 ml. Aliquots of these solutions were transferred to the cuvettes for analysis. Several fluorescence readings were taken over a period of days in order to identify trends which might relate to the settling of particulates or tracer adsorption. Concentrations were assigned on the basis of the minimum fluorescence readings obtained. These results are recorded in the April 1974 report to the U.S. Atomic Energy Commission.³

Although the reported concentrations are of the magnitude anticipated, the unexpected distribution of samples containing rhodamine and the lack of evidence firmly establishing the presence of rhodamine emphasizes the need for further examination of these solutions. Use of a spectro-

photofluorometer permitted scanning the fluorescence region and therefore identifying rhodamine from its fluorescence spectrum. This work was completed on an Aminco-Bowman Spectrophotofluorometer, which is capable of giving a distinct spectrum for a 0.6 ppb rhodamine solution. Since earlier measurements suggested concentrations which were generally less than 0.3 ppb (3 ng in 10 ml sample), several samples were combined, concentrated to 10 ml and examined for the characteristic rhodamine fluorescence peak at 580 nm. Samples suggested by the previous analysis to contain less than 0.1 ppb tracer were also combined for analysis on the spectrophotofluorometer. This solution was spiked with 12 ng of rhodamine and concentrated to 10 ml. Since it contained particulate material "representative" of the field samples, it was intended to indicate the contribution of surface adsorption to the analytical problem. The compositions of these combined samples are summarized in Table 1.

RESULTS

The absence of a distinct fluorescence maximum at 580 nm for solution 1 has failed to confirm the presence of rhodamine in rain samples collected following in-cloud release. In addition, failure to observe the fluorescence maximum with a spiked sample (5 days after preparation) emphasizes the pronounced influence of surface adsorption on tracer studies employing this material. This study has therefore failed to support our previous findings and illustrates the questionable merits of rhodamine for in-cloud scavenging studies.

TABLE 1. Combined Samples from March 31, 1972, Test.

Combined Solution	Combination of Field Samples	ng Rhodamine Indicated by Previous Work (3)	ng Rhodamine Added
1	18, 29, 26, 429, 432	13	0
2	22, 24, 27, 435	<4	12

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CIRRUS SCAVENGING AS A MECHANISM FOR THE PRODUCTION OF RADIONUCLIDE CONCENTRATION MINIMUMS BETWEEN 6 AND 9 km

J. A. Young, L. Wendell and N. A. Wogman

The chemical and radiochemical composition of atmospheric aerosols is being measured to aid in assessing the present and future hazards to the environment presented by radioactive and nonradioactive contaminants. The rates and mechanisms by which airborne particulates are mixed in the atmosphere, and transported to and deposited on the earth's surface are also under study. One facet of this program involves the measurement of radionuclide concentrations above, below and in cirrus. These measurements indicate that the scavenging of radionuclides by cirrus, followed by the settling of cirrus ice particles, may result in considerable transport of radioactivity.

INTRODUCTION

Since 1967, vertical profiles of radionuclide concentrations have been measured from 1.5 to 3 km to over 18 km in air filter samples collected by RB-57 aircraft. This is part of a program to determine the rates of atmospheric mixing and deposition on the earth's surface. Most of these profiles were measured either near

Albuquerque, New Mexico at 35°N, 107°W; south of Spokane, Washington at 46°N, 117°W; or near Barbados, British West Indies from 12 to 18°N and 54 to 59°W. The most surprising feature of these profiles has been the regular occurrence of concentration minimums between 6 and 9 km. These minimums occurred in about 40% of the samples collected near Albuquerque and Spokane, and 50% of those

collected near Barbados. The radionuclide concentrations in some of these 3 or 6 km samples were only a few percent of the concentrations at lower or higher altitudes.

Some of the less striking minimums could have been due to the predominantly horizontal advection of air from locations of lower radionuclide concentrations. However, it is unlikely that the horizontal advection of unscavenged air could have caused concentrations more than an order of magnitude lower than the concentrations at higher or lower altitudes, since the horizontal concentration gradients are generally not great enough. It is also unlikely that the transport of high-concentration stratospheric air to altitudes below 6 km could have caused apparent minimums at 6 and 9 km, since the concentrations below 6 km were generally normal, and the concentrations at 6 or 9 km were below normal on days that minimums occurred.

It appears that precipitation scavenging is the only mechanism that could have caused the very low concentrations measured at 6 to 9 km. Scavenged air at these high altitudes could have resulted from: 1) deep convective activity, 2) large-scale ascending motions of scavenged air associated with large-scale cyclonic activity, or 3) scavenging by cirrus, followed by the settling of the cirrus ice particles. The northward transport of scavenged air from the towering cumuli of the intertropical convergence zone could possibly have caused the minimums near Barbados. However, deep convective activity is rare near or west of Spokane, especially in the fall or winter when some of the most striking minimums have occurred, so deep convection is unlikely to have caused the minimums near Spokane.

In order to test the possibility that cirrus scavenging can cause concentration minimums, an RB-57 aircraft was used to collect air samples above, below, and in cirrus located near Albuquerque, New Mexico on November 30, 1973; February 19, 1974; and March 13, 1974. Back trajectories are being calculated for the air sampled on these days to determine whether any concentration minimums measured could have actually been caused by cyclonic activity rather than the cirrus.

RESULTS

On November 30, 1973, air samples were collected 3 km, 1.5 km, and 150 km above and below; and then at the center of a cirrus deck located near Albuquerque. The concentrations of the nuclear-weapons-produced radionuclides ^{95}Zr , ^{95}Nb , ^{103}Ru , ^{106}Ru , ^{125}Sb , ^{137}Cs , ^{140}Ba , ^{141}Ce , and ^{144}Ce , and the cosmogenic radionuclide, ^7Be , 1.5 km above the cirrus were an order of magnitude lower than would be expected, based on the concentrations measured in the other samples. The concentrations at the center of the cirrus were an order of magnitude higher than would be expected (Figure 1). The concentrations 150 m above and below the center of the cirrus were near normal, probably because the samples were collected partly in and partly out of the cirrus. The radionuclide concentrations in the sample collected 3 km above the cirrus appeared to be about normal. The 30 trace elements measured in the samples also showed a concentration maximum at the center of the cirrus and a minimum above it. However, the minimum above the cirrus was less pronounced, and the concentrations near the base of the cirrus appeared somewhat enhanced. Correlation studies of the ratios of some of these trace elements are being used to determine if a significant fraction of this dust is of extraterrestrial origin.

The back trajectory for the November 30, 1973, case was followed for 42 hr. The fact that the temperature along the trajectory was a constant -39°C is a good indication that there was no vertical motion along this path. The nearest storm center was over 1000 miles away during the period of this trajectory, and a stationary front through the northwestern states remained well to the north. It is therefore unlikely that the air sampled could have been scavenged by a recent traverse through a cyclonic storm system.

These results strongly indicate that the cirrus was responsible for the scavenging of around 90% of the radionuclides from a layer at least 1.3 km thick and possibly less than 2.8 km thick. It is possible that a thicker layer was scavenged, but that wind shear resulted in the transport

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FIGURE 1. A Comparison of Selected Trace Elements, Fall-out Radionuclides, and Cosmogenic Radionuclide Concentrations as a Function of Altitude on November 30, 1973.

of the scavenged air in this thicker layer to another location.

The concentration 1.3 km above the cirrus of the 15-hr half-life radionuclide ^{24}Na , which is produced at a relatively constant rate in the atmosphere by cosmic rays, was about one-half of the average value measured at that altitude on previous flights (Figure 1). This suggests that the air had been scavenged on the order of 15 hr earlier and had had time to grow back to 50% of the equilibrium. Since the measured concentrations have varied from flight to flight, this calculated time should not be taken as absolute. A settling rate of 1.3 km in 15 hr equals 2.4 cm sec^{-1} . It is interesting that, in contrast to the other radionuclides, the ^{24}Na concentrations within the cirrus were considerably below normal. This could indicate that the cirrus had scavenged radionuclides in considerable quantities earlier, but that at the time of sampling the scavenged ^{24}Na had mostly decayed away, and that large radionuclide-bearing ice particles were settling out of the cirrus at a rate sufficient to lower the ^{24}Na concentration in the cirrus below its production rate. The ^{24}Na concentration 2.8 km above the cirrus appeared to be somewhat depressed, suggesting that even at this altitude

the air might have been partially scavenged by the cirrus.

On February 19, 1974, a second vertical profile through a thick cirrus deck near Albuquerque was measured. The radionuclide concentrations at the center of the cirrus and right at the top of the cirrus appeared to be somewhat depressed (Figure 2). Vertical profiles measured at that location on previous flights have generally shown an exponential concentration increase with altitude. Unfortunately, the sample taken at the top of the cirrus was taken partly in clouds, and the sample collected 1.5 km above the cirrus showed no evidence of a concentration depression. It is possible that there could have been a layer of very low concentrations less than 1.5 km thick right above the cirrus. The low concentrations within the cirrus could indicate that larger ice particles were settling out of the cirrus, taking radionuclides with them.

Two back trajectories were constructed for February 19, the first starting at the beginning of the sampling period and the second starting at the end. The later trajectory runs farther south than the earlier one because of the influence of an advancing wave in the upper-level flow, associated with a low center and an eastward moving cold front. However, the trajectory positions were ahead

of the surface frontal positions and therefore should have been well ahead of the front at higher levels. The storm center was well to the north of both trajectories, and the temperatures were constant along them, indicating very little vertical motion. Therefore, recent scavenging of the sampled air by a traverse through a cyclonic storm was not likely.

On March 13, 1974, another vertical profile was taken, this time through two cirrus decks. The radionuclide concentrations 0.3 km and

possibly 0.9 km above the lower cirrus deck and those within the upper cirrus deck appeared to be somewhat depressed (Figure 3). The concentrations 1.2 km above the upper cirrus deck did not appear to be below normal.

CONCLUSIONS

The profile taken November 30, 1973, indicates that the scavenging of radionuclide-bearing aerosol particles by cirrus may result in considerable downward transport of radioactivity. However, the evidence for

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FIGURE 2. A Comparison of Selected Fallout Radionuclides and Cosmogenic Radionuclide Concentrations as a Function of Altitude on February 19, 1974.

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FIGURE 3. A Comparison of Selected Fallout Radionuclides and Cosmogenic Radionuclide Concentrations as a Function of Altitudes on March 13, 1974.

cirrus scavenging was much less clear-cut on the February 19 and March 13, 1974 flights. It is difficult to estimate the net downward transport of radioactivity which might result from cirrus scavenging, since the frequency of cirrus scavenging, the horizontal extent and thickness of the scavenged layer, and the distance the radionuclides might be transported downward by the ice particles are difficult to determine. The results from the November 30 flight, however, indicate that 90% of the radionuclides were scav-

enged from a layer at least 1.3 km thick on that date. Samples collected near Barbados at altitudes differing by 600 m at around 9 km often showed the same very low concentrations, while other samples collected 3 km above or below these samples showed some evidence of depressed concentrations. Samples collected at each of the corners of a 5° latitude and longitude square east of Barbados often showed the same very low concentrations at 9 km, showing that the scavenged layers affect a large horizontal area.

THE ENTRAINMENT OF TRACERS NEAR THE SIDES OF CONVECTIVE CLOUDS

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

The Metropolitan Meteorological Experiment (METROMEX), in addition to the study of city atmospheric concentrations and downwind concentration decreases of trace elements, sulfates, SO₂, freons and CO in St. Louis, also involves an experiment on entrainment of tracers near the sides of convective clouds. Inert tracers are released near the sides of these convective clouds, and followed to determine if the air at these locations is entrained and reaches regions of precipitation scavenging. Iridium, rhenium, gold, europium, and tantalum were released sequentially at 15,000 to 16,000 ft near the edges of convective towers on August 11, August 17, and August 18, 1974. Some of the precipitation samples collected on August 17, 1974 have been analyzed for rhenium and gold, and the measured concentrations were significantly above background.

Tracers were released sequentially at 15,000 to 16,000 ft near the sides of convective clouds on August 11, 17, and 18 in 1974 (Table 1). Following the tracer releases, samples of rain were collected for analysis using a 40 by 50 mile network of 187 rain collectors located east of St. Louis. The samples are now being analyzed for tracers and other elements by neutron activation.

On August 11, convective cells developed over St. Louis. Iridium, rhenium, gold, europium, and tantalum were released sequentially at 15,000 ft over a 30-min period, near the western edge of one cell as it moved eastward over the precipitation network. Each individual tracer was released over a 2-min period. Indium and lithium were also released into

TABLE 1. Tracer Releases

Tracer	Start Time (CDT)	Burn Time (min)	Troy, Ill. Coordinates	Altitude (k ft)	Approximate Grams Released
August 11, 1974					
Iridium	1520	2.5	242°/19	15	12
Rhenium	1525	2.5	242°/19	15	105
Gold	1535	2.5	242°/19	15	153
Europium	1551	2.5	204°/11	15	71
Tantalum	1556	2.5	203°/9	15	229
August 17, 1974					
Tantalum	1811	2.5	198°/18	15	229
Iridium	1819	2.5	198°/14	15	8
Rhenium	1826	2.5	183°/13	15.5	70
Europium	1833	5	183°/13	16	71.2
August 18, 1974					
Iridium	1704	2.5	073°/17	15	5
Rhenium	1713	2.5	073°/17	15	35
Tantalum	1724	2.5	020°/10	15	458
Gold	1736	5	073°/12	15	153

the updraft of this cell by the Illinois State Water Survey. Unfortunately, this cell died soon after it reached the network, and only around a dozen rain samples were collected from it. However, convective cells later formed east of this cell, and it is possible that the tracers may have entered these cells. Several rain samples were collected from these cells near the eastern edge of the precipitation network.

On August 17, a series of convective cells oriented in an east-west line developed just south of the precipitation collection network. Since these cells were drifting east-south-east, it did not appear at first that they could possibly reach the precipitation network. However, a cell eventually developed on the northern edge of the line which looked as though it might cross the southwestern corner of the network. The Battelle aircraft was therefore sent up to investigate. By the time the Battelle aircraft reached the area this cell had died, but another had developed over the southwestern part of the network, and was moving eastward. Tantalum, iridium,

rhenium, and europium were released at 15,000 to 16,000 ft near the edge of this cell over a 27-min period. Following the release of the tracers a considerable number of rain samples were collected from the southern portion of the network. It is quite possible that this cell was unaffected by the St. Louis metropolitan area.

On August 18, an east-west oriented line of convective cells again developed and moved east-southeast, but this time to the north of the precipitation network. It appeared at first that the cells would miss the northeast corner of the network by a considerable margin. However, one large cell began to veer southward, and since it appeared that this cell might cross the northeast corner of the network, the Battelle aircraft was sent up to investigate. Before the aircraft reached the area, a convective cell developed over the north central part of the network and moved south southeastward, so iridium, rhenium, tantalum, and gold were released near the edge of this cell at 15,000 ft over a 38-min period. A considerable number of rain samples

were collected from this cell following the tracer releases. This cell was probably influenced by St. Louis or Alton-Wood River.

Sixteen of the rain samples collected on August 18 in the region of maximum expected deposition have been analyzed for rhenium and gold. The measured rhenium and gold concentrations are shown in Figures 1 and 2, respectively, along with the posi-

tions of the other precipitation collectors. Calculations indicate that 50% of the rhenium and 5% of the gold released were deposited in this region. The lower gold deposition could have been caused by release of the gold 23 min after the rhenium release, at a time when the convective cell showed signs of weakening. It is also possible that the maximum gold deposition occurred in a different region.

FIGURE 1. Rhenium Deposition in 10^{-12} g cm^{-2} on August 18, 1974.

FIGURE 2. Gold Deposition in 10^{-12} g cm^{-2} on August 18, 1974.

PRECIPITATION SCAVENGING IN A SQUALL LINE:
NUMERICAL EXPERIMENTATION

Carl E. Hane

A squall line thunderstorm numerical model is described. The model is two-dimensional and time dependent with parameterized microphysics. The model is applied to an actual squall line situation which occurred on August 21, 1972, over St. Louis, Missouri. An aperiodic series of developments occurs in the model calculations with maximum updraft speeds during developments reaching ~ 25 m/sec. A time is picked when a typical circulation pattern is occurring. The motion field, cloudwater distribution, and rainwater distribution are assumed steady for purposes of precipitation scavenging calculations. A pollutant plume is allowed to enter the storm circulation from various locations and heights surrounding the storm and the resulting redistribution of pollutant is examined with the assumptions: 1) the pollutant travels with the air and does not interact with the liquid water, and 2) the pollutant enters the rain and/or cloud to varying degrees and is carried to the ground. The possibility of including the pollutant in a much less passive way in the storm circulation (i.e., inclusion in the time-dependent equations) is desired.

As a part of the on-going research effort in precipitation scavenging by the Atmospheric Sciences Department, a numerical model of squall line convection has been utilized to estimate internal cloud circulation patterns. This information is being used to study incorporation of pollutants into such clouds from various locations relative to the cloud. In addition, various environmental profiles of pollutant concentration are being assumed in order to include the possible range of natural occurrences. The initial effort investigates the redistribution of such pollutants by the cloud air motions with no interaction considered among pollutant, rain and cloudwater. A second step studies the interaction of pollutant concentrations in the atmosphere and in the fallen rainwater. A third step will involve the inclusion of pollutant in the cloudwater.

The model used to calculate air motions and other cloud properties is a two-dimensional time-dependent squall line thunderstorm model. The model will not be described in depth here; model equations and other details may be found in an earlier paper.¹ The model variables are the two components of air motion in the plane of propagation of the storm, the air temperature, and the mixing ratios of water vapor, cloudwater, and rainwater. Integrations are carried out over a rectangular grid with total dimensions of 25.6 km in the horizontal and 12.8 km in the vertical, with 400 m grid intervals in both the horizontal and the vertical. The general procedure for running the model is to begin the calculations with no cloud present, superimposing a perturbation in the motion field upon the observed environmental winds for the case in question. The observed vertical distributions of temperature and moisture

for the case in question are applied to the entire horizontal extent of the grid in the initialization. Important boundary conditions include no vertical motion through the upper or lower boundary, and time invariant vertical distributions of horizontal motion along the lateral boundaries. The calculations are done in a relative horizontal wind field; i.e., the motion of the storm is subtracted from the total motion field.

The squall line thunderstorm model has been applied using the environmental conditions existing in the St. Louis area on August 21, 1972. The storm which occurred on that day possessed several squall line characteristics. The rainfall pattern is shown in Figure 1, as measured by the raingage network operated by the Illinois State Water Survey. The analysis is done at 0.25 in. intervals beginning at 0.25 in., except in the south St. Louis area where

isolines representing values greater than 1 in. are 1.5, 2.0, and 3.0 in. The heavy lines show the positions of rain sampling arcs employed for precipitation scavenging measurements. Amounts in excess of 3 in. were measured in southeast St. Louis and directly across the Mississippi River in Illinois. The elongated rainfall maximum in the northern section of the area resulted from the movement of a separate cell toward the east-northeast. The rainfall pattern in the south part of the city represents the movement of individual cells in an easterly direction, but with development of new cells quasi-continuously on the right flank. This resulted in a propagation toward the southeast.

The wind, temperature, and moisture profiles were measured at approximately 1400 CST, a few hours prior to the storm's occurrence. The

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FIGURE 1. Rainfall (Inches) for 21 August 1972 Storm. The heavy lines show the positions of rain sampling arcs employed for precipitation scavenging measurements.

winds which were used in the two-dimensional model calculation were the components of the wind in the direction of storm motion [taken to be from the northwest (315°)]. The resulting wind profile is shown in Figure 2. The wind speeds plotted as a function of height in this figure are speeds relative to the ground. Actually two wind profiles were used in the model calculations, one profile to the left and one profile to the right of the storm, so as to produce low level convergence and upward motion. The profile shown in Figure 2 is actually the mean of these two profiles. The temperature, water vapor mixing ratio, pressure, and air density are shown as functions of height in 400 m increments in Table 1.

The model calculations were initiated by allowing the area of upward motion to produce a convective cloud. The evolution of the cloud is represented by the time variations of several parameters shown in Figure 3. Included in the figure are the time evolution of maximum upward motion,

maximum downward motion, and maximum rainwater mixing ratio. The model was run for about 60 min until a fairly regular oscillatory time evolution of the cloud circulation was obtained. A time was chosen (about 55 min) when a typical cloud circulation was occurring. The motion field, rainwater mixing ratio distribution and cloudwater distribution were then assumed to be fixed in time, for purposes of experimentation with pollutant inclusion into the storm. The stream function representing the motion field is shown in Figure 4. Shading in the figure is in increments of 2000 kg/m-sec; arrows indicate direction of air motions and closer spacing of isolines indicates higher wind speeds. A pollutant distribution was then introduced in the lower 2 km of

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FIGURE 2. Component of the Wind in the Plane of Motion of the Storm as a Function of Height for the 21 August 1972 Storm.

TABLE 1. Variables of State and Water Vapor Mixing Ratio as a Function of Height Used as Environmental Conditions for Model Calculations. Based upon observations taken at Lambert Field, St. Louis, Missouri on 21 August 1972 at 1417 CST.

Ht. (km)	P (mb)	T ($^\circ$ C)	q_v (g/kg)	ρ (kg/m ³)
0.0	990.	35.2	15.542	1.1082
0.4	947.	30.4	14.526	1.0775
0.8	905.	26.0	13.305	1.0458
1.2	865.	21.6	12.193	1.0147
1.6	826.	17.5	11.503	.9828
2.0	788.	14.0	11.216	.9492
2.4	751.	11.8	8.691	.9134
2.8	716.	8.7	6.264	.8812
3.2	682.	5.7	4.429	.8492
3.6	649.	4.2	4.238	.8130
4.0	618.	2.7	3.825	.7782
4.4	588.	1.1	3.317	.7450
4.8	559.	-1.1	2.707	.7146
5.2	532.	-3.6	2.182	.6861
5.6	505.	-6.9	1.844	.6600
6.0	480.	-9.8	1.511	.6339
6.4	455.	-12.3	1.187	.6075
6.8	432.	-14.6	.896	.5816
7.2	409.	-16.7	.664	.5559
7.6	388.	-18.7	.448	.5312
8.0	368.	-21.1	.377	.5080
8.4	348.	-23.5	.307	.4857
8.8	329.	-25.9	.251	.4641
9.2	312.	-28.3	.207	.4433
9.6	295.	-30.7	.163	.4233
10.0	278.	-33.5	.131	.4045
10.4	263.	-36.3	.104	.3865
10.8	248.	-39.1	.077	.3690
11.2	234.	-42.2	.057	.3525
11.6	220.	-45.4	.045	.3368
12.0	207.	-48.7	.033	.3216
12.4	195.	-52.0	.021	.3069
12.8	183.	-54.2	.015	.2914

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FIGURE 3. Time Evolution of Maximum Upward Motion (m/sec), Maximum Downward Motion (m/sec), and Maximum Rainwater Mixing Ratio for the Model Storm.

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FIGURE 4. The Motion Field Within the Model Storm Represented by the Stream Function (kg/m-sec).

the atmosphere upwind from the storm. The pollutant was then allowed to travel with the air motion (motion field is that relative to the moving storm, translational speed has been removed) and not allowed to interact with the rain or cloud. The distribution was allowed to enter through the right boundary for a limited amount of time corresponding to the

finite width of a pollutant plume over an urban area (~22 km in this case).

The resulting redistribution of the pollutant by the storm circulation is shown in Figure 5 at four different times. The shading interval here is 5×10^{-8} g/g. It can be seen that most of the pollutant is transported to the upper atmosphere while another significant part is retransported to

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FIGURE 5. The Pollutant Distribution at Four Different Times Following Its Insertion into the Storm Environment: Upper Left, Time = 0 min; Upper Right, Time = 30 min; Lower Left, Time = 50 min; Lower Right, Time = 70 min.

lower levels in the storm downdraft. This portion which is transported to the lower levels would have ample opportunity to be scavenged by cloud and rain since it traverses both the updraft and downdraft. The portion of the pollutant which remains in the upper levels might not be scavenged so effectively. An examination of the circulation pattern within the storm reveals that the portion of the downdraft air having its source in the low-level inflow originates in the lowest levels of that inflow prior to its incorporation into the storm. Therefore, if it is true that pollutant traversing both the updraft and downdraft is scavenged most effectively, the fraction of pollutant which is ultimately scavenged depends to a large extent upon the vertical distribution of pollutant prior to its entry into the storm.

In order to investigate the effect of this vertical distribution factor upon the amount of pollutant entering the downdraft area as a result of air motions only, the vertical distribu-

tion was varied in a series of cases. The profiles are shown in Table 2.

The results of Case I at four times following initiation have been shown in Figure 5. The results of the other cases bear great similarity to Case I in the distribution of pollutant at these times; the most noticeable differences occur near the ground where the downdraft air delivers its pollutant load. Table 3 shows the relative amounts of pollutant in this region near the ground for the four cases considered. The amounts given are the total pollutant (in grams) in the layer between 0.2 and 1.8 km for each meter slab thickness. As the table indicates, that vertical profile with pollutant maximum near the ground results in the maximum total pollutant in the low-level downdraft air.

The next step in this ongoing research is to determine the extent to which the pollutant traversing various parts of the storm will be scavenged by hydrometeor water (below cloud scavenging). The term below

TABLE 2. Concentration Profiles Used in Model Experiments

Height (m)	Mixing Ratio (10^{-8} g/g)			
	Case 1	2	3	4
0	12	30	0	17
400	18	27	12	17
800	27	18	14	17
1200	30	14	18	17
1600	14	12	27	17
2000	1	1	30	17
2400	0	0	0	0

TABLE 3. Low-Level Pollutant Amounts as a Function of Initial Pollutant Vertical Profile

Case	Total Pollutant (g/m)
1	1328
2	1529
3	864
4	1210

cloud scavenging is somewhat misleading here, since this process also occurs within the cloud wherever liquid water travels at a speed other than that of the air in which it is located.

The means whereby this type of scavenging is taken into account will now be described. The mechanism is taken here to be the same as that whereby rain droplets coalesce with small cloud droplets. The concept is rather simple; the rate of incorporation of pollutant into the rain in a given volume of air is determined by the volume swept out by the assumed distribution of raindrop sizes. All pollutant in the swept out volume is assumed to be incorporated, i.e., the collision efficiency is unity. The resulting amounts of scavenged material should therefore be considered an upper limit, since collision efficiencies are in general less than unity.

In the squall line thunderstorm model which has been described here, the Marshall-Palmer (exponential decrease with diameter) distribution is assumed. The accretion term resulting from this assumed distribution will not be derived here, but the term itself may be written:

$$\left(\frac{dq}{dt}\right)_s = c\rho Q/\lambda^{3.5}, \quad (1)$$

where $c = 3.08 \times 10^2$ is a dimensioned constant, and

$$\lambda^4 = 2.51 \times 10^2/q_\ell. \quad (2)$$

The variable q_ℓ is the hydrometeor water mixing ratio. The above term is calculated at the midpoint of the trajectories of pollutant-bearing rain and pollutant-bearing air in the Lagrangian type scheme which is used for the time integrations. The term adds pollutant to the rain at a given point and subtracts it from the air, but by differing amounts, since in general the air and rain at a point arrive along different trajectories. An additional factor, the possibility of total evaporation of the rain and release of pollutant to the air, has so far been ignored.

A quantity which will result from these calculations is the total amount of pollutant in the rain which reaches the ground. Observations are available for comparison with these amounts (e.g., the downstorm variations of cross-storm averaged measurements of total pollutant might be compared with the calculated values). A meaningful comparison, however, cannot be made until experiments are conducted making measurements of scavenged material and the vertical profile of pollutant concentrations characteristic of the low-level inflow to the storm. Until this is done, the best method that might be employed is probably to run experiments over a wide range of possible pollutant profiles and to examine whether observations agree with any values within this range.

Results from the above-described second step are not available at the time of this writing, and work on a third step of this research has not begun. The third step will involve a parameterization of the pollutant interaction with the cloudwater (i.e.,

in the role of a nucleus for condensation or as a dissolved gas). This step is quite major compared to the steps described above, and will probably necessitate recalculating the full set of model equations, with additional time-dependent equations for pollutant concentrations in various media. The same comments concerning comparison of ground level amounts with observed amounts and distribu-

tions apply to this phase of the research.

ACKNOWLEDGEMENTS

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ANALYTICAL INVESTIGATIONS OF INERTIAL DEPOSITION OF
SMALL AEROSOL PARTICLES FROM LAMINAR FLOWS
ONTO LARGE OBSTACLES

PART C - POWER SERIES EXPANSIONS
AND THE CRITICAL STOKES NUMBERS

W. G. N. Slinn

Power series solutions to the particle momentum equation are sought to elucidate the nature of the solution and the numerical value of the critical Stokes number. This method yields Taylor's values for potential flows and suggests that a critical Stokes number for Stokes flow about a sphere does not exist. It is seen that power series methods have limited value because of the multivalued nature of the solution at large Stokes numbers. Empirical expressions for the critical Stokes numbers obtained from numerical analyses are used in a semi-empirical expression for the inertial impaction collision efficiency for flow about a sphere.

INTRODUCTION

Part A of this report¹ showed that regular perturbation techniques failed to yield solutions to the problem of inertial collection. In Part B, using the method of matched asymptotic expansions, solutions were found for the cases of potential flow about a sphere and a cylinder at large Stokes numbers. Further there appeared to be no restrictions to the method for applications to arbitrary flows about arbitrary obstacles. However, the labor involved rapidly becomes prohibitive. To circumvent these calculations and to seek an analytical continuation for all Stokes numbers, an investigation has been made into the analytical behavior of the solution at small Stokes numbers. This report summarizes results of studies of power series solutions for particle motion along the upstream axis of symmetry. In Part D a different technique will be explored for the small Stokes number case.

Incidentally, subsequent to these studies, C. N. Davies (personal com-

munication) has called the author's attention to investigations of a similar nature described in Davies' book on air filtration.² However, there it was improperly assumed that regular perturbation techniques could be used (see the equation above his Equation [67]). This leads Davies and the original authors, Stechkina, Kirsch and Fuchs³ to untenable and incorrect conclusions. In reality it is not the equations which fail, as they suggest, but (as was shown in Part A) the method of solution via regular perturbation theory. Further, they ignored the additional problems caused by the existence of critical Stokes numbers below which there is no inertial collection. This topic will now be addressed.

COMMENTS ON THE CRITICAL
STOKES NUMBERS

In a number of recent reviews⁴⁻⁶ Taylor's original analysis⁷ of the critical Stokes number problem is presented. Here only a brief outline will be given. For particle motion along the upstream axis of

symmetry or for any flow near a stagnation point the steady-state momentum equation (see Part A) reduces to the simple formulation of Newton's second law (nondimensionalized)

$$s u \, du/dz = s \ddot{z} = v - u \quad (1)$$

where $s = \tau U_0/R$ is the Stokes number, $u = \dot{z}$ is the particle's velocity, v is the fluid velocity and z is positive in the direction of the fluid velocity at infinity. Taylor approximated the fluid velocity near the stagnation point by

$$v = -c\rho = -c(z-1) \quad (2)$$

where $c = 1, 2, 3, 4/\pi$ for potential flow past a plate, cylinder, sphere and disk, respectively. Upon substituting (2) into (1) and using $u = \dot{z} = \dot{\rho}$ there results

$$s \ddot{\rho} + \dot{\rho} + c\rho = 0 \quad (3)$$

which is the same equation as for a damped harmonic oscillator. If a solution to (3) of the form $\exp(-\lambda t)$ is assumed then the indicial equation gives the roots

$$\lambda_{1,2} = \frac{1}{2s} \left[-1 \pm (1-4cs)^{1/2} \right] \quad (4)$$

whose behavior as s is varied is shown in Figure 1. "Critical damping" occurs at $s_* = 1/4c$ and these are the critical Stokes numbers.

Further discussion of this solution in the above-referenced reviews perhaps could be improved. All that need be done now, without the need to discuss specific initial conditions,⁴ is to note that if $s > s_*$ (i.e., small damping) then this "harmonic oscillator" can oscillate through the origin or, returning to the terminology of the problem of interest, the particle will collide with the obstacle. On the other hand, for $s < s_*$, the damping is so large that the oscillator's amplitude will just exponentially decay as it approaches the origin, and the oscillator's velocity will be zero at $\rho = 0$. Therefore the inertial contribution to the collision efficiency will be zero for $s < s_*$. Referring to the fact that in this case it requires an infinite time for the particle to reach the origin⁴ appears to be an unnecessary source of confusion, especially without mention that it also requires an infinite time for the fluid in potential flow to reach the obstacle along the upstream axis of symmetry. This is easily checked, for example by integrating $\dot{\rho} = -c\rho$.

Notice, however, it is not correct to infer that for $s < s_*$ no particles will be collected by the obstacle. If the above analysis is correct it implies only that there is no inertial contribution to collisions for $s < s_*$; the analysis applies only for particles with mass but no size. If the particles are spheres of radii a , then a particle will collide with

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FIGURE 1. An Indication of the Behavior of the Roots of the Indicial Equation as the Stokes Number Varies, for the Linearized Inertial Collection Problem for Potential Flow about Various Obstacles.

the obstacle along the upstream axis of symmetry when the particle's center is within a distance a of the obstacle's surface. This leads to a finite, nonzero collision efficiency as is shown elsewhere.⁸ Also, a particle of finite size is influenced by the fluid velocity not only at one point but by the mean fluid velocity over the volume occupied by the particle. (Obviously, though, one should evaluate the disturbance on the flow field caused by the particle, but this has been ignored. An equation that accounts for this effect is given in Fuchs⁴, p. 70.) Consequently even for $s < s_*$, a particle travelling along the upstream axis of symmetry will acquire the mean velocity of the fluid evaluated a distance a off the axis. This obviously results in a minimum collision efficiency of $\pi a^2 / \pi R^2$; i.e., $E \geq \kappa^2$, where $\kappa = a/R$ is the interception parameter. Further, in addition to interception, a host of other processes even in laminar flows can promote collisions; for example, particle diffusion, Coulombic and polarization forces and under appropriate conditions, thermo- and diffusio-phoresis. Therefore it should be kept in mind that although $s < s_*$ may imply no inertial contributions to collisions, still collisions can occur.

Nevertheless, since inertial contributions to collision efficiency are typically very large for $s > s_*$,⁸ it is important to ascertain s_* as accurately as possible. As an illustration of the above statement, for a 10 μ m particle colliding with a 1 mm raindrop, $E_M = \mathcal{O}(1)$, $E_N = \mathcal{O}(10^{-2})$ and $E_D = \mathcal{O}(10^{-6})$ where subscripts on the collision efficiency E stand for inertial impaction, interception, and diffusion, respectively. Consequently it is of interest to seek refinements and extensions to Taylor's analysis. As a start towards this, it is interesting to notice that s_* may even be larger than the values obtained by Taylor because, returning to the harmonic oscillator terminology, he has assumed that the restoring force extends to infinity and that it always increases linearly with the displacement. Instead, if use is made of the full expression for the fluid velocity, it is seen that it is a "soft spring" whose restoring force is limited. Physically it seems rea-

sonable that this overestimation of the restoring force may overestimate s_* . It is also of interest to find s_* for other than potential flows; physically, since potential flow is a fairly good approximation for the upstream flow at large Reynolds numbers, one would expect s_* to be larger for flows at smaller Reynolds numbers. Some of these topics will now be addressed.

COORDINATE EXPANSIONS FOR POTENTIAL FLOW

In Parts A and B of this report approximate solutions were sought which were expansions in the parameter s . Because of the difficulties encountered, especially at small Stokes numbers, it seems reasonable to try alternative expansions. One such was sought by Michael and Norey⁹ who tried the coordinate expansion.

$$u(s, z) \sim u_0(s) + u_1(s)z^{-1} + u_2(s)z^{-2} + \dots \quad (5)$$

If (5) is substituted into (1) and coefficients of similar powers of z are equated, then for potential flow about a sphere one obtains

$$u(s, z) \sim 1 + \frac{1}{23} \left\{ 1 + \frac{3!}{2} \frac{s}{z} + \frac{4!}{2} \left(\frac{s}{z} \right)^2 + \frac{5!}{2} \left(\frac{s}{z} \right)^3 + \dots \right\} + \frac{3s}{27} \left\{ 1 + 14 \left(\frac{s}{z} \right) + \dots \right\} + \dots \quad (6)$$

which corrects an apparent typographical error in their result. From (6) it is seen that a solution of the assumed form is not a converging power series in s ; however the correct number of terms can be retained so that it can be a useful asymptotic expansion in $(1/z)$ when $z < 0$. Incidentally, it is interesting that (6) is essentially a regrouping of the expansion which can be found by seeking a power series in s :

$$u(s, z) = v \left[1 + \frac{3s}{z^4} + \frac{6s}{z^5} \left(2 + \frac{5}{z^5} \right) + O(s^2) \right] \quad (7)$$

This result is zero at $z = -1$ for all s and therefore neither (6) nor (7) are particularly useful to illuminate s_* .

The above coordinate expansion does however suggest that a coordinate expansion be sought in powers of ρ , where $\rho = z + 1$ ($\rho = 0$ at $z = -1$). Then the governing equation is

$$su \frac{du}{d\rho} + u = \sum_{k=1}^{\infty} v_k \rho^k \quad (8)$$

where for potential flow about a sphere,

$$v_k = -(k+1)(k+2)/2 \quad (9)$$

and the series for v converges uniformly in $|\rho| < 1$. If a solution to (8) is sought of the form

$$u(s, \rho) = \sum_{k=0}^{\infty} u_k(s) \rho^k \quad (10)$$

then from equating the coefficient of ρ^0 there results

$$su_0 u_1 = -u_0 \quad (11)$$

in which there is the option to take $u_0 = 0$ and u_1 undetermined or u_0 arbitrary and $u_1 = -1/s$.

Consider first the case $u_0 = 0$. Then the coefficient of ρ^1 gives

$$u_1(s) = [-1 \pm (1 - 12s)^{1/2}] / 2s \quad (12)$$

In (12) it is again seen, as might have been expected, that the value $s = s_* = 1/4c = 1/12$ has special significance; indeed, unless $s \leq s_*$, then u_1 as given by (12) is a complex number. This apparently should be interpreted to mean that there is no solution of the assumed form when $u_0 = 0$ and $s > s_*$. This is a desired result because $u_0 = 0$ implies there is no collection (at $\rho = 0$). For $s \leq s_*$, then proceeding to higher order terms there results (for $k \geq 2$),

$$u_k(s) = \frac{v_k - s \sum_{i=2}^{k-1} i u_i u_{k+1-i}}{1 + s(1+k)u_1} \quad (13)$$

which gives the remaining terms in the solution. In particular, for $s = 0$, (13) gives $u_k = v_k$ and therefore $u = v$ provided the positive sign is chosen in (12). For $s = s_*$, (12) in (13) leads to

$$u(z, s = s_*) \sim -6\rho + 12\rho^2 + 34\rho^3 + \frac{370}{3}\rho^4 + \dots \quad (14)$$

which is zero at $\rho = 0$. In summary, for $u_0 = 0$, there is a solution of the assumed form provided $s \leq s_*$ and with $s \leq s_*$, no particles are collected by the sphere.

On the other hand, for $u_0 \neq 0$, then (11) gives $u_1 = -1/s$ and u_0 is arbitrary. Further substitution and reduction leads to

$$u(s > s_*, \rho) \sim u_0(s) - \frac{1}{s}\rho - \frac{3}{2su_0}\rho^2 - \frac{1}{su_0} \left(2 + \frac{1}{su_0} \right) \rho^3 + \dots \quad (15)$$

which apparently requires s to be sufficiently large if only a few terms are to be a useful approximation. But there is a major inadequacy of (15); namely, that u_0 is unspecified. Certainly it is important since it essentially gives the collision efficiency: $u(s > s_*, \rho = 0) = u_0$. Therefore it is worthwhile to try other methods which might establish u_0 .

One way that might be tried to establish u_0 is to return to a parameter expansion. However, if use were made of

$$u(s, z) \sim u_0(z) + u_1(z)s^{-1} + u_2(z)s^{-2} + \dots \quad (16)$$

($s \rightarrow \infty$), it would fail, just as failure was encountered in the regular perturbation analysis of Part A. Instead the uniformly valid result should be used; i.e., the solution to

$$U \frac{dU}{dZ} = V - U \quad (17)$$

of the form

$$U(s, Z) \sim U_0(Z) + \sigma^3 U_3(Z) + \sigma^6 U_6(Z) + \dots \quad (18)$$

where $Z = \sigma z$ and $\sigma = s^{-1} \rightarrow 0$. If (18) is substituted into (17) and the result evaluated at $z = -1$ then one obtains

$$U_0 = 1 \quad (19)$$

$$U_3 = -\langle e^\sigma / \sigma \rangle E_3(\sigma) \quad (20)$$

$$U_6 = -\frac{1}{2} \left[e^{-\sigma} U_3^2 + \int_0^\infty \frac{e^\xi}{\xi^4} E_3^2(\xi) d\xi \right] \quad (21)$$

etc. These results are interesting, in particular the expansion of the exponential integral E_3 to see the occurrence of the log terms:

$$\sigma^3 U_3 \sim -\sigma e^\sigma \left[\frac{1}{2} - \sigma + \frac{\sigma^2}{2!} \left(\frac{3}{2} - \gamma - \ln \sigma \right) + \frac{\sigma^3}{3!} - \frac{\sigma^4}{2 \cdot 4!} + \frac{\sigma^5}{3 \cdot 5!} - \dots \right] \quad (22)$$

but the results are also overwhelmingly complex.

Instead of proceeding in this manner it is suggested that (16) be used, recognizing that it fails for large z , and that after the σ^3 term there appears a term $(\sigma^3 \ln \sigma)$. Then one obtains

$$u(s, z) = 1 - \frac{1}{2z^2} - \frac{1}{2z} \left(1 + \frac{1}{4z^3} \right) \frac{1}{s^2} + \dots \quad (23)$$

which is the same result as obtained in Part B (except here the coordinate system is reversed and $\theta = 0$). For small z , let $\rho = z + 1$ and expand (23):

$$u(s, \rho) \sim \left[1 - \frac{1}{2s} + \frac{3}{8} \frac{1}{s^2} + \frac{11}{48} \frac{1}{s^3} + O\left(\frac{1}{s^3} \ln \frac{1}{s}\right) + \dots \right] + O(\rho). \quad (24)$$

Upon comparing (24) with (15) it is seen that this ploy has yielded the first few terms of $u_0(s)$. It is interesting that the first three terms of $u_0(s)$ are the same as those in the expansion of

$$u_0(s) = \frac{s^{1/2}}{s+1}, \quad s \gg 1. \quad (25)$$

However, this procedure has not really helped us much because the results (24) and (25) are useful only for $s \gg 1$. Thus they do not help establish the form of the solution, and in particular of $u_0(s)$, for s near s_* . In Part D a different technique, the Oseen uniformization method, is used and it is found to yield more valuable results. In Part E our exploration of the use of the methods of strained coordinates and of multiple scales will be described.

CRITICAL STOKES NUMBERS FOR OTHER FLOWS

The above was for the case of potential flow, especially potential flow about a sphere, but this latter restriction was unnecessary. It is also of considerable interest to seek the critical Stokes number for other flows. Some of the new difficulties that enter can be seen in the case of Stokes flow about a sphere. If Taylor's method is utilized then the appropriate approximate equation is

$$s\ddot{\rho} + \dot{\rho} - \frac{3}{2}\rho^2 = 0. \quad (26)$$

This is not the equation for an oscillator since the "restoring force," $(3/2)\rho^2$, is unidirectional, always forcing the particle in the direction of the free stream. Thus for no case will the motion be oscillatory near $\rho = 0$.

The same failure of this method will appear for all flows which do not portray a restoring force. With this failure of Taylor's method it seems reasonable to try the method developed here which led to (12) for the case of potential flow. Thus if a solution of the form $u = \sum u_k \rho^k$ is tried in the momentum equation (1) there results

$$s \sum_{\ell=1}^{n+1} \ell u_\ell u_{n+1-\ell} = v_n - u_n \quad (27)$$

FIGURE 2. An Indication of the Behavior of u_1 (in $u = \sum u_n p^n$) as the Stokes Number and as v_1 vary.

or not collisions occur, i.e., whether (33), (which predicts collisions) or whether (31) or (32) (which do not), are valid thus appears to depend only on the particle's initial velocity. Clearly this suggestion that a critical Stokes number does not exist for Stokes flow deserves further study. This will be done in Parts E and F.

For practical applications the best that we can now provide for the inertial contribution to the collision efficiency is derived from an empirical fit through the numerical results for s_* ^{8,10}. Two such, for flow about a sphere, are shown in Figure 3. For practical applications in the precipitation scavenging problem it is of little significance whether s_* for Stokes flow is infinite or finite (e.g., the finite value of about 12/10 obtained numerically by Langmuir and Blodgett¹¹) but in other practical problems, such as in-cloud droplet coagulation or in dry deposition it can be of great importance¹² and clearly it is of substantial value. If one or the other of these empirical expressions for s_* are used in the expression for the collision efficiency suggested in Part B that has the right large s behavior (at least for potential flow) then the contribution to the collision efficiency by inertial impaction can be approximated by

$$E_m = \left(\frac{s - s_*}{s + c} \right)^{3/2} \quad (34)$$

where $c = 2/3 - s_*$. This expression was used elsewhere in this volume¹¹ and adequately reflects both numerical and laboratory data.

SUMMARY COMMENTS FOR PART C

The problem of inertial collection at small Stokes numbers is doubly difficult because of the failure of regular perturbation techniques and because of the existence of critical Stokes numbers. What we have managed to do in the above is to learn a little more about the solution, using power series representations of solutions in the four limiting cases of the variable: $(z \rightarrow -\infty, s \rightarrow 0)$, $(z \rightarrow -1, s \rightarrow 0)$, $(z \rightarrow -1, s \rightarrow \infty)$ and $(z \rightarrow -\infty, s \rightarrow \infty)$, at least for the case of potential flow about a sphere. In addition, (13) yields a solution near the sphere for $0 \leq s \leq s_*$. However the most interesting

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FIGURE 3. Empirical Estimates of the Critical Stokes Numbers' Dependence on the Reynolds Number from the Numerical Studies of Beard and Grover¹⁰ and Taylor's Analytical result⁷ for potential flow about a sphere. As is discussed in the text it is not clear whether as $Re \rightarrow 0$, $s_* \rightarrow \infty$ or as $Re \rightarrow 0$, s_* is the value shown in the figure, obtained by Langmuir and Blodgett.¹¹

case, $z \rightarrow -1$, $s \rightarrow s_*$ from $s > s_*$ is elusive. For the case of Stokes flow about a sphere there is a strong suggestion that a critical Stokes number dependent only on the equation does not exist; whether or not collisions occur appears to depend only on the initial conditions. That there is a strong dependence on the initial conditions is to be expected for the nonlinear equation with which we are dealing. As will be discussed further in Part E, the Equation (1) is Abels' equation which is a generalization of the Riccati equation.

One of the sources of difficulty with the power series method used herein can perhaps be seen more clearly from a different physical problem. Mathematically the momentum equation also describes the particle's motion inside the obstacle; for example in the case of potential flow about a sphere, the sphere's boundary is simply a location where the fluid's

radial velocity is zero and there the upstream velocity is balanced by a doublet at the origin. Thus a similar physical problem is the vertical motion of, say, a tennis ball falling into an upward jet of fluid. As a result it is easy to see that u is as sketched in Figure 4. There it is seen that some solution curves (in the case of interest here, those with $s > s_*$) spiral into the point $u = 0$, $z = -1$. From this picture one clearly sees a source of difficulty with power series solutions: for $s > s_*$, u is multivalued at $z = -1$ and it is usually impossible to obtain multivaluedness from a power series. Further when $u = 0$, $du/dz \rightarrow \pm\infty$. It might be thought that it would be better to attempt to solve the equation for the spiral but

even in the (u,v) hodograph plane where the range of the variables is finite, the equation for the spiral is highly intractable. For potential flow about a sphere it is

$$\frac{1}{r} \frac{dr}{d\theta} = \left[\sin^2 \theta - \sin \theta \cos \theta + 3s \sin \theta \cos \theta (1 - r \cos \theta)^{4/3} \right] / \left[\sin \theta \cos \theta - \cos^2 \theta - 3s \sin^2 \theta (1 - r \cos \theta)^{4/3} \right] \quad (35)$$

where $u = r \sin \theta$, $v = r \cos \theta$. Fortunately, as will be shown in Part D, progress can be made in a different way.

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FIGURE 4. A Qualitative Indication of the Solution to the Momentum Equation for Motion Along the Upstream Axis of Symmetry for Potential Flow About a Sphere, Including an Indication of the Solution "Inside" the Sphere.

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ANALYTICAL INVESTIGATIONS OF INERTIAL DEPOSITION
OF SMALL AEROSOL PARTICLES FROM LAMINAR FLOWS
ONTO LARGE OBSTACLES
PART D - APPROXIMATE UPPER BOUNDS FOR THE COLLISION
EFFICIENCIES AT SMALL STOKES NUMBERS

W. G. N. Slinn

A general implicit solution, valid to first order in Stokes number, is found for the particle momentum equation and leads to graphical representations of the particle velocity fields. The Oseen uniformization technique is applied, and approximated to yield solutions for the collision efficiencies at small Stokes numbers for potential flow about a sphere and a cylinder, and for Stokes and Oseen flows about a sphere. An analytical continuation for an approximation to the collision efficiency for the case of potential flow about a sphere is presented and compared with numerical results.

INTRODUCTION

In this series of progress reports the following have been discussed: Part A showed that regular perturbation methods failed to yield a solution for the motion of a particle immersed in a laminar flow field about an obstacle; in Part B the method of matched asymptotic expansions gave a solution valid for large Stokes numbers; and Part C used power series methods in an attempt to learn more about the nature of the solution along the upstream axis of symmetry, especially for Stokes numbers near their critical value, s_* . In this report, an ad-hoc uniformization technique will be introduced which gives further information about the solution at small Stokes numbers, but this time in the whole field, not just along the upstream axis of symmetry. In subsequent reports other perturbation techniques (the method of multiple scales, method of composite expansions, and the method of matched asymptotic expansions for small s) will be applied to the problem.

AN IMPLICIT FIRST ORDER SOLUTION

An obvious solution to the particle momentum equation

$$\vec{s}\vec{u} \cdot \nabla \vec{u} = \vec{v} - \vec{u} \quad (1)$$

valid to first order in the Stokes number s can be seen by rewriting (1) in the form

$$\vec{u} + \vec{s}\vec{u} \cdot \nabla \vec{u} = \vec{v} \quad (2)$$

where, again, $\vec{u}$ is the particle continuum's velocity field and $\vec{v}$ is the fluid's. The LHS of (2) can be recognized as the first two terms of the Taylor expansion of $\vec{u}$ about the value of $\vec{u}$ at $\vec{r}$. Then the implicit but predictive solution to (1) to first order in s , is

$$\vec{u}(\vec{r} + \vec{s}\vec{u}) = \vec{v}(\vec{r}). \quad (3)$$

To see the application of (3) consider first the motion along the upstream axis of symmetry where (3) becomes $u(z + su) = v(z)$ or

$$u_{i+1} \equiv u(z_i + su_i) = v(z_i) \equiv v_i. \quad (4)$$

In Figure 1 equation (4) is used to evaluate $u(z)$ graphically for $s = 1/2$ and for the arbitrary velocity field shown. This result shows clearly the nature of the solution, much like the motion of a sloshing liquid in a pitching tank. The u -field is nonuniformly phase lagged

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FIGURE 1. An indication of the Nature of the Solution, (4), to the Particle Momentum Equation, (1), Along the Upstream Axis of Symmetry for an Arbitrary Fluid Velocity Field $v(z)$ and $S = 1/2$. The u -field is essentially a nonuniformly phase-lagged image of the v -field.

from the v -field. In Figure 2 equation (4) is used to evaluate $u(z)$ for the case of potential flow about a sphere and with $s = 1/2, 1/3$ and $1/5$. Actually the use of (4) with such large values of s [since terms $\theta(s^2)$ in the Taylor expansion have been ignored] is of questionable validity but they were chosen large so that the calculation would be shorter and there would be little question that "collisions" would occur. The u -curves were started at $z = -5$ using $u(z, s)$ given by the large z small s series found in Part C.

The question naturally arises now whether or not (4) can be used to ascertain analytically the critical Stokes numbers and the nature of the solutions near $s = s_*$. In the graphical evaluations of Figure 1 and 2 it is clear that $u(-1)$ will be positive and therefore particles will collide with the obstacle unless, near the stagnation point, the step size su is equal to or smaller than the distance from the stagnation point. Unfortunately, though, this observation is insufficient to enable the extraction of s_* because to obtain u one needs v at this distance, which requires that we know the distance, for which we need u , etc. However, we can at least determine bounds for s_* . To obtain these we note that for the boundary

conditions of interest, certainly $v \leq u \leq U_0$ for all s and $z < -1$. Therefore obvious (second estimate) lower and upper bounds on u , $\check{u} \leq u \leq \hat{u}$ can be defined via

$$\check{u}(z + sv) = v(z) \quad (5)$$

$$\hat{u}(z + sU_0) = v(z) \quad (6)$$

in which the step sizes have been forced to take the artificial sizes sv and sU_0 , where $U_0 = 1$ in the nondimensionalization used but it is convenient to retain the symbol. These bounds for u are shown in Figure 3 for the case $s = 1/2$, for potential flow about a cylinder. In this case the calculations were started from $z = -5$ with an expression similar to that developed in Part C for a sphere; for a cylinder the expression is

$$\begin{aligned} u \sim & 1 - \frac{1}{z^2} - \frac{2!s}{z^3} - \frac{3!s^2}{z^4} \\ & - (4!s^2 - 2 \cdot 1) \frac{s}{z^5} \\ & - (5!s^3 + 3!s - 5 \cdot 4) \frac{s}{z^6} - \dots \quad (7) \end{aligned}$$

The upper bound on u , $\hat{u}$, predicts that collisions occur for all $s > 0$. In contrast, $\check{u}$ predicts collisions

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FIGURE 2. The Solution, (4), for Potential Flow About a Sphere at $s = 1/5$, $1/3$ and $1/2$.

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FIGURE 3. Upper, $\hat{u}$, and Lower, $\check{u}$, Bounds for u , Obtained from (5) and (6) with $s = 1/2$. The u curve is obtained from (4).

only for large enough s . This can be seen analytically from (5) if we look for the critical s value which gives a last step length, $\check{s}_*v$, which is the same as the distance from the stagnation point, δ . For potential flow, i.e., when near the stagnation point $v = c\delta$, this gives $\check{s}_*c\delta = \delta$, which satisfies that above criterion if $\check{s}_* = 1/c$. Thus we obtain the bounds

$$0 \leq s_* \leq 1/c \quad (8)$$

where, again (see Part C) $c = 1, 2, 3, 4/\pi$ for potential flow about a plate, cylinder, sphere and disk, respectively. Thus in Figure 3 we see that $\check{u}(s = 1/2, z = -1)$ is zero, as is predicted by the bound (8). For the case of Stokes flow about a sphere, with $v = 3/2 \delta^2$ then the criterion gives $3/2 \check{s}_*\delta^2 = \delta$ which, for

$\delta \rightarrow 0$ predicts $\check{s}_* \rightarrow \infty$. Thus for Stokes flow it is again unclear whether or not a critical Stokes number exists: $0 \leq s_* \leq \infty$ (cf. Part C).

AN AD-HOC UNIFORMIZATION TECHNIQUE

It may appear that with the above analysis we have even retrogressed, from the analysis in Part C where we had $s_* = 1/4c$ for potential flow, to the case now where only bounds on s_* have been established. Also one should question the use of the solution $u(z + su) = v(z)$ for s values large enough to satisfy $s > s_*$. Further no new information has been gained about the analytic behavior of the solution near $s = s_*$. However, at least the stage has been set for appreciating the significance of ad-hoc uniformization techniques. The classic example of such uniformization is Oseen's method for approximately solving the problem of viscous flow about a sphere: whereas Stokes completely neglected the inertia terms $\check{v} \cdot \nabla \check{v}$ in the fluid equations, Oseen approximated them by $\check{U}_0 \cdot \nabla \check{v}$ and subsequently it has been shown¹ that this leads to a uniformly valid first order approximation to the solution for the flow about a sphere at low Reynolds numbers. In this section a similar ad-hoc uniformization technique will be explored, but just for motion along the upstream axis of symmetry.

Consider the case of the lower bound for u , $\check{u}$, defined via

$$sv \, d\check{u}/dz = v - \check{u} \quad (9)$$

Although (9) is a linear ordinary differential equation, extracting its solution can become complicated. For example, for the case of potential flow about a cylinder one obtains

$$\check{u}(s, z) = \frac{1}{s} \left(\frac{-z - 1}{-z + 1} \right)^{1/2s} e^{-z/s} \int_{|z|}^{\infty} e^{-\zeta/s} \left(\frac{\zeta + 1}{\zeta - 1} \right)^{1/2s} d\zeta. \quad (10)$$

This result has not been explored fully. Apparently it describes the full spiraling motion of the particle (see Part C), which explains why the result is zero at $z = -1$. It can be recast in terms of Whittaker functions² but the re-

sult is complicated and at this time does not appear worth exploring in depth.

Of more value is the solution obtained by overestimating the streaming term; i.e., the solution for the upper bound defined via

$$sU_0 \, d\hat{u}/dz = \hat{u} - v \quad (11)$$

where again $U_0 = 1$. The solution to (11) for potential flow about a sphere is

$$\hat{u}(z, s) = 1 - \frac{1}{s^3} \frac{\exp(-z/s)}{(z/s)^2} E_3(-z/s) \quad (12)$$

where E_3 is the familiar exponential integral

$$E_n(\zeta) = \int_1^{\infty} \exp(-\zeta t) dt/t^n \quad (13)$$

Notice that this Oseen uniformization gives us exactly the same answer as the method of matched asymptotic expansions (cf. Parts B and C), at least to first order in $1/s$. For small s the asymptotic expansion of (12) yields (as in Part C)

$$\hat{u}(z, s) \sim v(z) + \frac{3s}{z^4} \left\{ 1 + \frac{4!}{3!} \frac{s}{z} + \frac{5!}{3!} \frac{s^2}{z^2} + \dots \right\}. \quad (14)$$

For potential flow about a cylinder one obtains

$$\hat{u}(z, s) = 1 + \frac{1}{s^2} \frac{\exp(-z/s)}{(z/s)} E_2(-z/s) \quad (15)$$

which can be expanded similarly.

The above results are quite encouraging. It is tempting to accept them as the analytical continuations we have been seeking from the large s results, valid for all s , at least to first order in $1/s$ at large s and to first order in s at small s . However there remain a number of questions. First an exploration must be made to see if, as in the viscous flow problem, this Oseen uniformization is a uniformly valid first order approximation at small s . This will be explored in later reports using the

method of matched asymptotic expansions and other techniques. Also, the above results are only an upper bound for u obtained by replacing the streaming velocity with U_0 . It seems clear that they will fail for $s < s_*$ because as was seen in Part C, u is zero at $z = -1$ for $s < s_*$, and therefore we can not replace the streaming term by a nonzero constant. Thus at most it seems the above will give us a first order solution only for $s > s_*$ and it suggests (and we will assume in the sequel) that a more accurate upper bound can be found by replacing the convective acceleration term $\vec{u} \cdot \nabla \vec{u}$ by $\epsilon \vec{U}_0 \cdot \nabla \vec{U}$. Here $\epsilon = s - s_*$ and the symbol for $\hat{u}$ has been changed to U to reflect this different assumption. This assumption will be explored further in later reports. A further restriction of the above results is that they are applicable only along the upstream axis of symmetry. This restriction will now be removed.

APPLICATIONS TO THE COMPLETE FIELD

In the above, attention was focused on the particle motion along the upstream axis of symmetry. The methods developed will now be applied to the complete particle field. First it is of interest to look at the first-order graphical solution for the upper bound:

$$\hat{u}(\vec{r} + s\vec{U}_0) = \vec{v}(\vec{r}). \quad (16)$$

Some examples are shown in Figures 4 and 5 using fluid fields calculated by others.³⁻⁵ To obtain these results all one need do is plot the fluid streamlines about the obstacle and then displace the obstacle a distance s , upwind.

These results have a number of interesting features. For example, it is interesting to see a particle-free shadow develop behind the obstacle (which was alluded to in Part A). This particle-free region looks similar to the wake behind an obstacle immersed in a moving real fluid and leads to some speculations about the possible value of a two-fluid model for real fluids (see closing remarks). By identifying the streamline which bounds the particle-free shadow, one essentially has the value for the collision efficiency for this approximation. For example, in the case of potential flow shown in Figure 4,

since $2\pi\Delta\psi$ is the amount of fluid contained between two streamlines, then if the particle field is incompressible we have $E = 2\pi(4 \times .06 a^2 U_0 / \pi a^2 U_0) = 0.48$ for $s = 1/2$. This will be compared with analytical results later in this report. The cases for finite Re (Figure 5) will be returned to in Part F. It is also of interest to see the point where this particle-free region separates from the obstacle. This will be analyzed below, but for now it is interesting to observe that it seems to occur roughly where real boundary layers separate. Finally it is of interest to compare qualitatively the variations in the particle field caused by variations in s versus Re . Qualitatively it appears that the particle field depends very much more strongly on s than Re and if one sought parameters of equal sensitivity then it would not be unreasonable to try s and $\ln Re$.

If analytical solutions for the approximate upper bound are sought, new difficulties appear. The equation is

$$\epsilon \hat{k} \cdot \nabla \vec{U} = \vec{v} - \vec{U} \quad (17)$$

where, again, $\epsilon = s - s_*$ and $\vec{U}$ is the corresponding approximation to $\hat{u}$. The formal solution to (17) is

$$\vec{U} = \frac{e^{-z/\epsilon}}{\epsilon} \int_{-z}^{\infty} d\zeta \vec{v}(\zeta, z = -\zeta) e^{-\zeta/\epsilon} \quad (18)$$

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FIGURE 4. Graphical Solution for the $\vec{u}$ Field, from (16) with $s = 1/2$, for Potential Flow About a Sphere. The fluid field was taken from Batchelor.³

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FIGURE 5. Graphical Solution as in Figure 4, Using the Flow Fields from References 4 and 5.

where the off-axis distance ϖ is a fixed parameter in the integrand. For potential fluid fields (18) can be written as

$$\vec{U} = \frac{\hat{k}}{\epsilon} \phi(\varpi, z) + \frac{e^{-z/\epsilon}}{\epsilon} \left[\hat{e}_{\varpi} \frac{\partial}{\partial \varpi} - \frac{\hat{k}}{\epsilon} \right] \int_{-z}^{\infty} \phi e^{-\zeta/\epsilon} d\zeta \quad (19)$$

where ϕ is the velocity potential and $\hat{e}_{\varpi}$ is a unit vector perpendicular to the axis of symmetry. The difficulty is that these integrals are not easy to evaluate. On the upstream axis of symmetry they reduce to the exponential integral function. If the integrals are evaluated by parts then what is obtained are asymptotic expansions of $\vec{U}$, for large or small values of ϵ . This can be done, but instead, solutions of this form are now sought from the original equation (17).

In particular for large ϵ or small $\Sigma = 1/\epsilon$, a solution to (17) is sought of the form

$$\vec{U}(\epsilon, \vec{r}) \sim \vec{U}_0(\vec{r}) + \Sigma \vec{U}_1(\vec{r}) + \dots, \quad \Sigma \rightarrow 0 \quad (20)$$

The result (for this upper bound) is exactly the same, to first order in $1/s$, as was obtained via the method of matched asymptotic expansions in Part B. Thus we obtain (as in Part B, but here the coordinate system is reversed) for potential flow about a sphere

$$\vec{U} = \hat{k} + \Sigma \frac{\hat{e}_r}{2r^2} + O(\Sigma^2) \quad (21)$$

which predicts that collisions will occur until $\theta' = \cos^{-1}(\Sigma/2)$ off the upstream axis of symmetry; i.e., until almost $\pi/2$ on the windward side of the sphere. For larger angles the particle velocity field is positive at the sphere's surface.

Now, for some new results, we try

$$\vec{U} \sim \vec{U}_0 + \epsilon \vec{U}_1 + \epsilon^2 \vec{U}_2 + \dots, \epsilon \rightarrow 0 \quad (22)$$

and obtain the general solution

$$\vec{U} = \vec{V} - \epsilon \frac{\partial}{\partial z} \vec{V} + \epsilon^2 \frac{\partial^2}{\partial z^2} \vec{V} - + \dots \quad (23)$$

For potential flow about a sphere (23) yields

$$\begin{aligned} \vec{U} = \vec{V} - \frac{3\epsilon}{r^4} \left[\left(\cos^2 \theta - \frac{1}{2} \sin^2 \theta \right) \hat{e}_r \right. \\ \left. + \sin \theta \cos \theta \hat{e}_\theta \right] + O(\epsilon^2) \end{aligned} \quad (24)$$

which degenerates to the solution found earlier for motion along the upstream axis, see (14), if in (24) $\theta = \pi$ and $\epsilon = s$. To first order in ϵ , (24) predicts collisions out to $\theta' = \pi/4$. For Stokes flow about a sphere, (23) gives

$$\begin{aligned} \vec{U} = \vec{V} - \frac{3}{2} \frac{\epsilon}{r^2} \left[\left(1 - \frac{1}{r^2} \right) \left(\cos^2 \theta \right. \right. \\ \left. \left. - \frac{1}{2} \sin^2 \theta \right) \hat{e}_r - \frac{\sin \theta \cos \theta}{r^2} \hat{e}_\theta \right] \\ + O(\epsilon^2) \end{aligned} \quad (25)$$

which gives no collision to $O(\epsilon)$ since at $r = 1$, $U_{1r} = 0$. This is another hint that a critical Stokes number does not exist for Stokes flow, but more work is required and this will be explored more fully later. For Oseen flow about a sphere (23) yields to $O(\epsilon)$

$$\begin{aligned} U_r = v_r - \epsilon \left[\frac{3}{2r^4} \left(\frac{1}{2} \sin^2 \theta - \cos^2 \theta \right) \right. \\ \left. - \frac{3 \cos \theta}{r^3 \text{Re}} + \left\{ \frac{3 \cos \theta}{2r^2} \left(1 + \frac{2}{r \text{Re}} \right) \right. \right. \\ \left. \left. - \frac{3}{4r^2} \sin^2 \theta \left(1 + \frac{r \text{Re}}{2} \right) \right\} \exp \left\{ -\frac{r \text{Re}}{2} \right. \right. \\ \left. \left. \left(1 - \cos \theta \right) \right\} \right] \end{aligned} \quad (26)$$

For $r = 1$ and small Reynolds number this gives

$$U_r(1, \theta) = \frac{3}{4} \epsilon \text{Re} \cos \theta \sin^2 \theta + O(\epsilon) \quad (27)$$

which predicts that collisions occur over the entire upstream side of the obstacle (except, surprisingly, at the stagnation point) at least for $s > s_*$ which, however, is unknown. This will be explored further in Part F.

THE COLLISION EFFICIENCY

To obtain an estimate for the collision efficiency based on these approximate solutions for an upper bound on $\vec{U}$, the particle density must first be found. This is governed (see Part A) by

$$\vec{U} \cdot \nabla N = -N \vec{V} \cdot \vec{U} \quad (28)$$

where the choice has been made, which seems consistent, to solve just for an upper bound to n , labeled N , found by using $\vec{U}$ instead of $\vec{u}$ in the continuity equation. But from (17) we have

$$\epsilon \frac{\partial}{\partial z} D = -D \quad (29)$$

where $D = \nabla \cdot \vec{U}$ and therefore

$$D = D_0 \exp(-z/\epsilon). \quad (30)$$

Consequently if the dilation, D , is zero at infinity, then it is zero everywhere within the field. Consequently in this Oseen uniformization technique, the particle field is incompressible or, from (28), N is a constant along particle lines: N_0 for lines emanating from infinity; zero for lines emanating from the obstacle (which defines the particle-free shadow).

Now, to obtain the collision efficiency for small ϵ , the flux of particles (dictated by U_r at $r = 1$) is integrated over the sphere where U_r is negative. Then the result is normalized by the flux of particles to the obstacle if the particle lines were undeflected (see Part B). The results are: for potential flow about a cylinder, $\hat{E} = \epsilon$; for potential flow about a sphere, $\hat{E} = (4/3)^{1/2} \epsilon$; and for Oseen flow about

a sphere, $\hat{E} = (3/8) \epsilon \text{Re}$, where $\epsilon = s - s_*$ and $\text{Re} = RU_0/\nu$. It is encouraging that this result for potential flow about a sphere gives $\hat{E} = 0.481$ which is the same value as was obtained from the graphical solution, Figure 4. Further, for potential flow about a sphere, if we replace the factor $(4/3)^{1/2} = 1.15$ by unity (the solution is an approximate upper bound for E) then coupling this result with the result for large s found in Part B, we can easily find an analytical continuation for the approximate upper bound for the collision efficiency:

$$\hat{E} = \epsilon / (1 + \epsilon) \quad (31)$$

where $\epsilon = s - s_*$, with $s_* = 1/12$. Of course there are many other possibilities besides (31); others will be given in Part F. In Figure 6 the suggestion (31) is compared with numerically calculated collision efficiencies.⁶⁻⁸ Clearly, as an upper bound, (31) appears quite attractive.

SUMMARY COMMENTS FOR PART D

The analysis of Part D started from recognizing the obvious solution to the momentum equation, valid to first order in s , $\vec{u}(\vec{r} + s\vec{u}) = \vec{v}(\vec{r})$. Applying this to motion along the upstream axis of symmetry gave graphical results which clearly indicated the

nature of the solution. Also it led to obvious lower and upper bounds for u , identified as $\hat{u}$ and $\hat{u}$, respectively. Although these led only to rather loose bounds for the critical Stokes number, s_* , they did again suggest that $s_* \rightarrow \infty$ for Stokes flow and, more importantly, they led to an application of Oseen's uniformization technique. Using this, the result equivalent to $\mathcal{O}(1/s)$ to that obtained by the method of matched asymptotic expansions (Parts B and C) was found for $\hat{u}$. This suggested that an approximation for $\hat{u}$ could be found, for the whole flow field, if the convective acceleration term, $s\vec{u} \cdot \nabla \vec{u}$ were replaced by $\epsilon \vec{U}_0 \cdot \nabla \vec{u}$ where $\epsilon = s - s_* > 0$. $\vec{U}_0$ is the uniform upstream velocity, and $\vec{u}$ symbolized this approximation for $\hat{u}$. This led to approximations for the collision efficiencies for potential and Oseen flow; the collision efficiency for Stokes flow was found to be zero to first order in ϵ . (One can obtain $\hat{E} = (3/4)\epsilon^2$ for Stokes flow but it is probably not valid to use the theory to generate a solution to order ϵ^2 .) It is encouraging that the results for potential flow are close to other's numerical results, and to the graphical result found from $\hat{u}(\vec{r} + s\vec{U}_0)' = \vec{v}(\vec{r})$.

Perhaps some comments are appropriate to try to justify seeking the collision efficiency by the Oseen

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FIGURE 6. A Comparison of the Approximate Upper Bound for the Inertial Collision Efficiency, (31), with Numerical Solutions⁶⁻⁸ for Potential Flow About a Sphere.

uniformization technique, when clearly it gives an upper bound for the inertial contribution to the collision efficiency. A first comment must be that, given the difficulty of the problem, a theoretician is thankful for any method that gives reasonable answers. Further, though, it is a reasonable contention that obtaining an upper bound is a valuable result because (as was emphasized in Part C) other physical processes can contribute to the collision efficiency and if an upper bound for the inertial contribution is found which is smaller than the contribution from other effects, then the result allows us to disregard the inertial contribution. However, of more importance than the above, is the suggestion that in fact the Oseen uniformization technique gives the correct solution to order $(1/s)$ at large s and to order $(s - s_*)$ at s near s_* . Indeed it was seen that this was true for the large Stokes number limit; in Parts E and F our investigations into the small s limit will be reported.

It may even be worthwhile to attempt to defend this entire analysis, given the available numerical results. In this regard, comments on the advantage of analytical over numerical results will not be repeated even though, in the author's opinion, these arguments are sufficient to justify the approach taken. Instead three other topics will be mentioned. One of these relates to possible contributions to the theory of singular perturbation analysis. On the one hand the problem represents a marvelous opportunity to test the range of applicability of various new techniques,

such as the methods of multiple scales, strained coordinates and matched asymptotic expansions. This will be discussed further in Part E. On the other hand, there are hints that this problem may yield the first known practical example (cf. reference 1) where perturbation solutions can be found at both extremes of the perturbation parameter. A second reason why the author believes the analysis should continue is because basically the analysis is of Abels' equation which for the past 150 years has failed to submit to analysis except in a few restricted cases. To the author's knowledge singular perturbation techniques have not previously been applied to this equation and therefore new results obtained may contribute to the solution of other important practical problems described by Abels' equation. Finally and of most interest to the author is that behind this analysis may lie a two-fluid model for real fluids. Hints of this were seen in the graphical solutions where pseudo wakes and flow separation seemed to be occurring for the $\bar{u}$ -field. Here the $\bar{u}$ -field is the particle field, i.e., a field in which mass is important and which is coupled to the $\bar{v}$ -field. The rudiments of a two-fluid model for fluids emerges by considering first a fluid in which inertial terms are ignored (e.g., Stokes flow) and then coupling a $\bar{u}$ -field to this $\bar{v}$ -field, such as via $s\bar{u} \cdot \nabla \bar{u} = \bar{v} - \bar{u}$. In this $\bar{u}$ -field, inertial effects dominate. Hopefully later studies will explore this model in depth as well as probe deeper into the problem of the Reynolds number dependence of the collision efficiency.

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CREATION OF A DATA-PROCESSING CODE
FOR USE WITH THE BNW AIRCRAFT SAMPLING SYSTEM

J. M. Hales and J. W. Slood

This report describes briefly a new software package for processing aircraft data, which has been developed during the past year. The package is available in a highly user-oriented form, and is expected to function as an essential component of the aircraft sampling program in the Atmospheric Sciences Department.

The majority of data that is acquired aboard the BNW Cessna Aircraft is recorded in digital form on magnetic tape using a Metrodata DC 620 recorder. Data recorded in this fashion may be subsequently treated in a number of ways, depending upon the user's need and the type of data involved. Until the present time, however, we have not possessed a versatile, general-purpose method of routine data processing; this report describes a procedure developed during the past year to fulfill this need.

A typical operation using the aircraft tape acquisition system involves the following steps:

1. Recording of data on Metrodata tape,
2. Translation of information from Metrodata tape to industry-compatible tape, and
3. Processing of the data on the industry-compatible tape using a large batch computer.

The second step of the operation is carried out by physically mounting the Metrodata tape on a Data-General NOVA minicomputer, which performs the necessary translation operations and writes the corresponding output on industry-compatible tape.

The third step is presently accomplished on a CDC 6600 CYBER computer - a step that involves the software (in the form of a FORTRAN-IV program) developed under this project and described in the following paragraphs.

Basically, the software is written in four segments, which include:

1. Reading of the tape by the CYBER and translation and storage in CYBER-compatible form,
2. Preliminary processing of raw data: culling of bad data and (if desired) specific time averaging,
3. Final processing and printing of "standard" data, and
4. Final processing and printing of special data.

Here, "standard" data are defined as data that normally are recorded for all experiments; these include the following:

- time
- indicated temperature
- indicated air speed
- pressure altitude
- carbon-strip humidity sensor
- 2 VOR (Variable Omni Range) stations
- DME (Distance Measuring Equipment), and
- Dew-point hygrometer

These inputs are processed by applying precision calibrations and appropriate corrections within the third block of the software package. Position of the aircraft is calculated two ways at the option of the user (triangulating from the VOR's or using a VOR-DME combination), and is listed as x and y coordinates using one of the reporting VOR stations as a reference. Output from this segment consists of the following, which is expressed in tabular output and/or in plotted form:

- time
- true temperature
- potential temperature
- true airspeed
- true altitude
- relative humidity (RH)
- dew point, and
- aircraft location

The fourth segment of the software package is simply an "open end", which allows the user to incorporate any special data handling for additional raw data inputs that he may desire. This allows the system to be extremely versatile while allowing the individual researcher to gain access to the system in a minimal amount of time. The code is written in a highly user-oriented fashion to facilitate this aspect.

It is expected that this system will be used extensively from the present time on for processing of data from the Cessna 411. It will also be employed as a segment of processing operations for future hardware acquisition systems if and when such systems become available.

PROPOSED TERMINOLOGY FOR PRECIPITATION SCAVENGING

W. G. N. Slinn

A list of suggested definitions for concepts in precipitation scavenging is presented. The major feature of the proposed terminology is to abandon past usage of the words "washout" and "rainout."

INTRODUCTION

A number of researchers working in the area of precipitation scavenging have expressed dissatisfaction with the present terminology and jargon of the field. As a result the author, working with A. N. Dingle, R. J. Engelmann, D. F. Gatz, J. M. Hales and R. G. Semonin (personal communications) has formulated the set of definitions given below. Figure 1 illustrates the main concepts in the distinctions among the terms.

DEFINITIONS

I. General Definitions

1. Scavenging - removal; replaced in compound words by the word "out."
2. Precipitation Scavenging - the removal of any material from the atmosphere to the earth's surface by various types of precipitation.

Notes (i) If an abbreviation for "precipitation scavenging" is desired, it is recommended that the word "washout" be used and that washout's other definition be abandoned. The result will be consistent with most European usage of "washout."

- (ii) Precipitation scavenging generally refers to wet deposition as contrasted with dry deposition. However, there are a number of marginal cases (for example, dry deposition of fog

droplets) which are probably best described without further abbreviation.

- (iii) Fallout is a common term used either to describe the removal of nuclear bomb debris from the atmosphere or to identify the material itself. In some cases, fallout can be further distinguished as wet-fallout (fallout removed by precipitation scavenging) and dry-fallout (fallout removed by dry deposition).

II. Altitude Dependence of Precipitation Scavenging

Previously many authors have used the well-known terminology to distinguish below-cloud from in-cloud scavenging. It is suggested that use be made of the full expressions:

1. Below-cloud (or sub-cloud) scavenging--precipitation scavenging (washout) by hydrometeors whose growth has substantially terminated or, more simply, washout beneath the visible cloud which is a source of the precipitation.
2. In-cloud scavenging--precipitation scavenging (washout) which initiates during the formation and growth of cloud particles or, more simply, removal from within the visible cloud, which culminates in the deposition of material on the earth's surface.

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FIGURE 1. Dependence of Precipitation Scavenging (Washout) on: (1) Position, (2) Precipitation, (3) Pollutants, (4) Processes

In some cases it may also be convenient to identify:

3. Plume washout--precipitation scavenging of material from plumes.

Notes (i) Although the "long-form" (i.e., in-cloud scavenging) is not so convenient as the familiar "short-form," nevertheless, it is recommended because the word "rainout" is needed (see below) to more logically relate to rain scavenging.

- (ii) A special complication with the terminology for in-cloud scavenging arises because the attachment of a pollutant to cloud water does not necessarily result in the deposition of the pollutant on the earth's surface. It is suggested that the various processes by which the pollutant attaches to cloud water (or water attaches to the pollutant) be described with familiar terms such as diffusional attachment, phoretic capture, condensational growth, etc.,

and that only the total process which culminates in the pollutant reaching the ground, be described as in-cloud scavenging.

III. Precipitation Dependence of Washout

Especially for the case of below-cloud scavenging when the hydrometeor type can be easily identified, the dependence of precipitation scavenging on the hydrometeor type can be identified as in:

1. Rain Scavenging--obvious definition; expected to evolve to the word "rainout" when the previous use of this word had been abandoned;
2. Snow Scavenging (snowout); etc.

IV. Pollutants and Mechanisms

1. When it is desirable to identify both the scavenged material and the hydrometeor type then usage can follow the examples: particle snowout; gas rainout; etc.
2. If it is also desirable to identify the physical mechanism then usage can follow the examples: inertial, particle snowout; reversible, gas rainout; etc.

V. Precipitation Scavenging Rates (or Washout Rates)--a nonexplicit term used to describe all removal rates in precipitation scavenging processes.

Note: Care should be taken to explicitly define the type of washout rate being considered: number, activity, mass, etc., washout rate. Also, further distinction should be made as to whether the rates are differential or integral (for monodisperse versus polydisperse aerosols, respectively) and whether the washout rates are considered as space-point or space-averaged removal rates (the former, which may be called "washout rate coefficients," are used almost exclusively in the convective diffusion equation; the latter, which might be called just "washout rates," are observed "directly" in experimental work).

VI. Below-Cloud Scavenging Rates--obvious definition.

- (a) Rain scavenging (rainout) rate, snowout rate, etc.--below-cloud scavenging rates for specific hydrometeors.
- (b) Particle rainout rate, etc.--used when it is desired to identify both the scavenger and the scavenged material.

Note: The symbol Λ is frequently used for the below-cloud rain-scavenging rate (viz., rainout rate). From the above, the rainout rate coefficient for an aerosol particle of radius a at position $\vec{r}$ at time t would be $\Lambda(a, \vec{r}, t)$, and for the steady state, space-averaged rainout rate $\Lambda(a)$. For a polydisperse aerosol with size distribution described by the probability density function $f(a)$ then the n th-moment, space-point rainout rate coefficient would be

$$\Lambda_n(\vec{r}, t) = \int_0^\infty a^n \Lambda(a, \vec{r}, t) f(a) da.$$

VII. In-Cloud Scavenging Rates

Since in-cloud scavenging can be interpreted to be a multi-rate process, various intermediate rate coefficients can be identified. However, experimentalists may prefer to continue to use ψ as the observed, overall in-cloud scavenging rate. There does not yet appear to be a consensus on recommended symbols for individual rate processes. One possibility is to use various lower case Greek letters: α --attachment rate, β --cloud water removal rate, etc., and the frequent use of K for the volumetric coagulation rate should be noted.

VIII. Precipitation Scavenging (Washout) Concentration Ratios

The ratio of the concentration of the pollutant in precipitation sampled at some height z_1 , $\kappa(z_1)$ to its concentration in air at height z_2 , $\chi(z_2)$.

- Notes (i) If the dominant removal process can be identified, these washout ratios can be further distinguished as below-cloud versus in-cloud washout ratios.
- (ii) If suitable units are chosen so that the ratio is dimensionless and therefore the (physically real) concentration factor of about 10^6 is displayed, then the κ/χ ratios might be better termed "washout concentration ratios," or just "concentration ratios--" the word "concentration" being used for both its connotations.
- (iii) If convenient units are chosen so that the κ/χ ratios are of order unity (e.g., κ in $\mu\text{g cm}^{-3}$ or mg l^{-1} and χ in $\mu\text{g m}^{-3}$) but are dimensional (e.g., $\text{m}^3 \text{cm}^{-3}$) then they might be called just "washout ratios."
- (iv) If mixing ratios are used, e.g., $\kappa_p(z_1)$ the mass of pollutant per mass of water sampled

at height z_1 , and $\mu_p(z_2)$ the mass of pollutant per mass of air sampled at height z_2 , then the ratios might be called "washout mixing-ratio factors," or just "wash-out factors."

Particle volume distribution-- $v(a)$, $f_3(a)$ ($\mu\text{m}^3 \text{ cm}^{-3} \mu\text{m}^{-1}$)

Particle diffusivity-- D , Δ , Y ($\text{cm}^2 \text{ sec}^{-1}$)

Particle relaxation time-- τ (sec^{-1})

IX. Symbols

In lieu of presenting a list of recommended symbols, a short list of current usage and convenient units follows. Nevertheless, there is a tendency to recommend the first symbols that appear.

Precipitation rate-- p , J_0 , I , R (mm hr^{-1})

Total precipitation-- P , h (cm)

Cloud liquid water content-- L , λ , w (gm m^{-3})

Cloud water content-- W

Raindrop size: diameter-- D ;
radius-- R (mm)

Raindrop number distribution-- $N(D)$, $F(D)$ ($\text{m}^{-3} \text{ mm}^{-1}$)

Raindrop volume distribution-- $V(D)$, $F_3(D)$ ($\text{mm}^3 \text{ cm}^{-3} \text{ mm}^{-1}$)

Particle size: diameter-- d ;
radius-- a , p (μm)

Particle number distribution-- $n(a)$, $f(a)$ ($\text{cm}^{-3} \mu\text{m}^{-1}$)

Particle log-number distribution-- $\Delta n / \Delta \ln a$, $n(\ln a)$ (cm^{-3})

Drop terminal velocity-- v_t , v_s , w_d (cm sec^{-1})

Reynolds number-- Re

Nusselt number-- Nu

Sherwood number-- Sh

Schmidt number-- Sc

Lewis number-- Le

Peclet number-- Pe

Stokes number-- Stk , S , s , P

Interception parameter-- κ , a/R

Collision efficiency-- E , $\mathcal{E}$, $\mathcal{E}$

Rain scavenging (rainout) rate-- Λ (sec^{-1} , hr^{-1})

Snow scavenging (snowout) rate-- Σ , Λ (sec^{-1} , hr^{-1})

In-cloud scavenging rate-- Ψ (sec^{-1} , hr^{-1})

DRY DEPOSITION OF PARTICLES

Gravity settling and turbulent eddy diffusion processes are an important mechanism by which an atmosphere is depleted of particulate material. Past research at Battelle-Northwest has studied deposition with monodisperse particles, first in tubes transporting particles, then in wind tunnel studies. Earlier field studies determined by mass balance the depletion of particles from a plume.

Our recent work in the wind tunnel has related deposition velocity to particle size, average velocity, friction velocity, and the unique characteristics of surfaces. Models have been developed which take these variables and a large body of available data into account. Ongoing field studies have compared depositing with nondepositing tracers as aids in evaluating deposition. This technique, as well as direct turbulent flux measurements of pollutants, should expand the capability for future deposition studies over larger regions and onto varied surfaces.

A spectrum of investigations in 1974 advanced empirical and theoretical knowledge of dry deposition processes. Wind tunnel measurements extended the deposition data for submicron particles on various surfaces and at various wind speeds, and further validated the "three box" model. "Experiments of opportunity" permitted measurements of radon daughter deposition to a lake surface and measurement methods development for dry deposition to snow. Accuracy of dry deposition by mass balance computations was shown to be greatly limited even when using a dense three-dimensional sampling grid. An interesting new modeling concept was developed which allows reasonable predictions of deposition velocities from basic premises about the position of the particle in the atmosphere and the rate limiting transfer processes. Methods were developed to permit deposition measurements in field investigations planned for 1975.

- **RADIOISOTOPES AS PARTICLES AND VOLATILES**
- **ATMOSPHERIC DIFFUSION, DEPOSITION AND TRANSPORT PHENOMENA**
- **ATMOSPHERIC AEROSOLS AND TRACE GASES**
- **RADIOACTIVE FALLOUT RATES AND MECHANISMS**
- **BATTELLE MEMORIAL INSTITUTE PHYSICAL SCIENCES PROGRAM**

PARTICLE DRY DEPOSITION VELOCITIES

G. A. Sehmel and W. H. Hodgson

Particle deposition velocities are predicted from a steady-state one-dimensional mass transfer model. Dimensionless integral mass transfer resistances are used for the predictions. Predictions are made for particle densities of 1, 4, and 11.5 g/cm³ and a reference concentration height of 1 m. Ranges of variables considered are for individual particle diameters from 0.01 to 100 μ m, roughness heights from 10⁻³ to 10 cm, friction velocities from 10 to 200 cm/sec, and Obukhov's stability length criteria of ± 10 m.

INTRODUCTION

Particle removal rates from polluted atmospheres to environmental surfaces have been described by dry deposition velocities, which are a function of particle, surface, and air properties. Deposition velocities have been determined for field conditions, but data have never been organized into a unified approach to extend predictions to other test conditions. These removal rates are important in considerations of air pollution removal, to prevent exceeding the environmentally permissible upper limits of air pollutant concentrations. Deposition velocities have been summarized for several authors.¹ Unfortunately for predictive purposes, experimental deposition velocities have often been determined with limited control of experimental conditions. Particle diameters have not been adequately known, and hence experimental results have usually been reported with insufficient information to generalize deposition velocities to other field conditions.

A second limitation to generalization from field data is that particle

and gaseous removal rates are non-steady-state processes dependent upon both the mass transfer resistance of the upper atmosphere as well as the mass transfer resistance near the deposition surface. Historically, deposition velocities are a steady-state mass transfer approximation of the nonsteady-state mass transfer deposition processes. In field experiments, deposition velocities, v_d , have been defined as a deposition flux, N , divided by an airborne concentration, x . That is,

$$v_d = \frac{N}{x} \quad (1)$$

which has units of length/time usually reported as cm/sec. Airborne concentrations are usually measured at 1 m above the deposition surface.

This deposition velocity concept fails if particle and gaseous mass transfer is controlled by pseudo eddy diffusion in the atmosphere-surface exchange. It is the exception rather than rule that nonsteady-state mass transfer can be adequately described by a single point concentration measurement. If mass transfer occurs, airborne concentration profiles must, of necessity, be developed to act as

the driving gradient for mass transfer in a diffusion controlled process. Only in the nominal case of simple gravity settling will concentration profiles be constant.

In mass transfer processes the airborne pollutant concentration and concentration gradient at any point result from nonsteady-state processes which reflect the capability of the atmosphere to deliver pollutant from regions of higher concentrations to regions of lower concentrations. In relative terms as a function of height and constant flux, a large concentration gradient in a region reflects a large relative mass transfer resistance in that region. In contrast, a small concentration gradient in a region reflects a small relative mass transfer resistance. Furthermore, because of these concentration gradients, most field determined deposition velocities define only the surface mass transfer removal rates for the particular depositing plume test conditions above the reference concentration height. To extrapolate these field deposition velocities to new situations requires similitude between pollutant plume shape, particle diameters, wind shear, etc.

This paper presents an initial generalized technique for predicting particle removal rates by dry deposition from any airborne plume. Inherent in applying the method is the assumption that one can completely describe the pollutant plume concentration profiles as a function of time and space. Otherwise, the usual assumption of a deposition velocity applicable to a 1 m reference height will be required. Predicted deposition velocities are given for a 1 m reference height.

GENERAL MODEL

Pollutant motion in the atmosphere is described by the nonsteady-state continuity equation which, conceptually, can be separated into a multi-box deposition model as a function of height above the deposition surface. Mass transfer resistances are to be calculated to describe the particle flux through the boxes. Mathematical requirements are that concentrations are equal at the juncture of adjoining boxes. Ideally there should be at least three boxes describing the overall deposition process.

In box 1, airborne pollutant plume movement is described by standard meteorological equations. In this atmospheric turbulent layer, mass transfer processes are described by micrometeorological eddy diffusivities. In the overall deposition process, the plume approaches and interfaces with box 2. This idealized layer of box 2 is just above the vegetative canopy or surface elements in a region where transfer processes are modified by the presence of the canopy or surfaces. This interface region can extend to all scales from small vegetation and trees, to scales as large as cities.

Deposition velocities will be predicted through calculating integral diffusional resistances in each of the three boxes. Unfortunately, predictions must be limited to surface elements with small roughness heights since diffusional resistance correlation exists only for small surface roughness. Perhaps as additional data are obtained, further model generalization to both larger roughnesses and sub-boxes within box 3 will be predictable.

Model predictions of deposition velocities using boxes 2 and 1 will also be for the simplest case--steady state one dimensional flow from a uniform pollutant source. Extension of model concepts to other pollutant plumes is limited only by the reader's requirements and ability to describe pollutant concentration profiles caused by diffusion and transport at significant elevations above environmental surfaces. If the user has a plume surface depletion model available to predict concentrations at the interface of boxes 2 and 3, the user will not need the proposed simple model. Rather, the user should directly use the mass transfer resistance integral for box 3 in that more complex model.

Most diffusion transport equations still use a deposition velocity rather than a mass transfer integral for a boundary condition. Consequently, deposition velocities K_1 -m will be presented. The deposition velocity, K , is defined as

$$K_1 = \frac{N}{C} \quad (2)$$

In this case, the concentration, C , is for monodispersed particles and is

measured 1 cm above the deposition surface. The deposition velocity nomenclature K , rather than v_d , reflects that deposition velocities are a function of particle diameter. The subscript 1-m indicates a reference pollutant concentration at a 1 m reference height.

SIMPLE MODEL

Deposition velocities K assume particle deposition is described by a one-dimensional, steady-state continuity equation. Basic assumptions in this model are that particles diffuse at a constant flux from a uniform concentration of particles, that a relationship for particle eddy diffusivity can be determined, that the effect of gravity can be described by the terminal settling velocity, that particle agglomeration does not occur, and that particles are completely retained by the surface. Based upon these assumptions, the deposition flux to a surface is described by

$$N = -(\epsilon + D) \frac{dC}{dz} - v_t C, \quad (3)$$

in which v_t is the absolute value of the terminal settling velocity.

The deposition velocity is predicted from a dimensionless integral form of Eq. (3).

$$- \int_{C_z}^0 \frac{u_*}{N + v_t C} dC = \int_{z^+}^{r^+} \frac{dz^+}{\epsilon/\nu + D/\nu} = \text{Int} \quad (4)$$

in which ϵ is the particle eddy diffusivity, D is the Brownian diffusivity, ν is the kinematic viscosity of air, u_* is the friction velocity, $z^+ = zu_*/\nu$ is the dimensionless distance above the surface, and v_t is the terminal settling velocity. Integration limits are that particle concentration is C_z at a reference height of z cm and that particle concentration is zero at a dimensionless particle radius, r^+ , from the deposition surface. The right-hand integral is a resistance integral (negative in value) abbreviated as Int. Int contains the dimensionless eddy diffusivity, ϵ/ν , and Brownian diffusivity and hence Int quantifies only diffusional resistance between

the integration limits. This integral Int can be subdivided into

$$\text{Int} = \int_{z^+}^{z^+_{1-2}} \frac{dz^+}{\epsilon/\nu + D/\nu} + \int_{z^+_{1-2}}^{z^+_{2-3}} \frac{dz^+}{\epsilon/\nu + D/\nu} + \text{Int}_3 \quad (5)$$

where the first integral is Int_1 and the second integral is Int_2 . The limit z^+_{1-2} is the distance at which boxes 1 and 2 interface. Similarly, boxes 2 and 3 interface at z^+_{2-3} . These integrals will be evaluated after a relationship between deposition velocity and Int is shown.

The deposition velocity is obtained from an integrated form of Eq. (4) for the deposition flux in which

$$N = \frac{v_t C_z \alpha}{1 - \alpha} \quad (6)$$

in which

$$\alpha = \exp(-v_t \text{Int}/u_*). \quad (7)$$

Now the deposition velocity is defined in terms of the reference concentration, C_z at z cm height,

$$K_z = \frac{(-N)}{C_z}. \quad (8)$$

Thus, the deposition velocity at height z is

$$K_z = \frac{v_t}{1 - (1/\alpha)}. \quad (9)$$

As shown by Eq. (9), the lower limit of predicted deposition velocities is v_t . The reason for this limit is that if the diffusional resistance were large (Int is a negative number), α would approach infinity and $1/\alpha$ would approach zero. However, as diffusional resistance became relatively less, the deposition velocity would become increasingly greater than the gravitational settling velocity.

Int₃

Surface integral resistances Int₃ were evaluated by Sehmel and Hodgson² from wind tunnel determined deposition velocities. Deposition velocities, K₁, were determined for deposition of monodispersed particles onto four different surfaces. These were an artificial grass 0.7 cm in height, a smooth brass surface, crushed gravel 0.5 to 1.6 cm in diameter, and a water surface with wave heights up to 2.5 cm. Particle diameters were from 0.03 μm up to 30 μm for a particle density of 1.5 g/cm³. Friction velocities, u_{*}, ranged from 11 to 144 cm/sec and roughness heights, z₀, from 0.004 to 0.4 cm. In all cases, the reference concentration height for defining K₁ was 1 cm above the deposition surface which corresponds to a box 3. Consequently, values of Int₃ were evaluated from the K₁'s by

$$\text{Int}_3 = - \frac{u_*}{K_1} \ln \left(\frac{1}{1 - v_t/K_1} \right). \quad (10)$$

Least squares techniques were used to determine a dimensionless correlation for predicting Int₃ from the wind tunnel deposition velocity data for the four surfaces for nonreentrainment conditions. The dimensionless correlation is

$$\begin{aligned} \text{Int}_3 = - \exp \left\{ -23.667 + 5.555 \ln \frac{d}{z_0} \right. \\ - 0.07681 \left(\ln \frac{d}{z_0} \right)^2 + 0.9722 \ln \frac{u_*}{v_t} \\ + 0.03799 \left(\ln \frac{u_*}{v_t} \right)^2 - 2.254 \ln \frac{D}{u_* z_0} \\ \left. - 3.724 \ln \frac{\rho_p u_* d^2}{18 v z_0} \right\} \end{aligned} \quad (11)$$

in which d is particle diameter, z₀ is roughness height, and ρ_p is particle density of 1.5 g/cm³. Since experimental observations have not been made for other particle densities, the assumption is made that the surface integral resistance Int₃ is independent of particle density.

Int₂ and Int₁

Resistance integrals Int₂ and Int₁ for heights greater than 1 cm were evaluated using Eq. (5) and Bussinger³ et al., atmospheric diffusion correlations for stable, neutral and unstable conditions. The assumption was that particle eddy diffusivity was equal to the eddy diffusivity of air momentum. Since these correlations do not include any canopy effect on eddy diffusivity in box 2, Int₂ and Int₁ were combined into a single resistance integral. Resistance integrals were calculated from 1 cm up to 1 m, and combined with Int₃ in order to predict deposition velocities, K_{1-m}, for a 1 m reference concentration height.

DEPOSITION VELOCITIES AT 1 m

Deposition velocities, K_{1-m}, have been predicted⁴ as a function of particle diameter from 10⁻² to 100 μm, friction velocities u_{*} from 10 to 200 cm/sec, roughness height z₀ from 10⁻³ to 10 cm, particle density from 1 to 11.5 g/cm³, and atmospheric stabilities for Obukhov's lengths from -10 to +10 m. These predictions indicate that deposition velocities can vary several orders of magnitude from about 10⁻³ to 10 cm/sec and increase with both an increase in roughness height z₀ and friction velocity u_{*}. Roughness height and friction velocity are defined from the velocity profile above the ground surface:

$$u = \frac{u_*}{k} \ln \left(\frac{z + z_0}{z_0} \right), \quad (12)$$

where u is the measured velocity, z is the measured height above ground, and k is von Kármán's constant of 0.4.

Typical predicted deposition velocities are shown in Figures 1 through 3. In Figure 1, deposition velocities for a particle density of one are shown as a function of friction velocity for a constant roughness height of 3 cm. This is a typical height for the Hanford Reservation. Deposition velocities, K_{1-m}, increase rapidly with an increase in friction velocity from 10 to 200 cm/sec and are a function of particle diameter from 10⁻² to 100 μm. Minimum deposition velocities occur for

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FIGURE 1. Predicted Deposition Velocities at 1 m Height for Particle Density of 1 g/cm^3 and $z_0 = 3.0 \text{ cm}$.

particle diameters from about 0.2 to 0.4 μm . For smaller particles, deposition velocities increase because of increased mass transfer caused by Brownian diffusion. For larger particles, deposition velocities increase because of increased mass transfer caused by eddy diffusion and gravity settling. In all cases, predicted deposition velocities are greater than predicted by simple gravity settling, v_T .

Predicted deposition velocities are a minor function of atmospheric stability. There are two curves for each friction velocity. The upper curve is for an unstable atmosphere with an Obukhov's length of -10 m and the lower curve is for a stable atmosphere with an Obukhov's length of 10 m .

Deposition velocities are shown for a constant friction velocity of 10 cm/sec in Figure 2 and 30 cm/sec in Figure 3. Curves are shown for particle densities of $1, 4$, and 11.5 g/cm^3 and roughness heights of $10^{-3}, 10^{-1}, 3$, and 10 cm . In all cases, predicted deposition velocities are greater than predicted by simple gravity settling.

Deposition velocities increase with an increase in both surface roughness and particle density as well as with the previously shown increase in friction velocity. However,

deposition velocities are independent of particle density for small particles where Brownian diffusion controls mass transfer. Brownian diffusion is controlling in the particle size region in which the three density curves merge.

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FIGURE 2. Predicted Deposition Velocities at 1 m for $u_* = 10 \text{ cm/sec}$ and Particle Densities of $1, 4$, and 11.5 g/cm^3 .

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FIGURE 3. Predicted Deposition Velocities at 1 m for $u_* = 30$ cm/sec and Particle Densities of 1, 4, and 11.5 g/cm³.

CONCLUSIONS

Sufficient experimental data should always be recorded when experimentally evaluating deposition velocities so that experimental results can be extrapolated to other field test conditions. Required data include at least friction velocity, roughness height, particle size distributions, and atmospheric stability. In addition, airborne concentration should be measured at several heights in order to determine the vertical concentration profile. If the profile is known, one can determine the relative sensitivity of airborne concentration to height at the height selected for defining the deposition velocity. If airborne concentration is dependent on height, a better approximation than a single deposition velocity must be developed to include the atmospheric delivery capability. These data will be required if more realistic deposition velocity predictions or correlations are ever to be made with multibox mass transfer models.

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PENETRATION OF DEPOSITING PARTICLES THROUGH CRUSHED GRAVEL

G. A. Sehmel and W. H. Hodgson

Airborne particles deposit both on a canopy surface and on the underlying ground surface. The penetration was measured for 0.028 to 1 μm diameter particles depositing through a single layer of 3.8 to 5.1 cm diameter crushed gravel. Penetration increased with increasing particle diameter and decreased with increasing air velocity from 3 to 17 m/sec. The maximum measured penetration was 25%.

INTRODUCTION

Airborne pollutant particle removal rates at the air-surface interface are described by particle deposition velocities. These deposition velocities are defined as the total deposition flux divided by an airborne concentration. Although deposition velocities are a function of particle diameter,¹⁻³ particle density, friction velocity, and aerodynamic surface roughness, little is known about predicting the fraction of depositing particles which penetrate through surface canopies such as forests. Indeed, little is known about penetration even for simple surfaces as crushed gravel. Consequently, the objective of this paper is to report initial penetration measurements through a layer of crushed gravel.

EXPERIMENTAL

Deposition velocities have been measured in wind tunnel studies⁴ using monodispersed particles and different surface types. Currently, deposition velocities are being measured for 3.8 to 5.1 cm diameter crushed gravel. This is the most open surface evaluated to date and has permitted evaluation of particle penetration through the crushed gravel layer to the underlying surface.

The crushed gravel was sized for use as railroad ballast. The gravel passed through a 5.1 cm (2 in.) screen and was retained on a 3.8 cm (1.5 in.) screen. Individual gravel

varied in shape from flats to needles to rounds. Minimum gravel thicknesses were about 0.3 cm with maximum length about 5 cm.

Gravel was randomly placed in the 61 x 61 cm cross section wind tunnel so that there were no obvious open areas. The average depth was 5 cm which indicates that at most only one or two layers of gravel covered the floor. The floor width of 61 cm was covered with gravel for a total length of 7.0 m. Penetration was measured at four gravel test sections at tunnel lengths between 4.7 and 6.3 m. Each test section was located along the axis of the wind tunnel and was 22 cm wide by 30 cm long.

Penetration was measured from a mass balance of depositing uranine particles. Particles introduced into the wind tunnel were generated with a spinning disc aerosol generator* or a collision atomizer.** These particles are water soluble and the water solutions were analyzed fluorimetrically to determine the mass balance.

The gravel and the underlying surface were washed separately at each test section. Precleaned gravel was initially placed on 30 x 22 cm removable rectangular metal strips on the

* Spinning disc generator, Model 8320, Environmental Research Corp., St. Paul, Minn.

** Fluid atomization generator, Model 7300, Environmental Research Corp., St. Paul, Minn.

wind tunnel floor. Gravel also surrounded the test sections. After particle deposition from a maximum air velocity in the wind tunnel of either 3 or 17 m/sec was completed, the surrounding gravel was removed. Next, test section gravel was removed and deposited uranine dissolved in a water wash. Similarly, the corresponding underlying surface was removed and uranine also dissolved in a known volume of water. Wash solutions were analyzed fluorometrically for uranine.

RESULTS AND DISCUSSION

Penetration through the surface layer of gravel was determined for particle diameters from 0.028 μm to 0.9 μm for 3 and 17 m/sec maximum wind speeds in the wind tunnel. As shown in Figure 1, the fraction of particles deposited which penetrated to the underlying surface ranged from 2 to 25%. For both wind speeds, penetration increased with increasing particle diameter. However, average penetration was less for the higher wind speed.

Each data point shown is the average value, with the range of values for all test sections indicated by the error bars. No significant differences were shown between test sections. The error bars suggest that significant penetration differences

between wind speeds occurred only for particle diameters less than about 0.1 μm .

Penetration did not appear to depend on the amount of deposited particles. The deposition layer on either the gravel or underlying surface was small. The average coverage of the gravel (projected area) by deposited particles ranged from 0.03 to 1.0% and that for the underlying surface from 0.02 to 0.2%.

CONCLUSIONS

A significant portion of depositing particles can penetrate through the top layer of 3.8 to 5.1 cm diameter crushed gravel. The penetration is believed to be caused by air flow through the gravel. Extrapolated velocity profile measurements above the gravel surface suggest a significant airflow through the gravel. Similar penetrations could also be expected in vegetative canopies. However, in the taller vegetative canopy penetration would probably also reflect nonsteady-state deposition processes which usually occur in the atmosphere. Consequently, due caution is indicated in obtaining field deposition velocities and interpreting experimental data. Experiments must be conducted to determine total deposition for both steady and nonsteady-state deposition.

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FIGURE 1. Penetration of Deposition Particles Through Surface Layer of 3.8 to 5.1 cm Diameter Crushed Rock.

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DETERMINATION OF AEROSOL DEPOSITION RATES
ON A LAKE SURFACE USING RADON DAUGHTERS

J. A. Young

The chemical and radiochemical composition of atmospheric aerosols is being measured to assist in assessing present and future hazards to the environment presented by radioactive and nonradioactive contaminants. The rates and mechanisms by which airborne particulates are mixed in the atmosphere and transported to and deposited on the earth's surface are also under study.

The concentrations of the radon daughters ^{214}Pb and ^{214}Bi were measured in the atmosphere and in Lake Crescent water in conjunction with wind speed measurements to determine the rate of turbulent deposition of atmospheric aerosols on the lake surface as a function of wind speed.

INTRODUCTION

Average rates of deposition of atmospheric aerosol particles on the earth's surface by wet and dry processes have been determined reasonably well from measurements of radionuclides in rainwater. However, the effect of meteorological conditions, and the nature of the surface, on instantaneous rates of deposition of particulates by turbulent diffusion is still poorly known. The rate of deposition of atmospheric aerosol particles on a water surface over a rather short period of time can be determined as a function of meteorological conditions from the simultaneous measurement of meteorological parameters and the concentrations of the short-lived radon daughters ^{214}Pb (26.8 min) and ^{214}Bi (19.7 min) in the air and in the water. These radionuclides are produced by the decay of ^{222}Rn (3.8 d) gas which emanates from the earth's surface. They rapidly become attached to atmospheric aerosol particles and therefore serve as tracers of the subsequent behavior of these particles.

EXPERIMENTAL

The deposition rates of atmospheric aerosol particles on a lake surface are being determined as functions of wind speed from simultaneous measure-

ments of wind speed and the concentrations of ^{214}Pb and ^{214}Bi in air and water. The investigations were carried out at Lake Crescent, a very clear lake on the Olympic Peninsula, on September 26 and October 1 and 2, 1974. The experiments were performed aboard the Battelle inboard-outboard cruiser.

Air samples for ^{214}Pb and ^{214}Bi analysis were collected 2 ft and 14 ft above the lake surface using HiVol samplers to draw air through IPC filter paper at $1.1 \text{ M}^3 \text{ min}^{-1}$. Lake water samples between the surface and a depth of 20 ft were collected by pumping the water first through parallel filters and then through Al_2O_3 at a rate of $0.034 \text{ M}^3 \text{ min}^{-1}$. Measurements of ^{214}Pb and ^{214}Bi concentrations which were carried out in March of 1973 in Lake Crescent water showed that the ^{214}Pb and ^{214}Bi which deposited on the lake surface decayed essentially completely away before penetrating below 20 ft. All of the samples were counted on a Ge(Li) diode installed aboard the boat.

When the analysis of the data is completed, the ^{214}Bi and ^{214}Pb deposition velocities will be calculated from the measured inventories and the atmospheric concentrations and then plotted as a function of wind speed. Vertical diffusion rates in the lake will also be calculated.

DIRECT MEASUREMENTS OF PARTICULATE DEPOSITION
ON A SNOW SURFACE

P. W. Nickola and R. N. Lee

Zinc sulfide and rhodamine particulate tracers were released simultaneously for one-half hour during a period when the surface was covered by snow. Tracers were collected by air sampling during release and by snow sampling following release. Sensitive laboratory techniques for assaying the snow samples are described. Deposition velocities of about 0.1 cm/sec computed from the field measurements are more than an order of magnitude lower than deposition velocities previously measured over the same field grid. However, the previously determined velocities applied over the gross dry surface (including vegetation) while the velocities from the current approach apply over only the smooth snow surface.

INTRODUCTION

Measurement of dry deposition of a particulate by direct measurement of mass on a deposition surface is generally quite difficult. Considerable problems are encountered in collecting a representative surface area. Further, the relatively great bulk of soil and vegetation in a sample designed to merely collect the surface area presents an almost insurmountable task to the analyst.

The release of a tracer over a snow surface simplifies surface collection and analysis procedures considerably. The surface is a natural one (albeit not the most frequent one in most areas), and affords an opportunity to make direct measurements in a true outdoor atmosphere.

The simultaneous release of two particulate tracers over a snow surface at Hanford has permitted direct measurement of dry deposition. Currently, analysis of these samples is underway. This note briefly describes the sample analysis technique, and presents a cursory data analysis of the samples that have been reduced to date.

THE FIELD EXPERIMENT

Tracers ZnS FP2210* and rhodamine** were released from an elevation of 26 m for a period of 30 min. Air sampling on filters took place on sectors of three arcs concentric about the release point. Distances from the release point were 0.2, 0.8 and 1.6 km. Sampling elevation was 1.5 m.

Wind speeds during tracer release averaged 1.4, 1.9, 2.0 and 2.5 m/sec at elevations of 1.5, 3.0, 6.1 and 24.4 m, respectively. A temperature inversion was present over the snow surface, but near neutral thermal stability persisted above the 6-m elevation. Temperatures during the tracer release period averaged 18.5, 19.2, 19.0 and 18.3°F at elevations of 0.9, 6.1, 15.2 and 30.4 m, respectively.

Snow samples were collected with an essentially flat-bladed snow shovel. An attempt was made to collect about a 1 in. depth of snow, although the area of the sample (the

* Fluorescent particulate ZnS, with mass median diameter of about 5 μ m.

** Rhodamine B particulate, with mass median diameter of about 0.8 μ m.

13 1/2 x 17 1/2 in. shovel blade) was the prime consideration. Since the temperature remained well below freezing throughout the dispersal and collection period, no problem was encountered in scooping up samples and transferring them to polyethylene bags. Depth of snow on the ground was about 4 in.

In an effort to minimize snow sample contamination from use of the same shovel for all samples, the sampling was begun at the greatest distance from the source (3.2 km) and proceeded inward to the closest (0.2 km). A total of only 12 samples were collected in the afternoon following the field test. The subsequent day, 149 more samples were collected. The maximum temperature during this period was 20°F. The samples were then stored in a frozen state.

SAMPLE ASSAY

The "Rankin" assay technique employed for detecting the ZnS fluorescent particulate on the air filters has been used at Hanford for more than 15 years.^{1,2} Assay of the air filters for rhodamine is yet to be completed.

The rhodamine content of each snow sample was determined by removing a 5 ml aliquot from the melted sample. This aliquot, taken within 12 hr of melting, was placed in a glass cuvette for analysis using a Turner Model 110 Fluorometer. Laboratory samples stored at room temperature in plastic bags have been found to undergo a 15% reduction in concentration over a 3-day period. If one assumes a similar decay rate for the field samples, the above procedure should limit the loss of rhodamine to <5%.

The total zinc content of approximately half the snow samples reported in this paper was determined by concentrating the acidified solution and analyzing by conventional atomic absorption. This method has subsequently been abandoned in order to avoid possible concentration changes arising from sample transfer and contamination during prolonged exposure to the laboratory atmosphere. The procedure which has been adopted eliminates several sources of error and reduces the analysis time. This

procedure involves the addition of 0.5 ml of distilled concentrated nitric acid to the sample after removal of an aliquot for rhodamine analysis. The acidified sample is allowed to sit at room temperature for at least 24 hr to insure dissolution of zinc sulfide. An aliquot is then removed for analysis by flameless atomic absorption.

It must be emphasized that the zinc analyses outlined above provide a measure of the total zinc content of the sample. Each source of zinc must therefore be recognized and its contribution to the total zinc content estimated in order to determine the deposition velocity of ZnS. In addition to the tracer, the following potential sources of zinc must be considered:

1. Concentrated acid and contamination of the interior of the bags,
2. Precipitation scavenging of zinc-containing aerosol, and
3. Soil and plant particles.

Analysis of the concentrated acid and simulated field samples has confirmed the negligible contribution of both the acid and storage bags to the total zinc load of the samples. An estimate of the zinc derived from scavenging processes may be obtained from those samples collected at the fringe of the sampling arcs and corresponding to low or nonexistent air concentrations of the tracer. The limited number of samples currently available shows this to be approximately 10 ng/ml. This order is in good agreement with data reported for a Pacific Northwest station as part of an NCAR study³ of the metal content of precipitation. It would appear therefore that zinc leached from soil and plant particles during the acid treatment is of secondary importance.

OBSERVATIONS AND COMMENTS

Inasmuch as chemical analysis of the samples is still underway, no firm conclusions have been reached. However, based on the 62 above background samples already analyzed, some comments and tentative conclusions can be made.

Table 1 lists median deposition velocities (v_d) determined from

TABLE 1. Deposition Velocities Determined from Snow Samples and from Atmospheric Exposures Measured at an Elevation of 1.5 m.

Distance from Source	No. of Samples ZnS Rhod		Deposition Velocity, cm/sec			
			Median		Range	
			ZnS	Rhod	ZnS	Rhod
0.2 km	14	13	2.4	3.7	0-31	<0.6-19
0.8	14	13	0.09	0.16	0-0.7	<0.02-1.7
1.6	7	1	0.09	0.14	0-0.5	-

analysis of several samples from the three arcs where both air and snow samples were taken. (Although analysis of air samples for rhodamine has not been accomplished, the source-normalized concentration was presumed to be the same as for ZnS for this first approach.) The median values are given because a few large values heavily biased the means. The number of samples analyzed and the range of velocities computed are also given in Table 1.

The computed deposition velocities are considerably higher at 0.2 km from the source than at the greater distances. Although nothing obviously went astray during the tracer dispersal processes, gravitational settling may have played a role in the relatively large masses of tracer deposited at 0.2 km.

The v_d values measured at 0.8 and 1.6 km over this smooth snow surface are more than an order of magnitude below those determined through mass balance techniques by Nickola and Clark⁴ on the same field grid. However, the Nickola-Clark v_d values of 1.5 to 5.4 cm/sec applied to the total surface, including the 1- to 2-m tall sagebrush and lower vegetation. Further, the basic surface amidst the larger sagebrush elements was devoid of snow and, hence, was considerably rougher.

The indication that deposition velocities for the smaller and less dense rhodamine particles are greater than those for the ZnS is unexpected. A more careful analysis of the data than has been carried out to date is in order here. One possible source of error is in the rather gross as-

sumption that the zinc background was identical for each snow sample. In fact, the background is a function of the volume of snow collected despite the fact that the deposited mass is a function of surface area only. Another potential source of error is the assumption that the normalized rhodamine atmospheric exposures are identical to the ZnS exposures. Final data reduction and analysis will eliminate both these uncertainties.

One disconcerting observation is worth mentioning. As stated earlier, on the afternoon following the morning field experiment, 12 snow samples were collected. Samples from these same 12 locations were also taken on the following day. To date, 7 of these 12 pairs of samples have been assayed. In all seven cases, the v_d determined from the earlier date is as large or larger than that determined from the later date snow. The ratio of first day to second day v_d values ranges from 1.0 to 6.0 (with one ratio actually infinity since the second day's sample revealed only a background quantity of zinc). The median ratio was 2.5. A possible explanation for the difference is that about 0.8 inches of fresh snow fell on the night between snow sample collections. Although an effort was made to collect about an inch of snow surface on both days, the possibility does exist that the snow shovellers just did not dig deeply enough on the second day. Nonetheless, even if the mass of the tracers collected in the snow were multiplied by 2.5, the computed v_d values would still remain about an order of magnitude below the Nickola-Clark v_d values mentioned earlier.

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PROSPECTS FOR TRACER MASS BALANCE COMPUTATIONS
ON A DENSE THREE-DIMENSIONAL SAMPLING GRID

P. W. Nickola and G. H. Clark*

Despite the use of a densely instrumented sampling grid (including towers spaced at 8° increments) obtaining a precise mass balance through several vertical planes remains to a great extent fortuitious. This observation suggests caution in the specification of other mean plume characteristics developed on the basis of sparse sampling in space and/or time.

INTRODUCTION

As sampling of plumes with aircraft becomes more commonplace, the procedure, and the need, of specifying mean plume properties based on traverses through the real plume also becomes more commonplace. The temptation exists to presume that one or a few traverses through a plume are sufficient to permit a "good" definition of the mean plume. Generally speaking, this is not true. Nickola and Clark¹ and Brown, et al.² have touched on some of the statistical aspects of this time and space sampling problem.

In the past year, Nickola and Clark have further considered the problem of mass balance computations on the basis of exposures measured on a fixed three dimensional sampling grid. In this situation the plume is sampled continuously so that only the errors associated with spatial distribution of samplers remain. Yet obtaining a representative estimate of the mass of tracer passing a vertical plane remains a problem.

THE FIELD SAMPLING GRID AND FIELD EXPERIMENTS

The study reported here is an outgrowth of a field study designed to measure deposition of a particulate tracer (FP) by comparison with a non-depositing inert gas tracer (krypton⁸⁵). The experimental technique

involved a simultaneous half-hour release of both tracers for the same 26-m elevation source location.

The field sampling grid is depicted in Figure 1. Sampling of particulates took place on more than 600 filters placed on sectors of six arcs concentric (or nearly so) about the tracer release tower. The angle embraced by the sampling arcs was approximately 80°. The basic sampling elevation was 1.5 m. Distances of the sampling arcs from the release point were approximately 0.2, 0.4, 0.8, 1.2, 1.6 and 3.2 km.* Additionally five towers spaced at 8° increments were found on each of the arcs at 0.2, 0.8, 1.6, and 3.2 km. Samplers on these towers reached to elevations of 33, 42, 62 and 62 m, respectively. Vacuum for sampling was supplied by pumps powered by small internal combustion engines.

The grid instrumented with 127 krypton⁸⁵ detectors was restricted to three arcs. Basic sampling level again was at a height of 1.5 m. The azimuths embraced by krypton detectors were 58, 40 and 15° at 0.2, 0.8 and 1.6 km respectively. Tower-mounted detectors were operative only at 0.2 and 0.8 km.

Vertical profiles of wind speed, needed in the mass balance computations, were measured on the 122-m

* Arcs at 0.8, 1.6 and 3.2 km are actually concentric about a point 100 m due south of the release point. Thus, the distance from release point to arc varies with azimuth.

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FIGURE 1. The Hanford Diffusion Field Grid.

tower at the source point and on a nearby 24-m tower. Additional details of the experiments were reported by Nickola and Clark.³

ANALYSIS

As indicated in Figure 1, the field sampling network is one which could be considered reasonably dense. And since during the field experiment of interest the plume axis appeared to be nearly centered on the portion of the grid instrumented with towers, it was expected that a reasonable mass balance of tracers from sampling arc to sampling arc could be computed. Such was not the case.

Figure 2 shows results of various methods of computing mass balance for both the krypton and for the FP. Since vertical sampling of krypton was restricted to two arcs, there are fewer data points for krypton than for FP. The first two computational procedures identified at the top of Figure 2 were reasonably sophisticated.

The latter two were more naive approaches. All started, of course, with the same set of input data. Note that although there is a reasonably wide scatter (up to 16%) in the fraction computed for a given tracer at a given distance, the shape of all curves is consistent. It would seem that the input data force the same general conclusion irrespective of the computational approach.

Reaching the same conclusion irrespective of reasonable approach used could be considered comforting. However, note that all percentages are below 100%--and frequently well below.

The FP data plotted in Figure 3 show why the mass balances of the previous figure were consistently low. Tower mounted samplers on which the mass balance computations are based are spaced at 8° increments. However, at 1.5 m (the so-called "ground-level" sampler), spacing was smaller as is indicated at the top of Figure 3. At each arc, exposures from all ground

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FIGURE 2. Fraction of Released Tracer Passing Vertical Plane Versus Distance. Data based on tower and 1.5-m samplers.

level samplers were summed and multiplied by the appropriate spacing between samplers to obtain a crosswind integrated exposure (CIE). Then

CIE's were recomputed using only samplers at 8° increments--simulating spacing between towers. The number of CIE's which could be computed based on the limited sets of samples depended, of course, on the spacing between samplers.

Examination of Figure 3 shows a wide variation in computed CIE based merely on which sampler happened to be selected as the starting point in the 8° increment. For instance, at a distance of 0.40 km, "movement" of the starting point in sampling by 4° resulted in a more than two-fold change in calculated CIE.

The circled points connected by dotted lines in Figure 3 represent CIE's computed from ground level samplers at the actual tower locations. Note that at 0.20 and 0.84 km they are the lowest possible computed for those distances.

These circled data points are plotted as x's on Figure 2. Except at 3.2 km, note that there is reasonable agreement with the "objective Computer Program" points. If no directional shear or deposition were involved, the mass balance points and

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FIGURE 3. Fraction of Mean Crosswind Integrated Exposure (CIE) Determined from 8° Sampler Spacing as Percentage of CIE Determined from All Available Samplers at 1.5-m Elevation.

the ground level CIE's measured at the tower base should reasonably coincide when plotted on this normalized scale.

CONCLUSIONS

Despite what can be considered a dense sampling grid, the evidence of Figures 2 and 3 points out the fact that obtaining a mass balance near

100% is to a considerable extent fortuitous. This observation suggests using considerable caution in generating other mean plume characteristics (e.g., plume centerline concentration and location, plume σ_y or σ_z , crosswind integrated exposure) on the basis of a few samples which are sparse with respect to both space and time.

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SOME NEW MODELS FOR DRY DEPOSITION OF AEROSOL
PARTICLES FROM THE ATMOSPHERE

W. G. N. Slinn

An extended summary is presented of new models for dry deposition recently reported by the author: a new multilayer scheme is presented; deposition to a smooth surface is modeled assuming that particles are convected across the viscous sublayer by the leading edge of turbulent eddies; a simple flux balance gives an estimate of dry deposition in a canopy. Theory is compared with experimental results and adequate agreement is found by varying the free parameters of the models. Future studies should be able to establish these parameters from theoretical analyses.

INTRODUCTION

The two primary methods by which the atmosphere cleanses itself of pollution are known as precipitation scavenging (or wet deposition) and dry deposition. During this reporting period some new models were developed for dry deposition and it is the purpose of this extended abstract to indicate these results. Details can be found in the original publications.^{1,2}

MULTILAYER MODEL

One of the complexities of dry deposition is derived from the many orders of magnitude which the length scale spans. This is illustrated on the LHS of Figure 1. In an attempt to simplify the problem it is convenient to divide the total height into various layers, introduced here as shown in Figure 1. In each layer a fewer number of independent variables should be sufficient. In response to the frequent inquiry to identify the layer in which dry deposition is rate limited, one concludes qualitatively (see the RHS of Figure 1) or more quantitatively (see Figure 2) that dry deposition could be rate limited in any of the layers A through E. This depends on the height of the release, the stability of the atmosphere, the density of the canopy and detail of the pollutant

and the surface. For example, the deposition of noble gases is rate limited in the edaphic layer, whereas for stratospheric bomb debris it is rate limited by diffusion in the stratosphere or stratospheric-tropospheric exchange, i.e., in the A layer. In what follows attention will be focused on the C and D layer.

DEPOSITION LAYER

A number of models are presently available to predict dry deposition to a smooth surface. However, all rely on the convective diffusion equation near the surface where their applicability is open to serious question, and on either the particle, free-flight model or an effective particle diffusivity; therefore, the models are basically empirical fits to data. The model that has been developed during the reporting period relies on Joukowski's model of turbulence (see Figure 3) and the premise [following Owen³] that particles are convected across the viscous sublayer by the leading edge of Joukowski's "caterpillar tread" model of turbulent eddies. The result for the deposition velocity is

$$v_d = v_s + \frac{1}{\beta} \frac{u_*^2}{u} E_j \quad (1)$$

where v_s is the particle's settling velocity; β is a numerical factor $O(1)$;

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FIGURE 1. A Suggested Division of the Total Dry Deposition Problem and a Qualitative Indication of Where the Process May Be Rate Limited.

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FIGURE 2. Indications of the Time Scales Involved in the Dry Deposition Process, Evaluated Either from $\delta t = \delta z/v$ or $\delta t = (\delta z)^2/D$, and Illustrating that the Process May Be Rate Limited in Various Layers, Depending on Where the Displacement Time is Largest.

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FIGURE 3. A Schematic Illustration of Joukowski's "Caterpillar Tread" Developing in a Turbulent Boundary Layer.

u_* is the friction velocity; $\bar{u}$ is the mean fluid velocity above the boundary layer and E_j is the collection efficiency for a viscous jet impinging on a flat surface. Using a semi-empirical expression for E_j for a surface that is a much better heat conductor than air (so that thermophoresis is negligible) leads to

$$v_d = v_s + 10^3 \frac{\text{cm}}{\text{sec}} \left(\frac{\dot{m}''}{1 \text{ g cm}^{-2} \text{ sec}^{-1}} \right) + \frac{u_*^2}{\beta \bar{u}} \left[10^{-3}/St + \frac{\beta}{\gamma} Sc^{-0.6} \right] \quad (2)$$

where $\dot{m}''$ is the water vapor flux to the surface (positive if condensation); St is the Stokes number based on the time scale of the turbulent fluid, $St = \tau u_*^2 / \nu$ where τ is the particle relaxation time and ν is the kinematic viscosity for air; $Sc = \nu/D$ is the Schmidt number in which D is the particle diffusivity; and γ is another numerical factor $O(1)$. The result (2) is shown in Figure 4 using $\beta = \gamma = 0.4$ and compared with experimental data of Sehmel⁴ and Clough.⁵

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FIGURE 4. The Deposition Velocity as Given by Equation (2) and Some Data Points from Sehmel⁴ and Clough.⁵

Notice that (2) is evaluated using $\rho_p = 1 \text{ g cm}^{-3}$ but Sehmel's data is for $\rho_p = 1.5 \text{ g cm}^{-3}$.

CANOPY LAYER

Apparently no other models are available to interpret deposition in a canopy. A simple one developed during the reporting period follows from the flux balance shown in Figure 5. The result for the deposition velocity when steady state conditions are developed is

$$v_d = v_s + \frac{\epsilon}{1 + \epsilon} \alpha u_* + \frac{1}{1 + \epsilon} \frac{u_*^2}{\bar{u}_g} \frac{E_j}{\beta} \quad (3)$$

where ϵ depends on the canopy's filtration efficiency

$$\epsilon = H E_c' \bar{u}_c / (\alpha u_*). \quad (4)$$

Here H is the height of the canopy; E_c' is the filtration efficiency per unit length of canopy; $\bar{u}_c$ is the mean wind speed in the canopy; α is a constant $O(1)$; and $\bar{u}_g$ is the mean (geostrophic) wind above the canopy. For $\epsilon \rightarrow 0$, (3) reduces to (1). For $\epsilon \rightarrow \infty$, (3) becomes

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FIGURE 5. A Schematic Diagram for the Pollutant Fluxes in a Canopy. γ^{-1} in the figure should be replaced by β^{-1} to be consistent with notation in the text.

$$v_d = v_s + \alpha u_* \quad (5)$$

that is, the deposition velocity to an infinitely efficient canopy becomes independent of particle size. If simultaneous resuspension occurs it is suggested that the jet deposition in (3) be modified to yield

$$v_d = v_s + \frac{\epsilon}{1 + \epsilon} \alpha u_* + \frac{\delta(d)}{1 + \epsilon} \frac{u_*^2}{\bar{u}_g} \frac{E_j}{\beta} \quad (6)$$

where $\delta(d)$ depends on the particle diameter, d , and δ is zero for no effective jet deposition. Equation (6) is used to model Chamberlain's⁶ deposition data to artificial, sticky grass and to real grass shown in Figure 6. For this fit the parameters chosen were $\alpha = 1$, $\epsilon = 0.04$ and $\delta = 0$ (no effective jet deposition) for the lower curve and for the artificial grass, all three terms in (6) were used and with $\delta = \beta = 0.4$. Future work should be directed toward establishing these parameters theoretically.

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FIGURE 6. Deposition with Simultaneous Resuspension; Data from Chamberlain.⁶

SUMMARY

New models have been developed to describe dry deposition of aerosol particles in the deposition and canopy layers. Adequate fit to experimental

data has been achieved by varying parameters of the models and, of more importance, it appears that the physical models are adequate to allow evaluation of the parameters from entirely theoretical analyses.

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RESUSPENSION OF PARTICLES

Particles, once airborne, then deposited, can again become airborne when wind stresses are sufficiently high. Research in this area, to establish the important variables and their interrelationships, will be ultimately used to determine the exposure to man from such secondary sources and to set permissible limits on ground contamination. We are primarily using areas contaminated inadvertently with radioisotopes from various processes, but have also performed initial studies using a tracer placed on an instrumented plot of prairie-like terrain.

An important part of this work is to develop models which account for particles resuspended and deposited. One important variable is the wind speed, but the interaction between the primary particle of interest and the bulk soil particles is also very important. Diffusion and dispersion in the atmosphere, once particles are resuspended, are important in diluting the airborne material; hence, this work interfaces directly with diffusion studies. The physics of wind blown sand and fine particles is also studied to determine the mechanisms of resuspension and transport. Studies sponsored by others on the nature and release of radioactive particles in accidents and fires are closely related to DBER programs.

During 1974 good progress was made in several areas. The size characteristics of airborne dust as a function of wind speed were measured, and the frequency and diurnal variation in the United States compiled and analyzed. A possible explanation of certain anomalous profiles above a contaminated zone was found. An effort was made to correlate available resuspension rates with surface roughness. Modeling efforts continued with success in relating downwind exposure to resuspension and deposition sequences over large areas. Some initial measurements were made of DDT resuspended over a forested area after application of the pesticide.

- **PARTICLE RESUSPENSION AND TRANSLOCATION**
- **BATTELLE MEMORIAL INSTITUTE PHYSICAL SCIENCES PROGRAM**

SOME ASPECTS OF THE RESUSPENSION PROBLEM

W. G. N. Slinn

Three particular topics in the resuspension problem are addressed in this extended abstract of work reported elsewhere: an aerosol kinetic equation for the soil particle size distribution; the horizontal flux of particles during a dust storm; and relating the soil erodibility to the resuspension factor.

INTRODUCTION

One of the sources of atmospheric pollution is the entrainment of particles from surfaces. The total "resuspension" problem is complex, as is indicated in Figure 1. Recently the author has considered some features of this problem, generally those features associated with the solid lines on the left hand side of Figure 1. The purpose of this extended abstract is to briefly report on these studies. Further details can be found in the original publications.^{1,2,3}

SOIL PARTICLE SIZE DISTRIBUTIONS

Kolmogoroff⁴ has demonstrated theoretically that if an original soil particle is subjected to a large number of independent and random breakages then the resulting particles will have a log-normal distribution of their sizes. Since various data, e.g., see Figure 2, have demonstrated the inadequacy of this prediction, it is of interest to formulate the problem of soil particle breakage accounting for subsequent agglomeration of the shattered particles. This has been done during the reporting period, but the result will not be given here since the resulting equation is highly intractable. For the case of only breakage and no agglomeration it reduces to

$$\frac{\partial f}{\partial t} dm = -bf dm + \int_m^\infty dM P(M \rightarrow m) f(M, t) \quad (1)$$

where $f(M, t) dm$ is the number of particles at time t whose mass is from m to $m + dm$; b is the breakage rate and $P(M \rightarrow m)$ is the probability that upon breakage of a particle of mass M , one of the pieces will have mass m . It is interesting that for $P(M \rightarrow m) = 2 dm/M$ the steady-state solution to (1) is the Junge particle size distribution function:⁷

$$f(r) = c_2/r^4. \quad (2)$$

Further studies of this formulation will be made in the near future.

DUST FLUX DURING WIND STORMS

The total horizontal flux of dust during a dust storm is customarily divided into three components: creep, saltation and suspension. However, clearly this division lacks a physically rational and mathematically quantitative basis. During this reporting period a new estimate has been made for the total horizontal flux of dust. The estimate is based on the assumption that near the surface during a dust storm, the vertical flux of horizontal momentum $\tau' = \rho(u_*')^2$ is carried essentially

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FIGURE 1. An Indication of Some of the Complexities of the Resuspension Problem.

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FIGURE 2. Data from Bagnold⁵ and Chepil⁶ Showing Measured Size Distributions of Specific Sand (Bagnold) and Airborne (Chepil) Soil Particles.

by the particles. Then the horizontal flux of particles (in units, for example, of $g \text{ cm}^{-2} \text{ sec}^{-1}$) is given by

$$\Gamma = \tau' / (\bar{v}_s + \alpha u_*') \quad (3)$$

where $\bar{v}_s$ is the average settling velocity of the particles and α is a constant 0(1). From (3) and an estimate of the height of the saltation layer, the mass flux of particles per unit crosswind distance is found to be

$$q = c \frac{\tau'}{g} (v_s + \alpha u_*') \quad (4)$$

where g is the acceleration of gravity and c is a constant 0(1). Equation (4) reduces to Bagnold's result⁵ only if $\alpha u_*' \gg \bar{v}_s$ (i.e., intense winds and/or small particles). Insufficient data has been found to test (3) or (4).

To obtain the vertical flux of particles it was found that the convective diffusion equation inadequately portrays the experimental results and it was concluded that the best available procedure is use of experimental data to estimate the horizontal flux as a function of altitude. For example, Chepil⁸ gives that the weight of dust in unit volume of air varies as height according to

$$c = c_0 (z_0/z)^{0.28} \quad (5)$$

where c_0 depends on the erodibility of the soil, the wind speed, etc.

THE RESUSPENSION FACTOR

During the reporting period some advances have been made relating the soil erodibility to the suspension flux in general, and to the resuspension factor in particular. The soil erodibility, usually expressed in units of tons of soil per acre per month, is given in charts, tables and functional form as^{9,10}

$$\dot{E}'' = E(I', R', C', L', V) \quad (6)$$

where I' is the soil erodibility index; R' is the soil ridge roughness factor; C' is a climatic (or meteorological) factor; L' is the field length along the prevailing wind erosion direction; and V is the equivalent quantity of vegetative cover. By approximating the air concentration downwind of a strewn field, (6) can be transformed with the aid of a Gaussian plume model evaluated at a distance downwind of the field so that $\sigma_y \sigma_z \sim A_s$, where A_s is the area of the contaminant spill, to yield the resuspension factor $K = \chi/\chi_s$ (the ratio of air to surface contaminant concentrations). The result is

$$K \approx \frac{3.5 \times 10^{-9} \text{ cm sec}^{-1}}{1 \text{ ton acre}^{-1} \text{ mo}^{-1}} \left(\frac{r \dot{E}}{2\pi \bar{u} \delta} \right) \quad (7)$$

In Equation (7) r is the fraction of the horizontal flux which travels at the height of the sampler [which can be estimated from (5)], $\bar{u}$ is the mean wind speed at the sampler height and δ is the depth to which the contaminant has penetrated the soil. Other formulae, for the air concentration at other heights and downwind distances, are given in the more complete reports.^{1,2,3}

CONCLUSIONS

An aerosol particle kinetic equation has been suggested which may be able to explain the observed soil particle size distribution. For negligible coagulation and steady state conditions, the solution to the equation for a specific breaking process was found to be the Junge distribution. In future studies other solutions will be sought. The horizontal flux of soil particles during a dust storm was estimated and an expression for the resuspension factor in terms of the soil erodibility was presented. Future studies in this area should concentrate on relating measured soil erodibility and particle size distributions to measured airborne particle size distributions and turbulence characteristics.

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INITIAL PARTICLE RESUSPENSION RATES--A FIELD EXPERIMENT
USING TRACER PARTICLES

G. A. Sehmel and F. D. Lloyd

Resuspension rates of a submicrometer calcium molybdate particle were measured. The area onto which the particles were "seeded" was a lightly vegetated area about 23 m in radius. Airborne concentrations were measured vertically over the center of the plot up to a height of 6.1 m. Measurements indicated that the trace particles reached higher elevations. Resuspension rates were from 10^{-10} to 10^{-8} fraction per second.

INTRODUCTION

Hazardous materials such as plutonium have inadvertently been deposited on environmental surfaces. Concern for possible human exposure to these materials continues after deposition since removal rates by wind and mechanical stresses are unknown, but are needed to predict airborne concentrations from the potential sources. If resuspension rates are large, the material has the potential to again create a local or downwind airborne health hazard. Inhalation hazards have been historically evaluated in terms of a resuspension factor defined as the ratio of airborne concentration per unit air volume at breathing heights to the ground surface contamination per unit area. Resuspension factors are often in the range¹ of 10^{-5} to 10^{-9} m^{-1} . However, reported² resuspension factors have varied by over 11 orders of magnitude and have not yielded any generalized predictability scheme.

The use of resuspension factors is of doubtful value in developing generalized resuspension models, since resuspension physics are ignored. Airborne concentrations should be related to the extent of upwind and not local surface concentration. In addition, resuspension factors do not indicate how rapidly contamination is removed from the surface and transported downwind. Predictions of downwind airborne concentrations require that resuspension rates be determined.

This paper reports initial particle resuspension rates and resuspension factors from a controlled surface source strength of submicrometer tracer particles simulating pollutant contamination.

EXPERIMENTAL

Wind caused particle resuspension rates from environmental surfaces are being determined using inert tracer particles as a substitute for radioactive particles such as plutonium. An inert tracer is used since placing radioactive particles in the environment could develop into a potential hazard comparable to the potential hazard being simulated. There are several other advantages in using inert tracers; they can be deposited at a known source strength in terms of particle diameter, time at which particles were initially deposited, and quantity of tracer per unit area.

The tracer resuspension site is a lightly vegetated area on the Hanford Reservation which has a roughness height, z_0 , of 3.4 cm. On October 2, 1973, a tracer suspension of submicrometer calcium molybdate was sprayed on the ground in a circle of 22.9 m radius around a sampling tower 6.1 m in height. The average surface concentration on the ground was 0.62 g of elemental molybdenum per m^2 .

Airborne wind resuspended tracer was subsequently sampled with a wind-vane-oriented rotating cowl and cascade particle impactor system³ shown in Figure 1. Each system consists of

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FIGURE 1. Tracer Source and Resuspension Sampling Tower.

a high volume air sampler drawing air through a cascade impactor* at a flow rate of $0.57 \text{ m}^3/\text{min}$. The impactor separates "respirable" particles into nominal diameters for unit density spheres of 7, 3.3, 2.0, 1.1 μm and smaller particles on the back-up filter. Particles entering the cylindrical sample inlet of the cowl would either fall to the internal cowl surfaces or be drawn up into the cascade impactor. In this paper, particles falling in the cowl will be called "non-respirable" and particles entering the impactor will be called "respirable," although this definition is not in strict accord with lung physiological considerations.

* Andersen 2000, Inc., model 65-100 High Volume Sampler Head, P.O. Box 20769, Atlanta, Georgia 30320.

Rotating cowl-impactor systems were mounted at tower heights of 0.3, 0.9, 1.8, 3.0, and 6.1 m to determine resuspended tracer as a function of respirable particle diameter and height. Initially, from October 2 to November 4, and for a second experiment from November 16 to December 16, 1973, only one impactor was located at each height and was operated continuously. For the third experiment from January 16 to February 8, 1974, three impactor-cowl air sampling systems were located at each height as shown. In this last experiment, air sampling systems were turned on automatically as a function of wind speed. Wind speed was measured with the wind sensor located 2.1 m above ground. One air sampling system at each height was turned on for wind speeds between 1.3 and 3.6 m/sec, the second for wind speeds between 3.6 and 5.8 m/sec and the third for wind speeds greater than 5.8 m/sec. The maximum recorded wind gust was 20.1 m/sec.

The initial experiment was started the day tracer was deposited and periodically continued for 566 hr from October 2 to November 4. Samplers were deactivated during precipitation conditions.

At the end of the experiment, filters were weighed to determine the total mass of collected material at each stage. Afterwards, the filters were leached to remove the molybdenum tracer and the leach solutions were analyzed by x-ray fluorescence for molybdenum.

Sampling is continuing to determine resuspension rate changes with time and as a function of airborne soil particle diameter.

RESULTS

Tracer resuspension rates were calculated from a mass balance of airborne molybdenum. Vertical concentration profiles of airborne tracer showed the largest concentration at the lowest sampler, and the least concentration at the highest samplers. However, the resuspended tracer plume did extend above the top sampler. The effects of underestimating resuspension rates by plume noncontainment are presently believed to be small. More important to data interpretation is that concentration profiles did

not always show a continuously decreasing concentration from the lowest to highest sampler, either for total tracer collected in the impactor or for individual particle size collections within the impactor. These concentration profiles will be reported when additional confirming data are obtained. At present, resuspension rates for total respirable particles from the integrated concentration profiles and resuspension factors based on the airborne concentrations at 1.8 m height are reported.

Resuspension rates were calculated⁴ from true fluxes, ground surface concentration, and sampling time. Average resuspension rates for "respirable" particles were calculated for all particles entering each impactor. A resuspension rate for the "non-respirable" particles that settled in the cowl was also determined for the last experiment. In this case, soil collected in all three impactors at each height was composited and analyzed for tracer.

Resuspension factors, RF, were calculated using the respirable airborne concentration, χ , at 1.8 m height. The resuspension factor is the ratio of the airborne concentration per m^3 divided by the ground source concentration per m^2 .

Resuspension rates and resuspension factors increased rapidly with an increase in wind speed. The wind speed dependency was determined only for the third experiment. At the start of this test period, tracer had been on the ground for 3.4 months. Figure 2 shows resuspension data for this January 16 to February 8, 1974 sampling period. Data points are plotted at the lower limit of each wind speed interval. A horizontal line through each data point extends to the upper limit of that wind speed interval.

Resuspension rates for "respirable" particles increased from 2.0×10^{-10} fraction resuspended/sec for the 1.3 to 3.6 m/sec wind speed interval to 2.2×10^{-8} fraction resuspended/sec for the 5.8 to 20.1 m/sec wind speed interval. This is an increase of two orders of magnitude. The overall increase in resuspension rates is a non-linear function of wind speed. However, a linear approximation was made

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FIGURE 2. Resuspension Rates and Factors as a Function of Wind Speed.

between the upper two data points. For wind speeds greater than 3.6 m/sec, resuspension rates increased with wind speed to the 6.5 power.

A resuspension rate of 1.3×10^{-11} fraction resuspended/sec for "non-respirable" particles was determined for the entire sampling period of this third experiment. However, no wind speed dependency could be determined for these larger particles due to the sampling technique. Large particles could be blown into the cowls even if a high-volume air sampler was not operating.

The total source concentration on the ground appears to be diminishing rather slowly. Since there are 3.15×10^7 sec/year, only 0.1 of the source would be resuspended even if wind speeds were continuously greater than 5.8 m/sec for an entire year. Resuspension rates and the fraction of tracer removed in a year could be much greater if the soil were mechanically disturbed.

Resuspension factors also increase rapidly as a function of wind speed. As shown in Figure 2 resuspension factors are between 10^{-10} to 10^{-8} m^{-1} and are in the average range reported

in the literature^{1,2} for larger contaminated areas. However, these resuspension factors apply only to the center of a 22.9 m radius contaminated area. Resuspension factors for larger contaminated areas should be larger.

There appears to be some relationship between resuspension rates and resuspension factors. The resuspension factors are based upon the concentration at 1.8 m height and resuspension rates are based upon airborne concentrations integrated as a function of height to 6.1 m. As shown in Figure 3, a nearly linear relationship exists for the data obtained as a function of wind speed intervals. However, the relationship is not universal. Resuspension rates and factors for continuous sampling during the first two time periods do not show the same trend. For these two cases, resuspension rates decreased as the resuspension factors increased.

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FIGURE 3. Resuspension Rates as a Function of Resuspension Factors.

Comparisons including all data suggest that average resuspension rates were of the order of 10^{-10} fraction resuspended/sec for the entire 4 month sampling time.

CONCLUSIONS

Resuspension rates for test conditions were of the order of 10^{-10} to 10^{-8} fraction resuspended/sec. The resuspension source will not be significantly depleted for many years unless resuspension rates are increased by mechanically disturbing the soil. It is unknown if reported rates can be extrapolated to other field conditions of either contamination or vegetative cover. There is reason to anticipate successful extrapolation since resuspension factors are in the same range of 10^{-10} to 10^{-8} m^{-1} often reported in the literature. These are the first field resuspension rate data that have been reported for soils. Consequently with the supporting agreement of resuspension factors, these data are a basis upon which to make realistic resuspension rate estimates.

Although there are some apparent relationships between resuspension rates and resuspension factors, there are insufficient data to draw general conclusions. At most, the conclusion is that resuspension factors between 10^{-10} and 10^{-9} m^{-1} are comparable to resuspension rates between 10^{-10} and 10^{-8} fraction resuspended/sec.

Resuspension rates for nondisturbed soils are a nonlinear function of wind speed. For wind speeds greater than 3.6 m/sec, resuspension rates increased with wind speed to the 6.5 power. For soils disturbed during worker activity, the same rapid increase in resuspension may also occur with a greater availability of contamination for resuspension. This increase suggests that even if average airborne concentrations are below maximum permissible concentrations, inhalation exposure could become significant during periods of high winds.

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EXPERIMENTAL DESIGN FOR DETERMINING TRACER RESUSPENSION RATES AS A FUNCTION OF WIND SPEED AND ATMOSPHERIC STABILITY

G. A. Sehmel and F. D. Lloyd

Equipment is being installed in the field to determine tracer resuspension rates as a function of both wind speed increments and atmospheric stability. This experiment will permit evaluation of resuspension rate reproducibility and determination of upward diffusion parameters for resuspending particles.

INTRODUCTION

Particle resuspension rates¹ have been determined using tracer particles at one site as a function of wind speed increments. Those are the only rates evaluated under field conditions. Resuspension rates increase with increasing wind speed but only one resuspension rate for each wind speed could be determined for any time period. Consequently, reproducibility of resuspension rates for the same time period is needed in order to determine confidence limits for resuspension rates. Furthermore, both resuspension rates and upward diffusion parameters for resuspension

are needed to model downwind transport of resuspended material. The objectives of this experiment are to:

- Determine resuspension rates as a function of wind speed at a second site from that initially used.
- Determine by replicate experimentation resuspension rate confidence limits at this second site.
- Determine the upward diffusion coefficients of resuspended particulates as a function of both atmospheric stability and wind speed increments.

EXPERIMENTAL DESIGN

The design for this field resuspension experiment is schematically shown in Figure 1. This figure is a view of an annular tracer source area and air sampling towers located in and downwind of the source. The annular area radii are 15 and 45 m, located around a central air sampling tower 37 m in height. The annulus extends from 188° to 330°.

High volume air samplers and cascade particle impactor-cowl² systems will be mounted on the towers to determine the total airborne tracer flux and thus tracer resuspension rates. Air samplers will be turned on automatically as a function of selected wind direction sectors, wind speed increments, and temperature lapse rates to indicate atmospheric stability.

There are two sets of air sampling towers radiating out from the central 37 m tower. Towers from one set are indicated by circles and are located on 200°, 225°, and 250° radial lines from the central tower in order to determine resuspension rates for high wind speeds which come from about 225°. The other set of air sampling towers indicated by triangles are located on 275°, 300°, and 325° radial lines, in order to determine resuspension rates for lower speeds of the prevailing winds which come from about 300°.

There are four air sampling towers for each wind direction with the central tower used for both wind directions. The three outer 9 m high towers are 7.5 m from the tracer edge. These three towers are designed to determine tracer resuspension rate reproducibility between cross wind sites. The four air sampling towers along both 225° and 300° are designed to determine tracer resuspension rate changes with distance in the respective wind direction. Air samplers from these four towers will determine

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FIGURE 1. Plan View Annular Resuspension Experiment with Source and Towers.

vertical airborne concentration profiles of resuspended tracer, which will be used to determine upward diffusion parameters for resuspending particles.

SUMMARY

A field resuspension experiment has been designed to measure particle resuspension rates during high wind speeds which come from about 225° and for lower speeds of prevailing winds which come from about 300°. Air sampling towers have been erected in the selected tracer area. Equipment assembly is directed toward beginning the experiment before the high wind months of February-March. The tracer area is covered mainly with cheat grass.

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INITIAL CORRELATION OF PARTICLE RESUSPENSION RATES
AS A FUNCTION OF SURFACE ROUGHNESS HEIGHT

G. A. Sehmel

Particle resuspension rates from a smooth surface were experimentally measured for monodispersed particles as a function of time. At a constant friction velocity, resuspension rates decreased nonexponentially with time. This decrease under controlled conditions suggests that the concept of a resuspension weathering exponential half-life may be questioned under some conditions. An attempt is made to grossly correlate resuspension rates with aerodynamic roughness heights ranging from a smooth surface to that for a forest. An approximate correlation is indicated but caution is recommended in its use since many variables have been assumed constant.

INTRODUCTION

Particle resuspension rates are needed to predict airborne concentrations of particle pollutants originating from surfaces in nature, and potential downwind health hazards. Unfortunately, too little is known about resuspension physics to permit reliable prediction of resuspension rates. Even resuspension rates for controlled laboratory experiments have not been reported. Consequently, this paper reports resuspension rates obtained as a function of friction velocity and resuspension time for resuspension of monodisperse particles from a smooth surface. Subsequently, available resuspension rate data will be plotted as a function of surface roughness to develop an initial guide for predicting resuspension rates in the environment. It is recognized at the outset that several other parameters may be simultaneously affecting resuspension. Some of these are humidity, the chemical nature of particulates, electrostatic effects, particle involvement, soil-canopy matrix, mode of application, and particle size distributions.

EXPERIMENTAL

Particle resuspension rates were measured in controlled laboratory ex-

periments using 10 μm diameter monodispersed uranine particles. Particles were generated with a spinning disc aerosol generator and were carried in air drawn upward into a 15 m long, 2.93 cm inside diameter vertical aluminum tube. The average flow velocity was 12.3 m/sec corresponding to a Reynold's number of 2.4×10^4 and a friction velocity of 70 cm/sec. Some of the particles were deposited on the 1.38 m^2 of internal tube wall surface and those still remaining airborne were collected on a filter at the exit of the 15 m length of tubing. After the deposition phase of the experiment was completed, the exit filter was replaced with a new filter. The resuspension phase of the experiment consisted of drawing clean air through the tube at higher constant air flow rates and collecting on the exit filter the resuspended particles from the tube walls. Periodically, air flow was stopped, and the filter was removed each time and replaced with a new filter. At the completion of the resuspension experiment each filter and the internal surface of the 15 m long tube were washed with water to dissolve uranine. Wash solutions were analyzed fluorimetrically for uranine. Resuspension rates were calculated from a uranine mass balance during the resuspension interval.

RESULTS

Resuspension rates ranged from 10^{-6} to 10^{-3} fraction resuspended/sec, but were dependent upon air flow rates and resuspension time. In Figure 1, resuspension rates are shown for a constant friction velocity of 1.24 m/sec. Resuspension rates decreased from 10^{-4} to 10^{-5} fraction resuspended/sec. After 121 min, 17% of the deposited particles had been resuspended. This decrease in resuspension rates in a nonlinear, nonexponential function of time. Consequently, the decay can not be described by a simple weathering exponential half-time as is often quoted¹ in the resuspension literature.

Resuspension rates increased with increasing air velocities. In Figure 2, resuspension rates decreased as previously shown with an increase in sampling time for a constant air velocity. The rate decreased from 3.3×10^{-4} to 1.9×10^{-5} fraction resuspended/sec during 40 min of an average flow rate of 16.7 m/sec. However, resuspension rates increased by a factor of 4.9, from 1.9×10^{-5} to 9.3×10^{-5} fraction resuspended/sec when the flow rate was increased only 9%. Afterward, the resuspension rate for the higher flow rate also decreased with time. At the end of 73 min, 23% of the originally deposited particles had been resuspended.

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FIGURE 1. Resuspension Rates of $10 \mu\text{m}$ Diameter Particles at a Constant Air Flow Rate Within a 2.93 cm ID Vertical Tube.

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FIGURE 2. Resuspension Rates of $10 \mu\text{m}$ Diameter Particles at Two Successively Larger Air Flow Rates Within 2.93 cm ID Tube (Air Flow Rate Increase is 8.8%).

The increased resuspension rate for a friction velocity of 0.97 m/sec suggests that turbulent eddies are penetrating to within at least $10 \mu\text{m}$ of the wall and resuspending particles. Ten μm corresponds to a dimensionless distance y^+ of only 0.7 from the wall surface. The quantity $y^+ = yu_*/\nu$ where y is the friction velocity, and ν is the kinematic viscosity. If eddies are penetrating this close to the wall surface, the often quoted² concept of a laminar sublayer next to the wall of thickness y^+ of 5 should not be applied in theoretically modeling particle resuspension.

Particles were resuspended at lower friction velocities following resuspension at higher friction velocities. In Figure 3, resuspension rates decreased from 1.1×10^{-3} to 5.3×10^{-6} fraction resuspended/sec in 13 min at a constant friction velocity of 39.3 m/sec. At a lower friction velocity of 1.55 m/sec the resuspension rate remained constant at 5×10^{-6} fraction resuspended/sec for times between 13 and 58 min. For a smaller friction velocity of 1.26 m/sec, resuspension rates again decreased with increasing resuspension time. However, when the flow rate was increased to a friction velocity

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FIGURE 3. Resuspension Rates of 10 μ m Diameter Particles at Four Successive Air Flow Rates Within a 15 m Vertical 2.93 cm ID Tube (Particles Initially Deposited at $U_* = 0.70$ m/sec).

of 2.28 m/sec after 133 min total resuspension time, the resuspension rate again increased. At the end of the experiment, 53% of the originally deposited particles had been resuspended.

DATA CORRELATION

Guidelines for estimating resuspension rates are needed based upon existing experimental resuspension rate data. Resuspension rate data have now been reported for uranine particles from a smooth surface, ZnS from an asphalt surface,³ submicrometer molybdenum tracer from a vegetated desert soil⁴ and for DDT from a forest.⁵

Each of these surfaces has a much different estimated surface roughness height, z_0 , from 4×10^{-3} cm for the smooth surface to 100 cm for a forest.⁶ Roughness height is calculated from the log-linear velocity profile and is the height at which the ex-

trapolated velocity profile reaches zero velocity.

Ranges of measured resuspension rates are shown plotted as a function of measured or estimated surface roughness heights in Figure 4. Resuspension rates may vary seven orders of magnitude from 10^{-10} to 10^{-3} fraction resuspended/sec. The practical significance of these numbers can be realized by remembering that a year is 3.2×10^7 sec. The data are interpreted to suggest that resuspension rates decrease with a surface roughness increase for the three smaller surface roughnesses. However, the measured resuspension rates for DDT sprayed on a forest are two orders of magnitude greater than for the desert soil. This is an unexpected and unexplained increase in resuspension rates. The possible explanation of the increase may be the increased resuspension caused by tree movement in the wind. Various other gross differences in controlling variables and experimental factors may be influencing the results.

CONCLUSIONS

Resuspension rates obtained for smooth surfaces in the laboratory were for larger friction velocities than usually occur in the atmosphere and are for a surface much smoother than most surfaces in nature. Nevertheless, the salient features shown from these laboratory resuspension experiments may also apply to outdoor terrain surfaces. These features are that: 1) resuspension rates increase with increasing wind speed, 2) resuspension rates decrease with time for a constant air flow rate, and 3) the decrease suggests that an exponential decrease in source strength should be questioned. These laboratory obtained resuspension rates agree qualitatively with the concept that resuspension rates should decrease with increasing surface roughness. The apparent correlation should be used with extreme caution, since the data are so extremely limited. Nevertheless, the correlation does give some justification for selecting resuspension rates until additional data are obtained.

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FIGURE 4. Initial Correlation of Wind Caused Resuspension Rates.

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AIRBORNE DUST SIZE DISTRIBUTIONS
AS A FUNCTION OF WIND SPEED

G. A. Sehmel

Airborne dust size distributions were measured as a function of wind speed increments, using particle cascade impactors. Airborne dust concentrations increased nonlinearly as wind speed increased. Increases are proportional to the 0.6 to 3.2 power of wind speed. Resuspended soil can contribute significantly towards exceeding air quality standards.

INTRODUCTION

Even though it is possible to theoretically model resuspension from surfaces, experimental data are needed to validate such models. One modeling concept in describing particle resuspension is that contaminants lose their identity as discrete particles, and thus, when resuspended, are attached to host soil particles. If the physics of soil resuspension were understood more adequately, it is anticipated that the same relationships could be used as a simple basis for developing predictive particle resuspension models. Consequently, soil resuspension was investigated by experimentally determining the rate at which airborne dust concentrations in the respirable size range increased with increasing wind speed.

EXPERIMENTAL PROCEDURE

Airborne concentrations of respirable sized particles were determined as a function of wind speed increments near a pond on the Hanford reservation. Particle sizing was with cascade particle sizing impactor-cowl¹ systems mounted on three air sampling towers at sampling elevations from 0.3 to 29.9 m. Three towers were used with two main towers east of the pond and a third tower upwind west of the pond. Prevailing winds were from the west. The smaller main tower had impactor-cowl systems at heights of 0.3, 0.9, 1.8, 3.0, and 6.1 m, which sampled winds from all direction.

The second tower east of the pond was 66 m north of the smaller tower. This second tower had impactor-cowl systems at heights of 0.3, 0.9, 3.0, 11, and 29.9 m, but in this case only sampled west winds coming from 170 to 340°. The upwind tower only operated concurrent with the taller main tower and had impactor-cowl systems at height of 0.3, 1.8, and 6.1 m.

Wind instrumentation located near the two main towers was used to turn on the impactor-cowl samplers as a function of wind speed and wind direction measured at a height of 2.1 m. Impactor-cowl samplers would be turned on for one of two groupings of wind speed increments. One grouping was for wind speeds between 1.3 to 3.6 m/sec, 3.6 to 5.8 m/sec, and 5.8 to 20.1 m/sec. The second grouping was 4.5 to 5.4 m/sec, 5.4 to 6.7 m/sec, and 6.7 to 9.4 m/sec.

Particle sizing was with Anderson 2000 Inc., cascade particle impactors* sampling at 0.57 m³/min. Airborne concentrations for each stage of the cascade particle impactors were determined by weighing the fiberglass collection media both before and after particle collection. In both cases, filters were allowed to equilibrate with laboratory humidity before filters were weighed.

Concentrations are reported as the number distribution, (number/cm³)/μm, evaluated at three particle diameters

* Andersen 2000 Inc., model 65-100
High-Volume Sampler Head, P.O. Box
20769, Atlanta, Georgia 30320.

of 1.1, 1.9, and 3.6 μm . These diameters are based upon the mid-points between 50% cut-off diameters of the cascade impactor of 0.78, 1.4, 2.3, and 5.0 μm for an assumed airborne, soil density of two g/cm^3 .

AIRBORNE CONCENTRATIONS IN SIZE RANGES

Increases in the airborne number distribution of particles as a function of wind speed are shown in Figure 1 for particle diameters of 1.1, 1.9, and 3.6 μm . In each case, the data point is shown at the lower limit of each wind speed increment. Wind speed increments are indicated by the horizontal lines extending to the right from each data point.

Each data point represents the average value over all sampling heights for the indicated tower. Although one expects some concentration changes with height, significant change in number distributions were not observed with the cascade-impactor sampling technique used. Samples were all obtained for selected wind speeds occurring within time periods from 23 to 135 days. It is anticipated that concentration changes with height will be measurable in planned short term measurements with optical particle counting and sizing equipment.

Particle number concentrations increased rapidly and reproducibly with an increase in wind speed. In the left portion of the figure, concentrations increased with wind speed to the 2.9 power for wind speeds between 4.5 to 9.4 m/sec. The same slope adequately represents data for all three particle diameters and sampling times. Concentrations in the summer months from May 6, to September 18, 1974, were greater than for the spring months from March 4 to April 25, 1974, but were less than for late winter months from January 16 to February 8, 1974, for different wind speed increments.

The remaining data in the three sections of the figure are for measurements using larger wind speed increments for air sampling. The solid lines in each of the three portions of the figure were calculated to have the same slopes while minimizing the data point deviations around the solid lines. Concentrations increased with wind speed to the 3.2 power.

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FIGURE 1. Airborne Soil Particle Concentrations as a Function of Particle Diameter, Wind Speed, Sampling Season and Sampling Site, 1974.

Airborne soil concentrations at the two towers east of the pond increased with wind speed to either the 0.6 or 2.9 to 3.2 power of wind speed. Any deviations between calculated curves for one tower can probably be attributed to seasonal variations of vegetation cover, temperature, and surface soil moisture content. There also may be differences in air concentration between these two sites which are 66 m apart. The differences may not be significant, however, since both tower sites were not operated simultaneously.

Differences in air concentrations were measured as a function of tower separation for the June 6 to July 5, 1974 data shown in the right portion of the figure. In this case, air was sampled simultaneously both upwind and downwind of the pond for 170° to 340° winds blowing across the pond. Upwind concentrations were greater

except for the $1.1\ \mu\text{m}$ data point at $5.8\ \text{m/sec}$. The significant difference is that upwind concentrations continually increased with wind speed to the first power as compared with increases to the power of 0.6 and 3.2 for the downwind tower. Reasons for differences between upwind and downwind concentrations can only be assumed at present. However, differences are probably caused by different wind speeds at the upwind and downwind towers, deposition of particles in the pond area between towers, and, probably of more significance, the open soil available for resuspension adjacent to the upwind tower.

AIRBORNE MASS LOADING

Average airborne mass loadings were calculated for each wind speed collected within impactors. Dust loading in $\mu\text{g}/\text{m}^3$ are shown as a function of wind speed increments in Figure 2. Dust loadings range from 13 to $360\ \mu\text{g}/\text{m}^3$ as shown for two lines representing sampling times from March 4 to April 25, 1974, and May 6 to September 18, 1974. Mass loadings of respirable particles increased with wind speed to the 2.9 power for wind speeds greater than $4.5\ \text{m/sec}$.

CONCLUSIONS

Airborne concentration of respirable dust particles increased as wind speed increased. For wind speeds above about $4\ \text{m/sec}$, concentrations increased with wind speed to about the third power. Concentration of $1.1\ \mu\text{m}$ particles ranged from 5 to $50/\text{cm}^3/\mu\text{m}$; $1.9\ \mu\text{m}$ particles ranged from 0.5 to $8/\text{cm}^3/\mu\text{m}$; $3.6\ \mu\text{m}$ particles ranged from 0.02 to $0.5/\text{cm}^3/\mu\text{m}$. In each case, particle concentrations

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FIGURE 2. Airborne Dust Loading Collected Within Impactors as a Function of Wind Speed.

increased an order of magnitude with increases in wind speed from 1.3 to $9\ \text{m/sec}$.

Number distributions when converted to total mass loading of respirable particles range from 13 to $360\ \mu\text{g}/\text{m}^3$. These mass loadings during short time intervals indicate that resuspended dust loadings during high winds can exceed annual and 24 hr ambient air quality standards² for particulates. The annual primary standard is $75\ \mu\text{g}/\text{m}^3$ and the secondary standard is $60\ \mu\text{g}/\text{m}^3$. The 24 hr primary standard not to be exceeded more than once a year is $260\ \mu\text{g}/\text{m}^3$ and the secondary standard is $150\ \mu\text{g}/\text{m}^3$. It is obvious by comparing measured dust concentrations in an unpolluted region that resuspended soil can exceed national air quality standards for particulates.

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RESUSPENSION OF PLUTONIUM AT ROCKY FLATS

G. A. Sehmel and F. D. Lloyd

In a July, 1973 field experiment at Rocky Flats, vertical airborne plutonium concentration profiles were determined as a function of respirable particle size. The principal sampling site was a 30-m meteorological tower which was instrumented at several heights with self-orienting, high volume cascade impactor-cowl systems. Wind speed instrumentation activated selected impactors as a function of wind speed. Airborne plutonium concentrations and concentrations on airborne soil were measured as a function of both height and wind speed. Below 10 m, concentrations decreased with height. Above this height to 30 m, an increase suggested that an elevated upwind source could be contributing to the airborne concentration. The maximum airborne concentration measured was 3.7×10^{-15} Ci/m³. The maximum concentration per gram of gross airborne soil was 50 pCi/g. All concentrations were far below the maximum permissible concentrations.

INTRODUCTION

As part of our general model development study to predict wind-caused resuspension of particulate surface contaminants, we initially analyzed reported airborne plutonium concentrations and their relationship to wind speed and direction at Rocky Flats, Colorado. The ground source of plutonium was an area near the plant where liquid from storage drums containing contaminated cutting oils and solvents had contaminated a relatively small area of soil. From material balance calculations it was estimated¹ that about 5000 gal containing 86 g of plutonium (5.4 Ci) had leaked sometime between initial use in 1958 and drum removal in 1968. In 1969, fill was applied to help contain the activity and the 120 m x 113 m rectangle on which the drums were originally stored was covered with an asphalt pad completed in November 1969. Since the primary ground source had been covered by fill and asphalt, the ground source for the analysis of airborne plutonium was plutonium which was resuspended and deposited prior to covering.

Initial correlation efforts demonstrated that airborne plutonium concentrations depend on wind speed and direction. Model predictions indicated that during weekly sampling periods at Rocky Flats sampling station S-8 from July 1970 to January 1971, the average airborne plutonium concentrations during each hour increased with wind speed to the 2.1 power.² Subsequent to this period, depletion of the source and certain mechanical operations including ditching are believed to have changed the source characteristic, such that new on-site measurements were desirable to describe resuspension and its relationship to wind speed and direction for this modified source.³ Information on size distribution of airborne plutonium-bearing particles was likewise needed to estimate the consequences in the environment of the resuspended fraction.

Objectives of this Rocky Flats resuspension experiment were to determine vertical airborne concentrations of plutonium and plutonium attached

to the mass of airborne soil, to determine particle size distributions of plutonium and associated airborne soil at three sampling sites, and to relate measurements to meteorological variables and surface contamination. Experimental results have been reported⁴ and are summarized here.

EXPERIMENTAL

Overall Experiment Design

Large volume air samplers were located and operated within the generally designated contaminated area east of the original oil storage area. Because we were interested in relating airborne concentrations at different heights with wind speed, samplers were placed at selected elevations. Cascade impactor samplers were used for particle sizing. Relationship of resuspension to wind speed was to be determined by windvane-oriented cascade impactor samplers which were energized only at pre-selected wind speed ranges. Each particle impactor system was assembled from a 0.57 m³/min flow rate Andersen 2000 Inc.* high volume cascade impactor and a windvane-oriented rotating cowl attached to the impactor. These impactors separate particles into nominal diameter ranges of 1.1, 2.0, 3.3, and ≥ 7 μ m which are stage 50% cut-off diameters for unit density spheres.

Sites

Airborne plutonium concentrations were measured at three tower sampling sites, A, AB, and B. Site A was near the originally contaminated oil storage area and site B was furthest away to the east. All air samplers at these two sites operated independently of wind speed. Site A was along the eastern security fence and site B was near the cattle fence. Site AB was between the eastern security fence and the cattle fence, and was 392 m from site A and 277 m from site B. At site AB, a 30-m meteorological tower was erected which was instrumented at several heights with high volume cascade impactor-cowl systems. Wind speed instrumentation turned on these impactor-cowl systems as a function of wind speed.

One group of impactor-cowl systems located at 0.3, 1, 3, 11, and 30 m heights operated for all wind speeds. A second group located at 0.3, 1, 2, 4 and 10 m heights operated for wind speeds at the 1 m height between 4.1 to 6.3 m/sec. The third group located at 0.3, 1, 4 and 11 m heights operated for wind speeds greater than 6.3 m/sec. (The maximum recorded wind gust was 9.8 m/sec.)

For comparison to airborne activity at plant sites during July 11 through July 21, and July 24 through 31, a background sampling site was also operated continuously from July 18 through August 2. This site was 8 miles into the mountains in the Cold Creek Valley at an elevation of 8100 ft. Elevations at sites A, AB, and B were about 5900 to 5950 ft. The background sampler was a cascade impactor mounted on the roof of a house in a forest. The sampler was about 20 ft above ground among trees about 30 ft tall.

RESULTS AND DISCUSSION

Airborne plutonium concentrations are reported in aCi/m³ (10^{-18} Ci/m³) of ²³⁹Pu in air and pCi/g (10^{-12} Ci/g) of ²³⁹Pu per g of airborne soil. The maximum airborne concentration was 3700 aCi/m³ (3.7 fCi/m³) ²³⁹Pu. The maximum concentration on the airborne soil was 50 pCi/g total airborne soil and 69 pCi/g of 2 μ m diameter impactor size fraction of airborne soil. One pCi/g corresponds to 2.2 dpm/g. All airborne concentrations were significantly less than maximum permissible concentrations⁵ in air of 2×10^{-12} μ Ci/cm³ of soluble ²³⁹Pu for occupational exposure in a 40-hr work week or of 6×10^{-13} μ Ci/cm³ or 600,000 aCi/m³ for nonoccupational exposure in a 168-hr week.

The functional relationship between resuspension and wind speed could not be developed as unequivocally as was initially anticipated as an experimental objective. This was due in part to the inadvertent loss of about a fifth of the samples in a step in the chemical separation of plutonium. Unfortunately, the higher wind speed samples (>6.3 m/sec) were among those which were lost.

* Andersen 2000, Inc., Model 65-100 High-Volume Sampler Head, P.O. Box 20769, AMF, Atlanta, Georgia 30320.

Background Sample Measurement

Fallout levels of ^{239}Pu entering the area were estimated from the cascade impactor operated in the mountains. There was no detectable activity on the 7, 3.3, and 1.1 μm stages and there were no radiochemical results for the 2 μm stage. The only reported plutonium activity was on the back-up filter which nominally collects only submicrometer particles. The airborne concentration was low at $4 \pm 88\%$ aCi/m³, and the concentration in the airborne soil on the back-up filter was $0.7 \pm 88\%$ pCi/g.

Elevated Source

In Figure 1, vertical concentration profiles of total plutonium are compared in this summary for site AB. In general, air concentrations calculated from impactor collection are greater than those calculated from high volume air sampler collection. It is not known if these concentration differences are caused by using two types of air samplers, sampling for nonidentical time periods, or if a true spatial horizontal concentration difference exists.

Both profiles indicate concentrations are greater at 30 m than at 10 m. This increased concentration with height is unexpected when one considers only ground resuspension as a plutonium source. For ground resuspension as a source, profile shapes should be similar below 10 m to 1) the dashed curve when resuspension occurs

near the samplers, or 2) the solid curve below 10 m when resuspension occurs at a distance from samplers and deposition depletes the airborne plume near ground. Both profiles suggest a secondary source at an elevated height.

Average Airborne Concentrations

Airborne plutonium concentrations are a function of both sampling height, soil, and particle diameter. Airborne concentrations in aCi/m³ are shown in Figure 2 for each stage of the cascade impactor. At site AB there are unexpectedly high concentrations for constant particle diameters at both 30 m and 3 m heights.

There was no plutonium in the upper stages of the cascade impactor back-ground site. We regard this as evidence that any plutonium activity associated with the larger particles is from resuspension. Resuspension of submicrometer particles is also occurring at Rocky Flats, but as is shown, increased concentrations are significant only at the 30 m height of site AB.

The complete data⁴ in general show a decrease in airborne activity with increasing distance from site A. This decrease corresponds to a greater distance from the oil storage area which was the principal source of the original ground contamination. However, significant deviation in the air concentration profiles with both distance and height did occur and at

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FIGURE 1. Site AB: Airborne ^{239}Pu Concentrations.

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FIGURE 2. Airborne ^{239}Pu Concentrations at Site AB as a Function of Impactor Collection Site.

present must be attributed to sampling "hot" particles which contained relatively more plutonium than average. This increase in airborne activity occurred at both site AB and B.

At site AB, "hot" particles may have been present in the $2.0\ \mu\text{m}$ size range as indicated in Figure 3. In this case, the concentration at 1 m of site AB is one to two orders of magnitude greater than at other elevations. More important to the "hot" particle concept is the concentration at the 10 m height at site B. This concentration of $230\ \text{aCi}/\text{m}^3$ was the highest concentration of $2\ \mu\text{m}$ particles measured at any location. Since this location was the most remote⁶ from both ground and the oil storage area, other relatively "hot" particles could also be escaping the plant boundary. However, due caution is indicated in interpreting this "hot" particle data. The total of 6 dpm collected on the $2\ \mu\text{m}$ stage or $230\ \text{aCi}/\text{m}^3$ is much less than the maximum permissible air concentration of $2 \times 10^{-6}\ \mu\text{Ci}/\text{m}^3$ (occupational). It is conceivable that the majority of this plutonium could have been attached to one soil particle.

Concentration on Airborne Soil

Plutonium-239 concentration on respirable airborne soil was determined to relate air concentrations in resuspension model development to similar ground concentrations if ground

contamination data become available. Airborne concentrations on soil particles in the respirable size range are shown as pCi/g of airborne soil in Figure 4. In general, pCi/g profiles are similar to profiles for aCi/m^3 . The same general exceptions occur in the pCi/g profiles. A secondary elevated source above 10 m is indicated for $7\ \mu\text{m}$, $2.0\ \mu\text{m}$, and back-up filter particles.

The maximum concentration shown in Figure 3 for $3.3\ \mu\text{m}$ particles is 34 pCi/g at the 3 m height of site AB. Similarly, the "hot" particle at 10 m of site B produced an average concentration of 69 pCi/g of $2\ \mu\text{m}$ particles.

Total Concentration

Even with the limited data, it is evident that airborne ^{239}Pu concentrations increased with wind speed. In Figure 5, total airborne concentrations are shown calculated from impactors operating continuously, for wind speeds between 4.1 to 6.3 m/sec, and for wind speeds from 6.3 to 9.8 m/sec. Concentrations for wind speeds from 4.1 to 6.3 m/sec are definitely greater than for the continuous samplers. However, $2\ \sigma$ counting statistic error limits are too large to determine the wind speed dependency.

Nevertheless, an attempt to approximate airborne plutonium concentration and consequently the resuspension rate dependency upon wind

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FIGURE 3. Airborne ^{239}Pu Concentrations from Impactor $2.0\ \mu\text{m}$ Stage Collections.

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FIGURE 4. ^{239}Pu Concentration on Airborne Soil at Site AB as a Function of Impactor Collection Site.

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FIGURE 5. Total Airborne ^{239}Pu Concentration at AB Site as a Function of Wind Speed.

speed was made for 7 μm stage collections at the 0.3 m height of site AB. The ^{239}Pu dpm on the 4.1 to 6.3 m/sec sample and the dpm on the 6.3 to 9.8 m/sec sample were subtracted from the dpm on the continuous sample in order to approximate the expected dpm for sampling winds up to 4.1 m/sec. An expected concentration was then calculated for wind speeds below 4.1 m/sec.

A straight line through all three data points suggests that ^{239}Pu resuspension rates increase with wind speed to the 5.9 power. However, extreme caution is needed in generalizing this tentative conclusion since 2σ error bars are very large. Also, the wind

speed dependency for smaller particles at 0.3 m height could not be estimated since there was less plutonium on the continuous impactor than on both the mid and high wind speed impactors. This nonuniformity probably reflects both a nonuniform source and also possible "hot" particle effects.

CONCLUSIONS

Both ^{238}Pu and ^{239}Pu resuspension occurred at Rocky Flats during July 1973, but all airborne plutonium was significantly below maximum permissible concentrations in air. However, ^{239}Pu was deposited on each stage of cascade impactors which indicates that most plutonium was resuspended while attached to larger host soil particles.

Airborne plutonium concentrations appear to have originated both from a secondary elevated source as well as from resuspended ground sources. The differences in vertical concentration profiles as a function of sampling site, sampling time, sampling method, as well as nominal particle size will make it difficult to satisfactorily model plutonium resuspension processes at Rocky Flats. This modeling difficulty will be especially true since there appear to be "hot" particles which contain relatively more plutonium than average airborne plutonium containing particles. Additional resuspension research as compared to monitoring efforts at Rocky Flats should consider using much larger arrays of air samplers in order to adequately define the resuspension rates of these "hot" plutonium particles.

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A POSSIBLE EXPLANATION OF APPARENT ANOMALOUS
AIRBORNE CONCENTRATION PROFILES OF PLUTONIUM
AT ROCKY FLATS

G. A. Sehmel

Airborne samples of plutonium were analyzed for isotopic content. These samples were obtained at elevations from 0.3 to 30 m above ground between the eastern security fence and cattle fence at Rocky Flats. Isotopic constituents are different at 30 m than for lower elevations, and samples of the higher elevations may represent a secondary source.

INTRODUCTION

A resuspension experiment¹ was conducted at Rocky Flats during July 1973, to determine airborne plutonium concentrations as a function of wind speed, particle diameter, and sampling site. The maximum airborne concentration of ^{239}Pu measured was 3.7×10^{-15} Ci/m³ which is significantly less than the maximum permissible concentration of 6×10^{-13} Ci/m³ for nonoccupational exposure in a 168-hr week.² The main sampling site was a 30 m tower east of the plant approximately mid-way between the eastern security fence and cattle fence. Airborne plutonium concentrations were measured at elevations from 0.3 to 30 m above ground. However, airborne concentrations did not decrease as expected from 10 to 30 m, but concentration increased

with elevation. This increase suggested that an elevated plutonium source could be contributing to airborne plutonium concentration in addition to plutonium resuspended from local ground sources. To resolve the possibility of an elevated source, plutonium in air samples collected at each height was also analyzed by mass spectroscopy for isotopic Pu content. Samples analyzed were metal discs upon which plutonium from air filter samples was electroplated after air filter dissolution, chemical separations, and tracing with ^{236}Pu for ^{239}Pu analysis.

RESULTS

Only seven samples of higher ^{239}Pu content were analyzed for isotopic plutonium content. These samples were

selected from plutonium collected on stages of cascade particle impactors mounted at 0.3, 1, 3, and 30 m elevations. Atom percentages of ^{239}Pu , ^{240}Pu , and ^{241}Pu are shown in Table 1 as a function of aerodynamic particle diameter.

There are differences in the isotopic plutonium compositions at 30 m compared to lower elevations of 0.3, 1, and 3 m. Isotopic compositions in these lower elevations might be similar to that on the surrounding contaminated ground if resuspension is the source of airborne plutonium. In this case, isotopic compositions of resuspended plutonium ranged from 5.73 to 5.97 a/o for ^{240}Pu and from 0.31 to 0.35 a/o for ^{241}Pu . These ranges include the 5.93 a/o ^{240}Pu and 0.33 a/o ^{241}Pu obtained for the back-up filter at a 30 m elevation. In contrast, the isotopic compositions for 2 and 3.3 μm particles at 30 m were outside these ranges.

At present there are no satisfactory explanations for the differences in isotopic composition at 30 m. Indeed it is difficult to conceive of processes by which isotopic ratios would vary so greatly as a function of particle diameter. Nevertheless, different isotopic ratios did occur for 2 and 3.3 μm particles at the 30 m elevation. One possible explanation of the high concentration and different isotopic ratio at 30 m might be unplanned low level plant releases. Prevailing westerly winds would have blown airborne plant releases toward the sampling site.

However, this does not seem too probable since the plutonium processed at Rocky Flats during July 1973 had an average isotopic content³ as follows:

^{239}Pu , 93.64 a/o

^{240}Pu , 5.89 a/o

^{241}Pu , 0.43 a/o.

Within the variation of individual samples, it is inconclusive that the plutonium at an elevation of 30 m (2 and 3.3 μm size fractions) might come from plant effluents. However, this possibility is not excluded.

Another observation of interest was that plutonium on individual soil particles was in particulate form much smaller than the impactor size diameter of the fraction collected. Fission track analysis⁴ of the original filter sample specimens showed that all plutonium present (assumed to be PuO_2) would have particle diameters of less than 0.25 μm .

CONCLUSIONS

An elevated source of airborne plutonium existed in July 1973, which was originally suggested by greater airborne concentrations at 30 m than at 10 m. Additional data on isotopic ratios of plutonium support the hypothesis that a source of airborne plutonium was present in addition to that of locally resuspended soil.

TABLE 1. Plutonium Isotopic Composition.

Sampling Height m	Impactor 50% Cut-Off Diameter μm	Atom %		
		^{239}Pu	^{240}Pu	^{241}Pu
30	3.3	94.74 $\pm$.14	5.02 $\pm$.10	0.24 $\pm$.05
30	2.0	90.9 $\pm$.3	8.6 $\pm$.2	0.5 $\pm$.1
30	*	93.74 $\pm$.07	5.93 $\pm$.12	0.33 $\pm$.07
3	7	93.94 $\pm$.04	5.73 $\pm$.11	0.33 $\pm$.06
3	3.3	93.72 $\pm$.10	5.97 $\pm$.12	0.31 $\pm$.06
1	2.0	93.93 $\pm$.06	5.76 $\pm$.11	0.31 $\pm$.06
0.3	7	93.83 $\pm$.19	5.82 $\pm$.11	0.35 $\pm$.07

*Back up filter

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RESUSPENSION OF RADIOACTIVITY INDUCED BY BURNING CROP STUBBLE

G. A. Sehmel and M. M. Orgill

Airborne particulates from burning fields outside the Hanford reservation were sampled with an aircraft-mounted air filter. Radioactivity concentrations in air samples collected from the smoke plume were compared to concentrations in ambient background air samples. Airborne concentrations of ^{7}Be and $^{95}\text{Zr-Nb}$ in the smoke were 1.2 to 4.6 times ambient background levels.

INTRODUCTION

Pollutant particles deposited on outdoor surfaces are resuspended by wind stresses¹⁻³ and mechanical activity.⁴ Burning contaminated vegetation and vegetation cover is another process by which pollutant particles might become airborne. Fire-induced resuspension of deposited radioactive particles may be of concern after long-term fallout of low-level contaminants has resulted in appreciable surface contamination. This condition might exist near a fuel reprocessing plant after many years of operation.

The objective of this experiment was to determine if accumulated chronic low-level radioactive surface contamination would be resuspended from burning crop lands. Resuspension was to be evaluated by determining if the concentration of radioactivity was greater in the smoke plume than in the surrounding air.

EXPERIMENTAL

During the fall, farmers in the Columbia Basin east of the Hanford Reservation burn acres of crop land to sterilize the land and destroy

accumulated biomass before the next planting. This crop land could have become slightly contaminated by depositing airborne radioactive particulates. These potential particulates include ^7Be from cosmic ray origin, fallout, and low-level releases of fission products from the Hanford Reservation.

Airborne particulate concentrations were determined from air filter samples collected on October 21 and October 22, 1974. Samples were collected on IPC filter paper supported in an aircraft-mounted filter holder. Ram air produced an average flow rate of $36.4 \text{ m}^3/\text{min}$ through the filters. Total flow through each filter was measured with an inline, negligible-pressure-drop vane flowmeter* for sampling times from 132 to 150 min.

Airborne concentrations of both smoke plume and background clean air were obtained the same day at elevations from about 150 to 180 m above ground. Sampling sites were over private land east of the Columbia River. On October 21, the burning fields were principally northeast of the Hanford Reservation near the Othello radar site at a location approximately 17 km from the Columbia River. This was a windy day with the daily average winds NW at 3.2 m/sec and a peak gust of 9.4 m/sec from the NW. Wind velocities were lower on October 22, with an average velocity of 2.0 m/sec from the west and a peak gust of 5.8 m/sec from the NNW. The burning field was approximately NE of the 300 area and about 2 km from the Columbia River. On both days background samples were taken between Eltopia and Sprague Lake which are about 20 to 130 km from the Hanford Reservation.

Filter samples were analyzed by gamma spectroscopy which indicated that both ^7Be and $^{95}\text{Zr-Nb}$ were present in the smoke sample and background sample for each day.

RESULTS

Both ^7Be and $^{95}\text{Zr-Nb}$ were identified on each filter sample. Concentrations in dpm for each radioiso-

tope were normalized to the lowest concentration, which was for $^{95}\text{Zr-Nb}$ in the background sample on October 22. Normalized airborne activity concentration ratios are shown in Table 1. Concentrations of ^7Be in the smoke plume are 1.2 to 1.3 times the concentration in ambient background air. Cosmogenic ^7Be deposited on the fields was released or resuspended to a degree during burning of the fields. Similarly, $^{95}\text{Zr-Nb}$ concentrations in the smoke plume were 1.4 to 4.6 times the concentration in ambient background air. In both cases, increased airborne concentration in the smoke plume indicates that surface contamination radioactivity is released to a degree from burning crop land.

Activity concentration ratios of $^{95}\text{Zr-Nb}$ to ^7Be were also calculated for each filter and are shown in Table 1. The consistency or variation in this ratio might shed light on the high 4.6 smoke/background ratio calculated for $^{95}\text{Zr-Nb}$ on October 22. The $^{95}\text{Zr-Nb}/^7\text{Be}$ ratios in the smoke plume were 1.8 for both days. Similarly the background ratio was 1.7 on October 21. However, the background ratio of $^{95}\text{Zr-Nb}/^7\text{Be}$ was only 0.4 on October 22.

There is not a completely satisfying explanation to the low $^{95}\text{Zr-Nb}$ concentration on October 22. However, one possibility is that wind velocities were lower and consequently any $^{95}\text{Zr-Nb}$ on the ground would not have been resuspended as readily by winds.

CONCLUSIONS

Increased radioactivity in smoke from burning crop lands over background air concentrations suggests that surface contamination can be

TABLE 1. Normalized Airborne Activity Concentration Ratios.

Sampling Date	Smoke/Background		$^{95}\text{Zr-Nb}/^7\text{Be}$	
	^7Be	$^{95}\text{Zr-Nb}$	Background	Smoke
10-21-74	1.3	1.4	1.7	1.8
10-22-74	1.2	4.6	0.4	1.8

* PR3 Flowmeter System, Sci-Med, Inc., Minneapolis, Minnesota.

released by fire. One implication of this conclusion is that deposition of low level airborne activity on vegetation may minimize wind-caused resuspension but subsequent fires in that contaminated vegetation will release accumulated activity during a short time period. The

possibility exists that the downwind transport and potential inhalation hazard could be significantly increased by fires in severely contaminated vegetation. Further experiments are advised to confirm these initial measurements.

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FREQUENCY AND DIURNAL VARIATION OF DUST IN THE
CONTINENTAL UNITED STATES

M. M. Orgill and G. A. Sehmel

Frequency of dust periods indicated by visibility less than 11 km were determined by analyzing existing weather data for 343 stations throughout continental United States. Contours of annual dust frequency are shown. Diurnal variation of resuspended dust often shows an afternoon maximum for a majority of stations.

INTRODUCTION

Studies concerning regional wind resuspension and translocation of dust are important for air pollution surveys, the nuclear-power industry, and agriculture. Specifically, future nuclear power reactors, chemical fuel-reprocessing and fuel-enrichment plants may be located in a number of different climatic regions where wind resuspension of radioactive materials may be possible. These could be urban, forested, agricultural, desert and range-land sites. It is important to know from a planning and operational standpoint, the frequency, duration and intensity of wind resuspension periods for these new sites, as well as existing operational sites. Consequently, the purpose of this study is to evaluate and quantify wind resuspension and natural wind-caused airborne dust loadings throughout the contiguous United States.

Most studies of wind resuspension of dust have centered around the Great Plains¹ without much consideration given to other areas of the country. Other regions of this country are generally known to experience significant dust storms^{2,3} but there are no comprehensive reports concerning the frequency and spatial distributions of dust resuspension for the entire country.

Consequently, National Weather Service and military surface observations on dust were analyzed for annual, monthly and diurnal periods. Frequency of dusty periods was based on hourly-weather observations summarized from 343 weather observing

stations that recorded dust, blowing dust and sand when the visibility was less than 7 miles (11.3 km). The data showed that most regions of the United States are susceptible to airborne dust due to wind resuspension with some localized exceptions.

PROCEDURES

Visibility as a measure of atmospheric opacity indicates to an observer the quantity of pollution (in this case, dust) in the air. Prevailing visibility is the greatest visibility equalled or exceeded throughout half of the horizon circle.⁴ It is a subjective measurement which depends upon individual ability to both detect and recognize distant objects seen against the horizon sky by day or to detect distant lights at night. Prevailing visibility is taken routinely at National Weather Service observation stations and military installations that have observers on duty.

Charlson⁵ has shown that prevailing visibility and particulate concentration are related when the particle size distribution remains constant and the relative humidity is less than 70 percent, or deliquescent particles are not present in significant numbers.

A theoretical visibility concentration relationship¹ can be derived by assuming that 1) visibility extinction is caused only by scattering, 2) airborne particles are of uniform size, and 3) the scattering area coefficient is constant. In this case,

$$C = \frac{57.2}{V} \text{ (mg m}^{-3}\text{)} \quad (1)$$

where V is prevailing visibility in km. A nearly identical empirical visibility concentration relationship at 1.8 m above the surface was found by Chepil and Woodruff,¹

$$C = \frac{56.0}{V^{1.25}} \text{ (mg m}^{-3}\text{)}. \quad (2)$$

National Weather Service and military surface observations of dust, etc., are only reported, unfortunately, when the prevailing visibility is less than 11.3 km. Substitution of 11.3 km into equations (1) or (2) gives a dust concentration from 2.7 to 5.1 mg m⁻³. Therefore, dust frequency data based on prevailing visibility refer to dust storms when the dust concentration is 3 to 5 mg m⁻³ or greater. A very severe dust storm may produce dust concentrations over 1000 mg m⁻³.³

Hourly observations of prevailing visibility and occurrence of dust, etc., have been collected over several years and are stored on magnetic tapes at the National Climatic Center. These data have been summarized for individual weather stations where the data are tabulated in terms of percentage frequency of occurrence for various weather elements for annual, monthly, and three-hourly periods. The data have been summarized over a 15- to 30-year period between 1940 and 1970.

These summaries for 343 weather observation stations were obtained from the National Climatic Center. The data for frequency of occurrence of blowing dust, etc., for annual, monthly and three-hourly periods were then extracted, plotted, and analyzed.

RESULTS

Spatial Distribution of Dust

The annual frequency of occurrence of resuspended dust from major dust storms is shown for the country (excluding Alaska and Hawaii) in Figure 1. In this figure, contour lines were drawn to approximate the annual dust frequencies at each station. These contour lines range from N (none) to 3% dust/year. Contour lines were drawn subjectively, but

with the goal of determining dust frequency patterns on a regional basis. Suspended dust occurs relatively infrequently, and therefore the percentage frequency is not very high. For example, the maximum percentage frequency for the country is 3.1% at Lubbock, Texas. This amounts to a total of 5133 hr in a 23 year period, or 223 dust hr per year on the average.

The spatial distribution of dust frequency can be discussed by dividing the country into five broad regions; West Coast and Inland Valleys, Western Mountainous States, Great Plains, Central States and East Coast.

West Coast and Inland Valleys.

The coastal and mountainous regions have little or no dust except for the southern part of California. Dust occurs in the southern valleys, basins, and deserts of California.

Western Mountainous States. Dust frequency is about the same magnitude for the Central and Eastern Washington, western Nevada desert and western Utah desert. Dust frequency is greatest in the southern Arizona desert. The high desert plateau of eastern Oregon is relatively free of dust but dust occurrence increases further east in the Snake River Valley. Though the observations are limited, dust frequency is apparently very low in the high mountainous terrain of all western states.

Great Plains. The highest dust frequency in the country is along a north-south line through west Texas, Oklahoma, Kansas and eastern Colorado approximately 300 km from crest of the Rocky Mountains. This line of high dust frequency also extends through Central Nebraska, and eastern South and North Dakota, but with lower frequencies.

Central States. The northern central states of Wisconsin, Michigan, Illinois, Indiana, Ohio have relatively low amounts of suspended dust. However, the southern states including Kentucky and Tennessee show higher frequencies than the northern states, but not as high as the western mountainous states. Evidently part of suspended dust in the southern states is due to dust fronts

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FIGURE 1. Annual Percentage Frequency of Dusty Hours Based on Hourly Observations from 343 Weather Observation Stations that Recorded Dust, Blowing Dust, and Sand when Prevailing Visibility was Less than 7 Miles (11.3 km). Total period between 1940 and 1970, approximately.

moving east from the dustier areas of Texas and Oklahoma.

East Coast. Dust frequency is very low among the Appalachian Mountains and northward into the New England States. A slight maximum in dust frequency is found generally leeward of the Appalachian Mountains in the eastern parts of South and North Carolina and southern Georgia.

Seasonal Variation of Dust

In certain regions of the country the seasonal dust frequency is bimodal with a primary maximum between March and May and a secondary maximum in the fall or early winter months. The seasonal, monthly and diurnal frequency of dust is shown in Figure 2 for four regions that experience a seasonal bimodal distribution.

Generally, however, maximum dust frequency occurs between the early and late spring months for the country as a whole. This is particularly true for regions east of the Mississippi River since dust frequency is very sporadic and infrequent during the remainder of the year. The charts showing the month to month variation in dust frequency are too many to present in this report. However, one of the distinguishing features of the monthly charts is that their general dust frequency spatial patterns, west of the Mississippi River, are very similar in pattern to the annual chart (Figure 1), except the magnitude of dust frequency varies from month to month. Apparently, in the western states, the sources of resuspended dust are generally fixed to certain regions of available soil and do not vary much in location during the year.

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FIGURE 2. Seasonal, Monthly, and Diurnal Variation of Dust Occurrence at Four Locations that have a Seasonal Bimodal Distribution of Dust Frequency.

Diurnal Variations of Dust

Our analysis to date shows that hourly variations of dust may appear in four types of diurnal curves. These are 1) afternoon maximum, 2) random, 3) mixed, and 4) late afternoon and evening maximum. Examples of all four are presented in Figure 3. The afternoon maximum is the more common diurnal variation and occurs at many locations. Lubbock, Texas, as well as the stations in Figure 2 illustrate this diurnal variation.

The random type is illustrated by Lufkin, Texas where the hourly variation is devoid of any significant peaks in dust frequency. Here the dust frequency distribution is probably due to the random occurrence of frontal or storm activity.

Hanford, Washington illustrates the mixed type, i.e., the random occurrence of storm activity superimposed on the afternoon maximum. The late afternoon and evening maximum type is interesting since it appears to occur most markedly at Bakersfield, California during the

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FIGURE 3. Four Types of Diurnal Variation of Dust Frequency. Lubbock, Texas: Afternoon Maximum; Lufkin, Texas: Random; Hanford, Washington: Mixed; Bakersfield, California: Late Afternoon and Evening Maximum (Warm Season).

spring and summer. Apparently, during these warm months a strong sea-breeze front invades the Bakersfield area and produces late evening duststorms of varying degrees.

DISCUSSION

These tentative results show that many regions of the country, such as

urban, agricultural, desert, valley, plain and forested areas are subject to resuspended dust in varying degrees. A continuing appraisal of the regional occurrence of dust due to major dust storms will be detailed in a separate report in the near future.

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SOME INITIAL MEASUREMENTS OF DDT RESUSPENSION AND TRANSLOCATION FROM PACIFIC NORTHWEST FORESTS

M. M. Orgill, M. R. Petersen, and G. A. Sehmel

The spraying of Pacific Northwest forests with DDT to control a tussock moth infestation provided an opportunity for initial measurements of DDT translocation in a forest environment by resuspension and volatilization processes.

Airborne DDT was sampled by a Cessna 411 aircraft-mounted high-volume sampler on 5 days. The air samples for 4 days contained significant amounts of p,p'-DDT, and o,p'-DDT, and p,p'-DDE except for background and high altitude samples. Calculated resuspension rates for particulate DDT on three of the sampling days were 1.0×10^{-8} , 2.5×10^{-8} and 7.7×10^{-8} fraction resuspended per second.

INTRODUCTION

Measurements of particle resuspension rates from forested areas are relatively unknown yet such measurements are important to predict translocation and potential downwind health hazards for particulate matter or radioactive materials that may become airborne again.

The recent (June-July, 1974) spraying of Pacific Northwest forests with DDT [1,1,1-trichloro - 2,2-bis (p-chlorophenyl) ethane] to control a tussock moth infestation provided a unique opportunity to utilize DDT as a tracer for studying resuspension and translocation processes from the forest environment. By using a high-volume sampler in an aircraft, the

vertical distribution of DDT airborne concentration over a forested region was determined. This was used with winds aloft to estimate the horizontal flux of DDT and a resuspension rate.

The forest resuspension rate, estimated from data from three sampling flights, varied from 1×10^{-8} to 7.7×10^{-8} fraction resuspended per second. Valley slope winds, convection, and convective turbulence were the predominant mechanisms for resuspension and translocation of DDT during the sampling days. Additional sampling 86 days after the spraying program had ceased indicated that airborne DDT concentration levels were back to background levels.

DDT SPRAY PROGRAM

During the week of June 10-14, 1974, the U.S. Forest Service¹ initiated their tussock moth control program by aerial spraying of three different forested areas in the Colville Indian Reservation in North-Central Washington. Table 1 indicates the general location, elevation and acreage of the spray areas, and the amount of DDT sprayed on the trees.

Spray application was by one or two helicopters flying as low as permissible over the tree crowns. The application rate of DDT was 0.75 lb acre, mixed with 1 gal of auxiliary hydrocarbon solvent and No. 2 fuel oil.

The DDT was applied under very light wind conditions in the early morning hours to prevent excessive drift and was terminated before 0800, PDT. The weather conditions were warm

and dry, with surface winds never exceeding 5 m/sec; ideal for spraying operations.

SAMPLING METHOD AND ANALYSIS

DDT is frequently detected in air samples at background levels² (19 fg m^{-3} to 115 fg m^{-3}). Various methods have been used to collect airborne DDT, and for this study we used an adaptation of an air sampling technique used by Bidleman and Olney² to determine the concentration of PCB, DDT, and chlordane in background samples collected in the Sargasso Sea atmosphere.

Air sampling was accomplished with a ram-air high volume sampler mounted in a Cessna 411 aircraft. The sampler consisted of a glass fiber filter (Type A, 293 mm diameter) and a backup filter of cylindrical polyurethane foam (6 in. dia. by 6 in. long). Airflow through the filters was monitored by a PR-3 Flow Sensor* and varied between 2 to 3 m^3/min , depending upon air speed and altitude. Sampling durations varied between 20 and 60 minutes, depending upon altitude. Glass filters and foam plugs were changed after each sampling period.

The sampling durations were 20 to 60 min to assure that enough material would be collected for analysis. However, this sampling time limited the number of sampling altitudes to four and at most five during the day. Sampling altitudes and locations were

*Sci-Med, Inc., Engineering Dept., Minneapolis, Minnesota.

TABLE 1. DDT Spray Areas and Amounts of DDT.

Date	Location	Mean Elevation (ft msl)	Acreage	Amount of DDT (Kg)
11 June	5 miles ESE of Keller	3000	1500	510 (1125 lb)
12 June	Nicholls Mtn. and vicinity	3000	3000	1021 (2250 lb)
14 June	Whitestone Ridge	3000	2100	714 (1575 lb)

generally based on an idealized model of valley-slope wind system and predicted winds. Generally, two samples were taken at the lower altitudes (500 to 1000 ft above trees) over or near the DDT forest source, and two samples above the ridges downwind from the source.

Before sampling, the glass fiber filters and polyurethane foam plugs were cleaned to remove possible interfering organic compounds. The filters were fired overnight to 400°C and the foam plugs were extracted (Soxhlet) with benzene for 24 hr. The DDT samples were extracted (Soxhlet) from the glass fiber filters and foam plugs with hexane for 24 hr. The hexane extracts were concentrated to 0.5 ml with a Kuderman-Danish apparatus. The DDT analyses of the DDT extracts from the filters and foam plugs were analyzed on a gas chromatograph fitted with an electron capture detector. Chromatograms for the filters and plugs were compared to standards and pesticide background levels in order to quantify the DDT results. The results from the foam plugs showed the samples to be slightly contaminated with PCB (polychlorinated biphenyl), but this interference did not significantly affect the overall results.

SAMPLING WEATHER

The weather conditions during four of the sampling periods, usually 4 hr each, were generally hot (85°F to 100°F) and dry with surface winds seldom exceeding 5 m/sec. This unfortunately was not ideal for typical wind resuspension, due to wind shear stresses and mechanical turbulence. Nevertheless, the wind and thermal conditions were favorable for the development of local valley-slope winds, convection and convective turbulence.

Pilot balloon observations, taken hourly during sampling, showed the presence of valley winds except on June 13, when lower-level winds were from the northeast in the Columbia River Valley. A hodograph with trajectories and wind speeds of pilot balloons during one of the four sampling days is shown in Figure 1. The development of a valley wind (south wind) in the Sanpoil River Valley is shown between 0850 PST and 1210 PST.

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FIGURE 1. Hodograph for June 12, 1974 Showing Trajectories, Direction and Speed of Pilot Balloons. Development of valley wind is shown between 0850 PST and 1210 PST.

RESULTS

Airborne DDT was sampled on four consecutive days during the first spray operations. On two of the days, June 11 and 14, sampling commenced 2 to 6 hr after the spraying operation had ceased. Sampling on the other 2 days commenced 26-30 hr after the spray operations. All the sampling flights were conducted between 0830 PDT and 1300 PDT.

Fifteen samples were obtained during the four sampling flight periods. All of the samples contained varying amounts of p,p'-DDT, o,p'-DDT, and a degradation product, p,p'-DDE. The detailed results of the analyses have been reported by Orgill, Petersen and Sehmel.³ Table 2 shows the sum of all DDT components in the particulate (filter) and vapor (foam plug) phases grouped according to sampling days. The background samples, taken on June 11, were collected over a forested and farm area upwind and west of the Keller spray area.

TABLE 2. Collection Dates, Locations, Altitudes and Total Concentration of DDT During Airborne Sampling Over Three Freshly Sprayed Forested Areas in North-Central Washington.

SAMPLE	DATE	LOCATION	MEAN ALTITUDE ft/msl	TOTAL PARTICULATE DDT ng/m ³	TOTAL VAPOR DDT ng/m ³	TOTAL AIRBORNE DDT ng/m ³
B1	JUNE 11	EAST OF	3300	< 0.01	< 0.028	< 0.038
B2	JUNE 11	NESPELEM	6000	< 0.01	< 0.028	< 0.038
S1	JUNE 11	SE OF	5500	0.089	LOST	?
S2	JUNE 11	KELLER	5600	LOST	0.212	?
S3	JUNE 12	SE OF	3500	0.106	0.593	0.699
S4	JUNE 12	KELLER	4000	0.023	0.075	0.098
S5	JUNE 12		5300	0.061	0.221	0.282
S6	JUNE 12		6000	0.272	0.140	0.412
S7	JUNE 13	NICHOLLS	2700	0.808	0.828	1.636
S8	JUNE 13	MOUNTAIN	3600	1.352	0.258	1.610
S9	JUNE 13		5300	0.102	0.052	0.154
S10	JUNE 13		6300	0.084	0.005	0.089
S11	JUNE 14	WHITESTONE	2700	14.882	3.882	18.764
S12	JUNE 14	RIDGE	3300	2.754	3.189	5.943
S13	JUNE 14		4500	1.331	1.750*	3.081
S14	JUNE 14		5300	0.057	0.291	0.348
S15	JUNE 14		8100	~0	0.204	0.204

*SAMPLE LOST VALUE INTERPOLATED

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DDT FLUXES

The horizontal DDT total particulate and vapor flux (wind speed times concentration) were calculated for June 12, 13 and 14 using the concentration data from Table 2 and mean wind profiles obtained from the pilot balloon observations. Figures 2 and 3 show the DDT particulate and vapor flux on June 12, 1974. The increase of particulate and vapor concentration and fluxes with height this day was probably the result of vertical currents, due to slope winds and convection, transporting the DDT to the higher levels.

The concentration profiles and fluxes for the last 2 days were different from those of June 12. Apparently, the sampling location of the aircraft was not totally in the DDT plume due to wind direction changes. Consequently the DDT concentrations were lower in the higher altitudes than might have been expected.

RESUSPENSION RATES

The DDT particulate flux data was used in approximating the rate of DDT translocation from the spray areas. The rate of particulate DDT removal past a given vertical area normal to

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FIGURE 2. Horizontal DDT (Total) Particulate Flux for June 12, 1974 as Calculated from DDT Concentration Profile and Mean Wind Profile.

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FIGURE 3. Horizontal DDT (Total) Vapor Flux for June 12, 1974 as Calculated from DDT Concentration Profile and Mean Wind Profile.

wind direction would be equal to the average concentration multiplied by the mean wind velocity. The actual rate of removal (resuspension) for a given sampling period and spray area was expressed as:

$$R.R. = \frac{\left[\sum_{z \sim 3000 \text{ ft msl}}^{z \sim 8000 \text{ ft msl}} (C \cdot V_{\text{wind}}) L_s \cdot \Delta z \right]}{\text{[Total amount of DDT deposited]}}$$

$$\left(\frac{\text{gm sec}^{-1}}{\text{gm}} \right) = \frac{\text{Fraction resuspended}}{\text{sec}} \quad (1)$$

where

C = the total particulate DDT concentration in the interval Δz

V_{wind} = mean wind speed through the interval Δz

L_s = the airborne cross-wind sampling distance downwind from the DDT spray area

Δz = height interval during sampling.

The airborne sampling distances (L_s) were based on a best estimate of the cross-wind distance downwind from the spray area where sampling was done. Wind directions were constant enough with height that L_s was chosen to be constant with height except on June 12. On this day L_s varied with height having a longer distance at the higher altitudes. The sampling distances varied from 6 to 19 miles and may be underestimated due in part to the light and variable winds present each day.

The particulate DDT resuspension rates for three of the sampling periods, June 12, 13, and 14, were calculated by equation (1). The first sampling day, June 11, was not included because the DDT concentration data were not complete enough. Computed resuspension factors were 1×10^{-8} , 2.5×10^{-8} and 7.7×10^{-8} fraction resuspended per second. The highest rate was computed on June 14, a period when sampling was initiated within 2 hr after spray application. The lower two rates were computed for

June 12 and 13, when sampling was initiated 26 to 30 hr after spray application.

DDT RESUSPENSION AFTER EIGHT-SIX DAYS

The U.S. Forest Service completed their DDT spray program in the Colville Unit on July 2, 1974. Our initial success in sampling of DDT in June and obtaining an estimate for the resuspension rate made it desirable to do additional sampling to estimate how the DDT airborne concentration and resuspension rate may vary with time. Unfortunately, the Battelle aircraft was not available for additional DDT sampling until late September. On September 26, an ideal resuspension day, an attempt was made to sample DDT over the Colville Control Unit but strong surface winds and turbulence in the area prohibited the aircraft from flying the desired sampling altitudes. On September 27, 86 days after the last spray operation, a successful sampling flight was conducted over the eastern sector of the Colville Indian Reservation.

Four samples, each of about 60 min duration, were obtained for four different altitudes--3300 ft msl, 3800 ft msl, 4800 ft msl and 6300 ft msl. The sampling flight path, generally, over and downwind from the sprayed timberland was based on the existing winds and flight turbulence levels. A weather observer on the ground radioed current pilot-balloon wind information to the aircraft so these data could be used to plan the sampling flight pattern.

Winds at ridge top levels were generally southwest at about 10 m/sec; at higher altitudes the winds were more westerly. In the valleys, the wind had a more southerly direction and was generally below 6 m/s. However, surface winds were sufficiently strong to the south of the Colville Indian Reservation that resuspended dust could be observed moving toward the northeast and the sampling region.

The results from the four sample filters and foam plugs showed that all DDT amounts were below or at the detection limits of 0.56 ng from filters and 1.5 ng from foam plugs. The maximum concentration that could be

calculated for total DDT was 0.02 ng/m³. Apparently, no airborne DDT, above background levels, was being translocated from the forested areas on this particular day.

DISCUSSION AND CONCLUSIONS

DDT is a somewhat complex atmospheric tracer to use in studying resuspension and translocation over forests. Nevertheless, the U.S. Forest Service's tussock moth control program has resulted in a unique opportunity to measure the distribution of DDT with height, as well as a first estimate of the resuspension and translocation from a forest area.

The calculated forest DDT particulate resuspension rates of 10⁻⁸ (and possibly 10⁻⁷) fraction resuspended per second are in the upper-range of wind caused resuspension rates (10⁻¹⁰ to 10⁻⁸) reported in field measurements at the Hanford AEC Reservation (a semi-arid region in Washington).³ It is not known at this point what microphysical resuspension mechanisms were in operation during the DDT sam-

pling periods but convection and convective turbulence were more predominant than mechanical turbulence, due to natural air movement. Since DDT has a relatively high vapor pressure, volatilization may have contributed indirectly to the resuspension by adsorption on airborne particulate matter.

All these factors are difficult to assess quantitatively at this point but do suggest qualitatively that the calculated DDT resuspension rates of 10⁻⁸ may be on the high side due to the complex nature of DDT. Additional measurements of DDT or some other tracer over a forest area are needed in order to verify these resuspension rates as well as to provide information on the effects of weathering and degradation on resuspension rates.

ACKNOWLEDGEMENT

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AN ASSESSMENT OF THE LONG-TERM EXPOSURE
DUE TO RESUSPENSION--PART II

T. W. Horst and C. E. Elderkin

An earlier model by the authors for the ratio between the exposure due to resuspension and the exposure due to direct atmospheric transport from a source is extended to account for radioactive decay of the hazardous material, and for a lower limit to the resuspension factor. These modifications are shown to affect the evaluation of the hazard of environmental release of plutonium only for a source which emits continuously for thousands of years.

INTRODUCTION

At present a special need exists to evaluate potential environmental hazards of plutonium. Resuspension into the air of hazardous material previously deposited on a natural surface is one potentially important pathway for plutonium to enter the human lung. A simple model, which applies most accurately in a situation which is horizontally homogeneous, was presented by Horst, et al.¹ to address this problem. This report modifies those results by explicitly accounting for radioactive decay and by limiting the loss of deposited material due to fixation by the soil.

ANALYSIS

Most available field data on resuspension is available only in terms of the resuspension factor,

$$K(m^{-1}) =$$

$$\frac{\text{Airborne concentration (units/m}^3\text{)}}{\text{Surface concentration (units/m}^2\text{)}} \quad (1)$$

Horst, et al.,¹ showed how this factor and its variation with time could be related to parameters which have a somewhat more fundamental physical basis, such as the resuspension rate, deposition velocity and soil fixation rate. For the purpose of the LMFBR impact statement,² the resuspension

factor was modeled as decaying exponentially with a half life of 50 days from an initial value, applying immediately after deposition, of $10^{-5} m^{-1}$. This occurs until a minimum value of $10^{-9} m^{-1}$ is reached after 664 days, when the resuspension factor has been assumed to remain constant. Thus

$$K(t) = \begin{cases} K_0 e^{-\beta t} & , t < t_a \\ K_m = K_0 e^{-\beta t_a} & , t > t_a \end{cases} \quad (2)$$

Consider first the passage of a puff of radioactive material with air concentration x_0 in a time T . If v_d is the deposition velocity, this cloud deposits a concentration $v_d x_0 T$ on the ground. The resulting air concentration due to resuspension, normalized by the initial air concentration, is thus

$$x_r/x_0 = \begin{cases} v_d T K_0 e^{-(\beta + \lambda)\tau} & , t < t_a \\ v_d T K_m e^{-\lambda t} & , t > t_a \end{cases} \quad (3)$$

where λ is the radioactive decay rate of the material. The subsequent exposure ratio is calculated as

$$E_r/E_0 = \frac{1}{x_0 T} \int_0^t x_r(\tau) d\tau \quad (4)$$

Using (3),

$$E_r/E_o = \begin{cases} \frac{v_d K_o}{\beta + \lambda} \left[1 - e^{-(\beta + \lambda)t} \right] & , t < t_a \\ \frac{v_d K_o}{\beta + \lambda} \left[1 - e^{-(\beta + \lambda)t_a} \right] + \frac{v_d K_m}{\lambda} \left[e^{-\lambda t_a} - e^{-\lambda t} \right] & , t > t_a \end{cases} \quad (5)$$

If we let the time of exposure become large compared to the radioactive half-life of the material and recall that $e^{-\beta t_a} = 10^{-4}$,

$$E_r/E_o = \frac{v_d K_o}{\beta + \lambda} + \frac{v_d K_m}{\lambda} e^{-\lambda t_a}, \quad t \rightarrow \infty. \quad (6)$$

Consider also, now, a continuous plume of material with concentration x_o which is depositing on the ground at a constant rate $v_d x_o$. The surface concentration resulting from deposition during the time increment $d\tau$ is then $v_d x_o d\tau$ and the air concentration due to resuspension at a time t after the initiation of the continuous plume is

$$x_r(t) = \int_0^t v_d x_o K(t - \tau) e^{-\lambda(t - \tau)} d\tau \quad (7)$$

Changing the variable of integration to the elapsed time since deposition, $(t - \tau) = s$, this becomes

$$x_r/x_o = \int_0^t v_d K(s) e^{-\lambda s} ds. \quad (8)$$

Substituting (2) into this equation gives results identical to (5), i.e.,

$$(x_r/x_o)_{\text{plume}} = (E_r/E_o)_{\text{puff}} \quad (9)$$

The exposure to the resuspended material during a period T can further be calculated and compared to the direct exposure $x_o T$ during the same period,

$$E_r/E_o = \frac{1}{T} \int_{t_1}^{t_2} (x_r/x_o) d\tau, \quad (t_2 - t_1) = T. \quad (10)$$

Three different cases must be considered and the results are shown below, using when possible the fact that $e^{-\beta t_a} = 10^{-4}$.

Case 1: $0 \leq t_1 \leq t_2 \leq t_a$

$$E_r/E_o = \frac{v_d K_o}{\beta + \lambda} \left\{ 1 - \frac{1}{(\beta + \lambda)T} \left[e^{-(\beta + \lambda)t_1} - e^{-(\beta + \lambda)t_2} \right] \right\} \quad (11)$$

$$E_r/E_o = \frac{v_d K_o}{\beta + \lambda} \left\{ 1 - \frac{1}{(\beta + \lambda)t_a} \right\}; \quad t_1 = 0, \quad t_2 = T = t_a \quad (12)$$

Case 2: $0 \leq t_1 \leq t_a \leq t_2$

$$E_r/E_o = \frac{v_d K_o}{\beta + \lambda} \left[1 - \frac{1}{(\beta + \lambda)T} e^{-(\beta + \lambda)t_1} \right] + \frac{v_d K_o}{\lambda T} \left[(t_2 - t_a) e^{-\lambda t_a} - \frac{1}{\lambda} (e^{-\lambda t_a} - e^{-\lambda t_2}) \right] \quad (13)$$

Case 3: $t_a \leq t_1 \leq t_2$

$$E_r/E_o = \frac{v_d K_o}{\beta + \lambda} + \frac{v_d K_m}{\lambda} e^{-\lambda t_a} - \frac{v_d K_m}{\lambda^2 T} (e^{-\lambda t_1} - e^{-\lambda t_2}) \quad (14)$$

DISCUSSION

Some special cases of solutions (11) - (14) are worth considering. Typical results have been listed in Table 1 for a selection of radioactive isotopes, using the resuspension parameters given prior to (2) and a deposition velocity of 1 cm/sec. Note from the equations that these results may be changed linearly for a different v_d (and also for a different K_o if K_m/K_o remains at 10^{-4}).

TABLE 1. Exposure Ratios for Selected Isotopes and Periods of Plant Operation.

Isotope	Radioactive Half-Life (yr)	Exposure Ratio, E_r/E_o , During:		
		Initial 1.8 Yrs of Operation (12)	Next 70 Yrs of Operation (14)	Infinite Operating Period (6)
Cm^{242}	0.44	0.44	0.48	0.48
Cm^{244}	19	0.55	0.63	0.63
Pu^{238}	92	0.55	0.63	0.66
Am^{241}	475	0.55	0.63	0.84
Pu^{240}	6,580	0.55	0.63	3.62
Pu^{239}	24,100	0.55	0.63	11.59

Eq. (12) represents the exposure ratio for the first 664 days of plant operation. Resuspension contributes an additional 50% to the exposure during that period. Due to the rapid (50 day half-life) weathering or fixation of the deposited material which makes it unavailable for resuspension, the exposure ratio increases only slightly during the next 70 years of operation. However, due to the finite limit (a factor of 10^{-4}) placed on the loss due to weathering, there is a gradual increase in the amount of material available for resuspension, and a consequent increase in x_r/x_o . This reaches a limit only at times which are large compared to both the weathering and radioactive half-lives. After that time x_r/x_o is equal to a constant value, (6). Thus for any exposure period which is large in the sense indicated above, or which begins at this large time

after the plant began operation, E_r/E_o for the plume is equal to (6), E_r/E_o for the puff. This increase is seen in the last column of Table 1. It is significant only for those isotopes which have extremely long half-lives and also requires the plant to operate for that period of time.

Neglecting radioactive decay and a lower limit for K, Horst, et al.¹ found that

$$E_r/E_o = v_d K_o / \beta. \quad (15)$$

Under those conditions the previous equations analytically approach this limit. More importantly, (15) gives $E_r/E_o = 0.62$ with the current values of the parameters. Table 1 shows that neglecting the considerations stated above was justified so long as the LMFBR system does not operate indefinitely.

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THE RESUSPENSION RATIO FOR A GAUSSIAN PLUME

T. W. Horst

The surface flux model is used to calculate the resuspension ratio (the air concentration due to resuspension divided by that due to direct atmospheric transport from the source) for the Gaussian diffusion plume from a single source. Assuming 1) a steady state situation during constant atmospheric conditions, and 2) that all deposited material is available for resuspension, the total air concentration is found to be the same as would be calculated by simply ignoring both deposition and resuspension.

The contribution of resuspended material to an airborne hazard relative to that of material arriving directly from the source may be formalized by means of a resuspension ratio

$$R = x_r/x_d, \quad (1)$$

where x_r and x_d are the air concentrations due to resuspension and due to direct atmospheric transport from the original source. Horst, et al.^{1,2} have discussed this quantity in the context of the nation-wide impact of the liquid-metal fast-breeder-reactor (LMFBR) program, assuming horizontal homogeneity on a large enough scale to justify the use of a resuspension factor. Here this constraint is eliminated, albeit in favor of new assumptions, and results are presented which include the horizontal variations associated with a single realization of a Gaussian plume.

In order to account for the contributions of deposition and resuspension to the total air concentration, the surface flux model^{3,4} is utilized. Consider the cross-wind-integrated air concentration x_0 of a nondepositing material, released at a rate Q from the point $(x = 0, y = 0, z = h)$ into a mean wind of strength u , which is blowing in the direction of positive x . Defining $\psi = ux/Q$,

$$\psi(x, z) = \psi_0(x, z, h) + \int_0^x S(\xi) \psi_0(x - \xi, z, h = 0) d\xi \quad (2)$$

where $S(\xi)$ is the net flux of material into the air from the ground, also normalized by the source strength Q . For the calculation of the direct air concentration ψ_d , S need only account for deposition,

$$S_d(x) = - \frac{v_d}{u} \psi_d(x, z_d) \quad (3)$$

where v_d is the deposition velocity and z_d is the reference height for deposition. The resuspended air concentration ψ_r is found from the difference between ψ_d and the total air concentration ψ_T ,

$$\psi_r = \psi_T - \psi_d. \quad (4)$$

The calculation of ψ_T requires S to account for both resuspension and deposition,

$$S_T(x) = \frac{\Lambda G}{Q} (x) - \frac{v_d}{u} \psi_T(x, z_d) \quad (5)$$

where G is the ground concentration of material and Λ is the resuspension rate. G/Q finally, can be calculated from¹

$$\frac{\partial}{\partial t} \frac{G}{Q} = \frac{v_d}{u} \psi_T(z_d) - (\Lambda + \alpha) \frac{G}{Q}. \quad (6)$$

Here α represents the rate at which the material on the surface weathers and becomes permanently fixed to the soil, unavailable for resuspension.

Since the actual values of Λ and α which pertain to a given situation are not well known,¹ a solution which does not depend on them may be quite valuable. We can first of all maximize the airborne hazard by neglecting the loss of resuspendable material due to weathering. We can further restrict our investigation to the steady state situation when G has increased to the point where the resuspension and deposition fluxes are equal. Thus

$$\frac{\partial G}{\partial t} = -S_T = 0 \quad (7)$$

and $\psi_T = \psi_0$. Hence the maximum, steady state air concentration is that which can be calculated by neglecting deposition and resuspension altogether! Further, the resuspension ratio is a function only of v_d/u and not α , Λ or G . Combining (1) - (4) and (7),

$$R(x) + 1 = \psi_0(x)/[\psi_0(x) - (v_d/u)]$$

$$\int_0^x \psi_d(\xi, z_d) \psi_0(x - \xi) d\xi]. \quad (8)$$

The right-hand-side of this equation has already been calculated by Horst⁴ for several combinations of atmospheric stability, v_d/u , and release height. The resulting values for the resuspension ratio are shown in Figure 1 for a selection of these parameters. The curve for Pasquill stability category D, $v_d/u = 10^{-2}$ and $h = 10$ m may be considered to be a reference curve with which to gauge the variation of R with release height (2 m and 100 m), stability (Pasquill A and F), and v_d/u (10^{-3}). Amato⁵ has calculated the values of similar steady state curves, including also the dependence on weathering, but limited to surface releases and downwind distances less than 700 m.

The large values for the resuspension ratio at large distances downwind of the source might be a cause

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FIGURE 1. Resuspension Ratio, X_r/X_d , Versus Downwind Distance as a Function of Stability, h , and v_d/u .

for concern, except for the fact that they are due in part to a rapid decrease with distance of the direct air concentration χ_0 . In fact, as stated above, the total air concentration is no greater than would be found by simply neglecting both deposition and resuspension. This could be a very valuable result except for two points. First, these results ignore the contribution of precipitation scavenging in creating a ground deposit as a source for subsequent resuspension. Secondly, they are based on a steady state under the influence of a single atmospheric stability. Since actual resuspension rates are commonly found to be on the order of 10^{-7}sec^{-1} or less, steady state is approached exponentially with a time constant greater than several weeks. The atmospheric stability, on the other hand, will normally follow a diurnal cycle and the mean wind direction will shift on a similar time scale. Thus the steady state solution will reflect an amalgam of stabilities and wind directions rather than a single stability and direction as presented here.

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RESUSPENSION FROM A UNIFORM AREA SOURCE

T. W. Horst

The air concentrations resulting from resuspension from a horizontally uniform area source are calculated as a function of atmospheric stability, deposition velocity, and distance from the upwind edge of the source. The resulting horizontal flux of material and the implied resuspension factor are discussed, as well as limiting values for the air concentration, horizontal flux and resuspension factor at distances large enough so that resuspension is balanced by deposition.

The air concentration of material resuspended from an upwind area source is an extremely complicated function of the source geometry, source history and current meteorology. In order to begin an investigation of the atmospheric diffusion and deposition aspects of this problem, a simple geometry was hypothesized: a uniform field of resuspendable material of infinite extent in the cross-wind direction. A ground concentration G was assumed to resuspend at a constant rate Λ and re-deposit with a deposition velocity v_d from the resulting air concentration χ . The relevant coordinates are the height z above the ground and the distance x from the upwind edge of the uniform field. In applying the results below to an actual problem the assumption of an infinite extent in the cross-wind direction is not as limiting as might be supposed, since diffusion plumes are actually quite narrow in the cross-wind direction and only sources within a narrow cone in the upwind direction contribute substantially to the air concentration at a given point.

Air Concentrations

Using the surface flux model,¹ the air concentration is

$$\chi(x, z) = \int_0^x [\Lambda G - v_d \chi(\xi, z_d)] D(x - \xi, z) d\xi. \quad (1)$$

Here z_d is the reference height for deposition and $D(x, z)$ is the mathematical description of the Gaussian plume from a ground level, cross-wind-infinite line source of nondeposition material,

$$D(x, z) = \frac{2}{\sqrt{2\pi} \sigma_z u} \exp\left(\frac{-z^2}{2\sigma_z^2}\right), \quad (2)$$

where u is the mean wind speed and the dispersion parameter $\sigma_z(x)$ is calculated from the formulas of Briggs.²

Figure 1 shows the resulting air concentrations at z_d , as a function of x , for the special case of no deposition and for stable, neutral and unstable atmospheric conditions (Pasquill categories F, D, A). χ has been appropriately normalized with the mean wind speed, resuspension rate, and ground concentration. As would be expected, χ increases less rapidly with an increasingly unstable atmosphere due to the increasing effectiveness of the eddies in carrying the material away from the surface. The curve for Pasquill A ends at a distance of 10 km from the upwind edge due to computational difficulties with evaluating (1).

The upper set of curves in Figures 2 and 3 show the reduction in χ resulting from dry deposition for Pasquill F and D for three values of v_d/u . The reduction for Pasquill A

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FIGURE 1. Normalized Air Concentration Versus Downwind Distance for a Range of Stabilities and No Deposition.

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FIGURE 2. Normalized Air Concentration and Horizontal Flux Versus Downwind Distance for Pasquill F Stability and Several v_d/u .

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FIGURE 3. Normalized Air Concentration and Horizontal Flux Versus Downwind Distance for Pasquill D Stability and Several v_d/u .

is considerably less: only 20% at 10 km for the moderately high v_d/u of 10^{-2} . It can be inferred from (1) that χ increases until the concentration at the surface reaches a limiting value,

$$\chi_L(z_d) \equiv \lim_{x \rightarrow \infty} \chi(x, z_d) = \frac{\Lambda G}{v_d}. \quad (3)$$

Beyond this point resuspension is exactly balanced by deposition and there is no net transfer of material into the air from the ground. This limiting process can be seen in the curves at the upper right of Figures 2 and 3. Note that these limiting values are approached only for area sources with extremely large dimensions. Thus these limits will actually seldom be realized and are most useful as upper limits of the effects to be expected.

An empirical expression can be proposed for the reduction in air concentration at a given x due to deposition,

$$\chi = \chi(v_d = 0) / \left[1 + \frac{v_d \chi(v_d = 0)}{\Lambda G} \right]. \quad (4)$$

This formula is based upon the known asymptotic limits for large and small values of v_d/u and fits the air concentration curves of Figures 2 and 3 quite well.

The Horizontal Flux

When the resuspension is balanced by deposition the horizontal flux of material in the downwind direction,

$$F_H(x) = \int_0^\infty u\chi(x, z) dz = \int_0^x [\Lambda G - v_d \chi(\xi, z_d)] d\xi, \quad (5)$$

also reaches a constant value. F_H , normalized with ΛG , is shown in the lower set of curves in Figures 2 and 3, referenced to the right hand ordinate. Note that it approaches an asymptotic value for Pasquill F, but not for Pasquill D. This is associated with the fact that $D(x)$ becomes constant at large x for Pasquill F but continues to decrease indefinitely with increasing x for Pasquill D. The former condition on $D(x)$ is necessary for χ to truly reach its limiting value χ_L , for the net transfer from the ground to the air to be shut off,

and hence for the horizontal flux to become constant. When $D(x)$ does reach a limit (due to σ_z becoming constant),

$$\lim_{x \rightarrow \infty} D(x, z) \equiv D_L(z), \quad (6)$$

the constant upper value for F_H can be predicted from (1), (3) and (5),

$$F_L \equiv \lim_{x \rightarrow \infty} F_H = \Lambda G / v_d D_L(z_d). \quad (7)$$

This is confirmed by the curves of Figure 2.

The Resuspension Factor

In order to describe resuspension, much use has been made in the past of

the resuspension factor $K \equiv \chi/G$. If the ordinate $u\chi/\Lambda G$ is interpreted as K measured in units of Λ/u , some valuable insights can be gained. For a given value of v_d/u the resuspension factor is directly proportional to Λ/u . K is an increasing function of distance, changing rapidly close to the upwind edge of the contaminated area and then at a lesser rate until the limiting value Λ/v_d is reached for very large sources. Depending on the application, this change with x at intermediate distances may be slow enough for K to be usefully constant. Some effects of a nonuniform G can also be inferred: a G which increases in the downwind direction will counteract the rise of K with distance shown here, and a G which decreases with x will enhance the rise of K .

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IMPACT AND SPECIAL STUDIES

(Papers in this section are not appropriate to categories of the previous sections.)

ENERGY CONVERSION

The expansion of conventional energy conversion facilities and the development of alternate methods to meet the energy requirements of the nation brings a need for increased understanding of environmental interrelationships. Traditionally, possible adverse effects of energy conversion facilities on the environment have been of paramount importance. That viewpoint is reflected in the first two papers in this section, which characterize the condensate plumes from cooling towers used to dissipate the waste heat of existing power plants. The third paper propounds a concept by which such waste heat can enrich energy resources, and cites deficient knowledge of atmospheric behavior as a problem in evaluating the concept. The three succeeding papers describe remote, ground-based sensing of radiation phenomena as related to atmospheric turbidity from natural and anthropogenic sources. Sensing techniques have the potential to characterize the aerosol, and for sensitive determination of the magnitude and quality of insolation. These studies, while assessing the environmental impact of current energy conversion facilities, will also help evaluate and optimize solar energy conversion methods.

BIOMETEOROLOGY

The distribution of rainfall over the diverse regions of the Arid Land Ecology (ALE) Reserve is an important factor in understanding differences in their respective plant and animal communities. The paper proposes a simple model to extend rainfall distributions observed during the past six years over a greater period.

- **COOLING TOWER AND COOLING POND ATMOSPHERIC IMPACT**
- **ATMOSPHERIC AEROSOLS AND TRACE GASES**
- **ECOLOGICAL MICROMETEOROLOGY AND CLIMATOLOGY OF ALE RESERVE**
- **BATTELLE MEMORIAL INSTITUTE PHYSICAL SCIENCES PROGRAM**
- **AEC/RL SPECIAL STUDIES PROGRAM**

COMPARISON OF DROPLET SIZE DISTRIBUTIONS IN
A CUMULUS CLOUD AND A COOLING TOWER PLUME

A. J. Alkezweeny and M. A. Wolf

An airborne droplet size spectrometer was used to measure droplet distributions in a natural cumulus cloud and in the plume of a mechanical-draft cooling tower. Observed mass-median diameters of both were in the range 10-15 μm and an obvious increase with altitude was noted in the cloud. Liquid water content ranged between 0.0065 and 0.0303 g m^{-3} in the plume and 0.0680 and 0.2472 g m^{-3} in the cloud, where the highest value occurred midway between the cloud base and its top. The analysis suggested that drift--droplets mechanically created within the cooling tower--constituted an appreciable portion of the liquid water observed at the large droplet end of the spectrum in the plume. Additional effort will be made in the future to separately delineate the drift and condensate spectra.

The intensified interest during the past several years about the effects of cooling towers on the environment was demonstrated by the 3-day symposium "Cooling Tower Environment-1974" dedicated to that subject.* The symposium participants agreed that experimental data generally are inadequate for evaluation of cooling tower effects. In particular, a need was recognized for data on plume characteristics, with which models for plume rise and persistence might be evaluated and improved. In answer to this need, a field program will be conducted at the Rancho Seco Nuclear Generating Station near Sacramento, California, early in 1975

to provide these necessary cooling tower plume data. A facet of that program is measurement of droplet spectra in the cooling tower plume. This report discusses preliminary results using the measurement system which will be utilized.

The system consists of the Axially Scattering Spectrometer Probe, the Particle Data System* and, in the preliminary experiments, a high speed printer. The probe was mounted on a Cessna 411 aircraft, as shown in Figure 1, with its isokinetic opening of approximately 4.0 cm diameter located about 60 cm from the fuselage. A beam of light from a He-Ne laser on one side of the opening illuminates a centrally-located area of 0.86 mm^2

* Proceedings of the symposium held at College Park, Maryland, March 4-6, 1974, is available as "Cooling Tower Environment-1974," AEC CONF 740302 from the U.S. Atomic Energy Commission, Washington, D.C.

* The Axially Scattering Spectrometer Probe and the Particle Data System were produced by Particle Measuring Systems, Inc., Boulder, Colorado.

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FIGURE 1. Axially Scattering Spectrometer Probe Mounted on Cessna 411.

within the opening. A silicon photodiode detector on the opposite side of the opening senses the forward-scattered light from droplets passing through the sampling area. A pulse height detector examines the output signal, and the droplet size is recorded in the system memory which has 15 equal size channels for each of the 4 size ranges available: 0.5-7.5, 1.0-15.0, 2.0-30 and 3.0-45 μm .

Although the system can obtain droplet counts for a framing period as short as 0.25 sec, it was necessary to operate during the preliminary experiments with a framing period of 4 sec, the shortest permitted when using the printer. An incremental magnetic tape recorder which subsequently replaced the printer per-

mits operation of the droplet spectrometer at its maximum rate. This was an important modification to the system for two reasons. First, it provides resolution of droplet distribution over relatively short distances and identifies the time duration of the traverse, both of which are essential for effective plume sampling. Second, it eliminates the present droplet count uncertainty. Due to the storage capacity of 9999 in each channel, it is necessary that the framing rate be sufficiently high to avoid exceeding that value within a frame. During a framing period of 4 sec, it is possible to sample greater than 10^4 droplets and under such circumstances, the coefficient cannot be determined.

Preliminary experiments were made during the spring of 1974. Penetrations of a cumulus cloud were made near Pasco, Washington, on April 25. Cloud base and top were at about 1300 and 2600 m, msl, respectively. The freezing level was at 2290 m. Penetrations were made over a period of approximately 40 min during which no obvious trend in conditions was recognized. Cooling tower measurements were made on May 16 at the Centralia Power Plant in western Washington. Plume penetrations extended over approximately an hour, during which the sky was overcast above flight altitude. There were occasional light showers. At the sampling heights of 200-245 m, msl, (about 100-145 m above the cooling tower exit), the ambient air was moderately turbulent and the aircraft temperature sounding showed an unstable atmosphere below 325 m, msl. Winds were light and variable, and the plumes were visible only immediately above the towers.

The results of the cumulus cloud sampling are shown by the curves and table of Figure 2. It appears that the mass median diameters increase with increased altitude throughout the sampling range, although the increases are greatest in the lower half of the cloud. These observations

are consistent with previous investigations.^{1,2} Mass median diameters increased from 10.4 μm at the lowest penetration to 14.7 μm at the highest penetration. All measurements were made using the 2.0-30 μm range of the spectrometer, and it can be inferred from the curves that larger droplet diameters were present but were not sensed. Had those droplets been included, higher liquid water content (LWC) would have been found at the greater altitudes, and it is possible that the maximum value would have appeared above 1980 m. The distributions, as observed, indicate condensation and droplet growth in the lower half of the cloud with continued growth but dilution by entrainment in the upper half.

A different distribution of droplets in the cooling tower plume is shown with the curve defined by dots in Figure 2, which is the mean of 4 plume penetrations between 200 and 245 m, msl (approximately 100-145 m above the tower exit). While the mass median diameter of 12.7 μm is comparable to values in the lower half of the cumulus cloud, the LWC is nearly an order of magnitude less and the proportion of droplet sizes above 20 μm is more than double the proportion found in the cumulus cloud.

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FIGURE 2. Cloud Droplet Liquid Water Distribution.

Figure 3 presents the droplet distributions for the four individual penetrations of the cooling tower plume. Mass median diameters range from 12.0 to 13.2 μm , with the lower altitude penetrations showing somewhat larger values. This inverse relationship relative to the cumulus cloud penetrations is more pronounced at larger droplet diameters, where the percentages of droplets greater than 20 μm are 31, 24, 20, and 18 at 200, 215, 225, and 245 m, respectively. Thus, unlike the cumulus cloud for which larger droplets contained increasing percentages of the total LWC as the altitude increased, the larger droplet component of the plume spectrum decreases with altitude. The total LWC appears to increase with altitude over the limited depth examined, but this is uncertain due to the variability of the cooling tower plume and the difficulty in assigning a valid traverse time to the narrow plume with a framing period of 4 sec. A traverse time of 3 sec was assumed in the calculation of LWC. Again, the largest droplets in the

plume were not sensed by the spectrometer operated at the 2.0-30.0 μm range, and the calculated LWC values are undoubtedly too low.

It appears, in light of the inverse relationships of the large droplet component to altitude, that different mechanisms operate for the cumulus cloud and the cooling plume. Whereas the increase of the large droplet component with altitude in the cumulus cloud indicates continued droplet growth, the decrease of that component with height in the plume suggests that the large droplet component consisted largely of drift--droplets mechanically created within the cooling tower. This implies that definition of the condensate plume also requires definition of the drift component in the plume. It may be possible to accomplish the latter with the spectrometer during periods of low humidity when the condensate component is absent. Future effort will be directed toward resolution of this problem.

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FIGURE 3. Cooling Tower Plume Droplet Distribution.

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A METHOD OF PLUME PHOTOGRAPHY

O. B. Abbey, D. W. Glover,
and J. M. Thorp

Photography as an aid to plume analysis has been utilized for some time in various ways. This report describes a transit-camera system which permits photography of the plume as well as precise angular measurements. Instantaneous or long exposure "integrated" plume photographs are possible.

Description of long time exposure photography of plumes is well covered by Culkowski;¹ and air quality researchers of the TVA describe a photo and transit system for a press type camera.²

Figure 1 illustrates a modified transit which has been incorporated for our studies, and permits mounting a 16 mm movie camera, a 35 mm single lens reflex camera or a 4 x 5 press type camera above the transit barrel. This arrangement permits sighting-in on reference objects and/or boundaries of the plume to an accuracy of one tenth of one degree or less, and photographing the same with very small errors of parallax in the vertical, and none in the horizontal.

The mounted camera may cause some deflection of the compass needle; however, the camera platform is made of nonmagnetic material and the transit is oriented before the camera is attached. After orientation, the mag-

netic compass is not used in any calculations and as long as the assembly is not moved, magnetic directions are not relevant.

The camera platform is designed to present a perfectly horizontal platform in the tilting (side to side) place whenever the transit is properly leveled. Platform adjustment to horizontality in the tipping (fore and aft) plane is made through two leveling screws.

Long time exposures are possible through the use of neutral density filters which can be used singly, in tandem, or in combination with colored Wratten type filters. Neutral density filters are available that block as much as 99% of the light. They do not alter color rendition, however photographic films all have a trait called "reciprocity failure" when they are exposed for time periods much greater than that for which they

FIGURE 1. Nonmagnetic Camera Platform, and 35 mm Camera Mounted on Transit.

are designed. As a result of reciprocity failure, the rated ASA speed of a photographic film emulsion is reduced when long exposures, say greater than 3 min, are used. With black and white films, the reciprocity failure requires a longer exposure than that indicated by light meter measurement; for color emulsions a color shift also occurs which will be difficult or impossible to correct for. Film manufacturers should be able to provide data regarding reciprocity failure for their individual films.

In field use, the transit mounted 35 mm or 4 x 5 camera will produce instantaneous and time exposed, "integrated," photographs of the plume. On a separate tripod, a horizontally leveled, but not precisely oriented 16 mm movie camera will be taking time lapse movies of the same plume at approximately one to two frames per min. The combination of photographs will provide a useful base for dimensional and historical analysis of the plume.

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A SUGGESTED CONCEPT FOR DYNAMIC WEATHER MODIFICATION UTILIZING LARGE ATMOSPHERIC HEAT RELEASES TO STIMULATE CUMULONIMBUS CLOUDS

W. G. N. Slinn

It is suggested that research be conducted to determine if large heat releases could be utilized to influence the location of cumulonimbus clouds. If this is possible, the additional rainfall could be used for irrigation, cooling, domestic purposes and hydroelectric power generation. Some details of a specific scheme are sketched.

INTRODUCTION

During these days of increasing demands for energy and decreasing availability of it, it is of interest to inquire about alternative energy sources. One such is familiar to all; namely, the energy in natural clouds. For example, the power represented just by the kinetic energy of a 10 msec^{-1} , 1 km^2 updraft of a cumulonimbus cloud (Cb) is about 1000 megawatts, which is comparable to the power output of a large electrical generating plant. Further, the thermal power released during condensation in the cloud, which is derived from the sun's investment of energy to vaporize the water, can be many hundreds of thousands of megawatts. This dwarfs even the largest conceived nuclear power park. But the questions are: How can this "cloud power" be harnessed and how can it be sustained?

PROPOSED METHOD

Addressing these questions and recalling the problems discussed by F. A. Gifford (personal communications) and his staff^{1,2} concerned with the possible environmental impact of nuclear power parks, leads one to consider the following concept. It is seen from Figure 1 how closely anthropogenic heat releases, especially from cooling towers, approach the power dissipated by cumulonimbus clouds. Considering that anthropogenic activities may already be modifying the weather³ and that future modification, especially from cooling facilities for nuclear power parks, may be even more significant, one is led to the concept shown in Figure 2. This illustration suggests that the (tens of thousands of megawatts of) heat and the (tens of thousands of tons per hour of) water rejected by a nuclear power park be used to stimulate a Cb above a mountain plateau. If,

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FIGURE 1. A Qualitative Plot of Power Dissipated by Various Natural and Anthropogenic Activities as a Function of Their Scale Size.

FIGURE 2. A Suggestion of How to Tap "Cloud Power" Through Stimulating a Cb with a Nuclear Facility's Waste Heat and Utilizing the Water for Cooling, Irrigation and Domestic Purposes and, Especially, for Hydroelectric Power Generation. Whether or not the heat pipes or paved parking lot would be of any advantage is not clear; however, there are suggestions in the literature (see text) that large (~10 thousand megawatts) cooling facilities could stimulate Cb's.

thereby, moist air is enticed up from the plains below then the energy previously squandered by the cooling facility could be used as an investment to tap the latent energy invested by the sun in vaporizing the water, and consequently produce extra rainfall on the plateau. This water could be used later as needed for cooling, irrigation and domestic purposes and, more specifically, for hydroelectric power generation.

BACKGROUND INFORMATION

Thus the concept is basically a plan for dynamic weather modification (although the updraft could be seeded if desired) and perhaps it is useful to note here a few other similar schemes that have been described. For example, on 20 March 1945, F. W. Lee received a U.S. patent (#2,371,629) for "an apparatus for forming artificial thermals for sailplane use...a black body positioned to permit solar rays to impinge thereon to heat said body and turn to heat the adjacent atmosphere thereby causing it to ascend."⁴ Although this may be the first U.S. patent for a dynamical weather modification scheme, there may have been an earlier scheme. There are speculations that in biblical times Babylonians burned their fields after harvest to provide a blackened area to enhance rainfall for the subsequent crop.⁵

Indeed, in a modern version of this idea Black and Tarmy⁵ described a proposal to create an artificial (i.e., thermal) mountain adjacent to a water body by asphaltting a considerable area, thereby stimulating the growth of cumulus clouds and the production of rain. Their theoretical predictions appear quite encouraging, "2 to 3 acres of arable land could be produced per acre of asphalt and the cost of the rainfall would be of the order of three cents per 1000 gallons." However, others have expressed reservations about this concept,⁶ suggesting that an asphalt cover would typically reduce the surface roughness. Therefore, except on very calm days the heated layer would be constrained within a boundary layer of even reduced depth, and also, unless moisture was released simultaneously, then even on relatively calm days the effect might be only to increase the height of the cloud base. However,

if this scheme could assist in the stimulation of Cb's then, returning to the concept shown in Figure 2, it may be of some advantage to surround the cooling towers with a large paved parking lot (since presumably one would be needed anyway!). But obviously this, as well as the entire concept presented here, requires some quantitative analyses.

Another method was described at a recent international weather modification meeting:⁷ "Vulfson and Levin (USSR) described the use of artificially-induced vertical jets of air for modifying convective clouds. Following the original idea behind Dessens' 'meteotron,' 12 jet engines are being used with a power of 500,000 kw to generate updraft velocities of 350 m sec^{-1} and an initial air temperature of 450°C (in 1974 the power will be increased by 20% to produce updrafts of 600 m sec^{-1} and temperatures of 1100°C). Field experiments carried out in the Ukraine during the period 1967-1969 indicated that for condensation levels about 1 km above the ground, clouds were produced above the jets and under favorable conditions these sometimes precipitated (artificial ice nuclei were sometimes injected into the jets)." Although Vulfson and Levin's studies are encouraging one wonders if the clouds could be stimulated more efficiently than with their almost-supersonic turbulent jet. In their case the drag losses would be significant; a moist buoyant plume, as from a cooling tower, would have lower velocity, larger volume to surface area and therefore smaller drag losses. Further, with condensation there is a continuous buoyant body force which also would enhance the possibility of Cb development at the desired location.

Similar inducements of Cb's by large heat and momentum releases have resulted from static test firings of the first stage of the Saturn V.⁸ Although the energy releases were large ($\sim 20 \times 10^{12}$ joules) the release times were short ($\sim 10^2$ sec) so that even though the power dissipated was large (~ 200 thousand megawatts or an order of magnitude more heat release than from the largest nuclear power park currently discussed seriously), the dynamic behavior of the plume would be expected to differ substantially from

the plume from a continuous source. Qualitatively the difference is between the behavior of a turbulent puff and a thermal plume,⁹ and probably most of the kinetic energy of the puff was lost by viscous drag. However, Cb's were induced.

CONCLUDING REMARKS

Upon further consideration of the proposed scheme to use waste heat as a seed for "cloud power" it seems that a major uncertainty is whether or not moist air could be enticed up from the low plains, onto the mountain plateau and into the Cb. That is, whether or not the natural up-slope winds and the cumulus clouds frequently associated with them could be modified by the presence of an anthropogenically stimulated Cb. It seems reasonable to consider using a natural channel such as a mountain valley. In this regard, the observation that "the presence of convective clouds (cumulus or cumulonimbus) over the valley may also cause local currents which completely overwhelm the valley-wind circulation"¹⁰ is an encouraging statement.

If the proposed scheme has merit, then the first task would be to identify sites which appear favorable from climatological, topographical, meteorological and power-economical viewpoints, and to model the proposed scheme both numerically and with laboratory simulations. Then, armed with this experience, field tests might be designed and executed to determine statistically significant results. Of course a major task would be to assess the environmental impact of the proposed scheme but then the following viewpoint should be considered: if nuclear power parks are contemplated and if their waste heat could stimulate Cb's, then why not stimulate the Cb's in a location where their consequences would be most beneficial? After all, clouds do represent the world's most astounding desalinization, irrigation, and electrical generating projects.

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ASSESSMENT OF CLIMATIC CONSEQUENCES OF
POWER PRODUCTION POLLUTION

R. A. Stokes and A. J. Alkezweeny

The objective of this study is to determine if the unique polarimetry capabilities at the Rattlesnake Observatory can be used for in situ measurements of the complex index of refraction of the atmospheric aerosol. The study tests the only conceived method for such in situ measurements, and will hopefully obtain unbiased data for input to the radiation portion of models for the earth's climate.

Measurements of the polarized intensity of backscattered radiation were made on three nights in May and June during different weather conditions. Direct aircraft sampling of the atmospheric aerosol layer responsible for the effect enabled us to compare theory with observations. We are now confident that the Rayleigh-scattered light can be reliably subtracted, and the remaining polarized intensity related directly to the properties of the aerosol particles. Our remaining objective is to extend the range of meteorological conditions occurring during the measurements in order to develop a technique with the widest applicability.

INTRODUCTION

In recent years considerable interest has been shown in the effect of air pollution on the climate. Changes in the amount and frequency of precipitation have been detected with increasing man activities^{1,2,3} and the problem of inadvertent weather modification as a result of air pollution has also been reviewed recently.⁴ The effect of aerosol particles on the climate can be divided into two parts: 1) the role of particles as ice and cloud condensation nuclei in cloud formation and precipitation development^{5,6} and 2) the role of particles in the scattering and absorption of radiation passing through the earth's atmosphere.

The scattering of light by aerosols is the result of either Mie scattering or Rayleigh scattering, depending on the ratio of the wavelength of the light to the effective

particle diameter. Both processes result in light which can be strongly polarized.

Except for a few qualitative interpretations, polarimetry has only recently been used effectively for studying planetary atmospheres. Primarily through the theoretical and observational work of Coffeen and Hansen,⁷ polarimetry was used to determine several of the aerosol properties of the Venusian atmosphere. The real part of the index of refraction, the mean particle radius and shape, and the size distribution were found using linear polarization measurements as a function of color and phase angle. These properties determine how sunlight is scattered by the aerosols. The fraction of the solar radiation that is able to enter the atmosphere or strike the surface can, therefore, be analyzed from the scattering properties determined by linear polarization measurements.

In special circumstances the scattered light may even be circularly polarized. In this case the chemical composition of the particles becomes an important variable since the circular polarization may be largely determined by the imaginary part of the index of refraction of the aerosol. Thus, if one were able to completely characterize the intensity and polarization state of scattered light as a function of wavelength and scattering angle, it should be possible to infer a wide range of aerosol properties by use of remote, ground-based sensing techniques.

Recently Harris⁸ calculated the polarization properties of aerosol particles as a function of scattering angle. The particle size distributions used in his calculation were based on his measurements in the Los Angeles Basin. His computations were carried out with different wavelengths of light, and different real and imaginary parts of the aerosol refractive index. The results show that the polarization depends strongly on the wavelength and the refractive index.

The refractive index of aerosol particles in the atmosphere is expected to be quite different from that of bulk material. One of the reasons for this is that the particle may act as a nucleus for chemical reaction; also, the particle may grow due to increase in the relative humidity. Therefore its optical properties may change considerably. The present state of the art for determining the refractive index of airborne particles requires that a sufficiently large bulk sample of the aerosol be collected and returned to the laboratory for analysis.⁹ However, this technique may change the particle characteristics and, hence, change its optical properties.

INSTRUMENTATION

The Battelle polarimeter uses a modulating element developed by Kemp.¹⁰ This is a piece of fused silica mechanically coupled to a 50-kHz oscillator (50 kHz being a resonant frequency of the silica bar). The process of compressing and stretching along the length of the bar produces a quarter-wave plate whose fast and slow axes alternate at the above rate. The action of the quarter-wave plate

changes circularly polarized light to linearly polarized light at 45° and 135° alternately. By orienting a linear polaroid (after the silica bar) at one of these two angles, we are able to detect a 50-kHz signal from the phototube for any circularly polarized beam that may be incident.

In practice we use a digital lock-in amplifier to count pulses produced by photon events. Counts accumulate in one counter during one half-cycle and in a second counter during the other half-cycle. The difference in counts divided by the sum of the counts yields the polarization.

For linear measurements we introduce quarter-wave plates (before the silica bar) whose axes are oriented 45° to each other such that the two linear polarizations represented by the second and third Stokes vectors are circularly polarized after passage through these plates. A circular polaroid is inserted into the beam (before the silica bar) to determine how effectively the system measures circularly polarized light.

The system may be able to detect one-handed circular polarization better than the other. To eliminate this asymmetry the linear polaroid sheet that follows the silica bar is rotated every other integration period. To check that the mirror does not introduce polarization upon reflection, we examined some of the polarized and nonpolarized standards of Serkowski.¹¹ In performing circular polarization measurements, residual stresses in the modulating element have a tendency to change linear polarization to circular polarization. This effect is cancelled by a rotation of the whole polarimeter. Circular polarization is so small, typically, that circular-to-linear conversion is not a problem in linear measurements and this rotation is not needed. Our results agree (within standard errors) on all stars examined. Use of a digital synchronous detection system to obtain signals enjoys an advantage over polarimeters employing analog detection schemes in that fluctuations in both sum and difference (due to sky changes and instrument drifts) are monitored simultaneously thus allowing highly accurate polarimetry on less than photometric quality nights.

The Battelle polarimeter was attached to the Cassegrain focus of the 31 in. reflecting telescope for this work. A 1000-watt projection lamp was placed at the focus of a 5-ft diameter searchlight to produce a nearly parallel beam of light. Polarization measurements of the light scattered by the aerosols over Rattlesnake Mountain were made by viewing the vertical beam at several angles with the telescope-polarimeter. Color measurements were also performed. Simultaneous measurement of the concentration and particle size distributions were made by Battelle's aircraft.¹² The measuring instrumentation consisted of a Royco Model 220 Optical Sensor, a pulse converter, a pulse height analyzer (PHA), and a digital printer. The optical sensor measured the 90° light scattering from a single particle; its output pulses were fed into the pulse con-

verter and then to the PHA, where the pulses were stored in its memory according to their height for the preset sampling period. Pulse height is proportional to the particle size. The system, which was calibrated with latex spheres of known sizes in the laboratory to relate the channel number in the PHA to the particle sizes, and is capable of simultaneously measuring size distributions of aerosol particles in the range from 0.3 to 5 μm diameter in about 100 channels.

PRELIMINARY RESULTS

The results of the polarization measurements for two nights in May and one night in June of 1974 are presented in Table 1 and Figure 1; 03/04 May, it will be noticed, are the dates the airplane samples were taken.

TABLE 1. Linear and Circular Polarization Results.¹

<u>Date</u>	<u>Filter</u>	<u>Linear or Circular Polarization¹</u>	<u>Scattering Angle</u>
5-02/03-74	r	.078±.007	25°
	B	.083±.002	
	r	.225±.007	39°
	B	.257±.005	
	r	.860±.149	71°
	B	.998±.102	
5-03/04-74	r	.052±.002	9°
	B	.063±.002	
	r	.263±.015	34°
	B	.284±.009	
	r	.450±.026	58°
	B	.533±.020	
6-11/12-74	r	.180±.007	42°
	B	.220±.003	
	r	.376±.010	57°
	B	.429±.008	
	r	.491±.008	70°
	B	.639±.009	
5-02/03-74	r	+.00143±.00041	71°
	B	+.00158±.00039	

¹Last two entries are the only circular polarization results

FIGURE 1. Linear Polarization of Light Beam Versus Scattering Angle.

Each night of observations shows significant differences from every other night. A summary of weather conditions precedes a description of polarimetric results on each night.

02/03 May

On May 1 cooler air was advected over Eastern Washington as a surface and 850 mb ridge moved from the Pacific Ocean over the area. This was followed by weak warm air advection starting during the evening of May 3. The air-mass

was dry so there was no precipitation over the area. The only clouds over the area were high-thin cirrus. Surface winds on top of Rattlesnake Mountain (3580 ft) were southwesterly 12-26 knots early on May 1, and shifted to northwest 15 knots as the ridge moved into Eastern Washington. The winds decreased to <5 knots and became variable on May 3 and 4. Temperatures on top of the mountain ranged from 38°F on May 1 to 50°F on May 4. The polarization curves for this night reveal a very high degree of polarization

at large scattering angles. The preponderance of Rayleigh scattering is revealed in the curves' shape in that a function

$$p(\theta) = \alpha \frac{1 - \cos \theta}{1 + \cos \theta},$$

where p is the linear polarization and α is a constant, fits the curves reasonably well.

03/04 May

On this date the airplane was flown and samples were taken at

altitudes of 4000, 5000, 6000, and 7000 ft. No significant change with altitude was observed as can be seen in Figure 2. A bit more dust in the atmosphere may be revealed by a suppression of the amount of polarization in this night's polarization curves at large scattering angles--an indication that Rayleigh scattering may have been less dominant.

11/12 June

There was an upper ridge over eastern Washington June 10 through

FIGURE 2. Particle Size Distribution Over Rattlesnake (May 3, 1974, 10:00 PM).

12, resulting in a westerly flow above 7000 ft. The winds below 7000 ft were light and variable. The surface winds on top of Rattlesnake Mountain (3580 ft) were variable <5 knots. Daytime temperatures on top of the mountain ranged from 73°F to 78°F. A distinct inversion layer could be seen at sunset. The layers which contained an appreciable amount of dust and practically no dust could easily be distinguished visually. Again this is evidenced in the polarization curve, as the least amount of linear polarization of all three nights is observed here at all angles.

It should be noticed that all three nights show the presence of Mie scattering, i.e., scattering by particles whose dimensions are $\geq$ the visible wavelengths. This can be seen in that the blue light polarization is always larger than the red. For a pure Rayleigh scatterer, the opposite is true because the Rayleigh optical depth is always greater at shorter wavelengths. The greater the optical depth the greater the chance of multiple scattering and depolarization of the light. Furthermore, based on the power spectrum of the incandescent light bulb and the wavelength responses for the polarimeter-photometer system, more than Rayleigh scatterers may be present because the red intensity response is significantly lower than that expected for a pure Rayleigh atmospheric layer.

Considering these latter two circumstances observed in our data, a good opportunity for subtraction of the Rayleigh component of polarization exists. After this subtraction is made, a comparison of the polarization due to the Mie scatterers (aerosols) and that expected from the measured size and number distribution should be made.

DISCUSSION

It is known that single scattering of unpolarized radiation cannot produce circular polarization except in the instance of aligned, elongated

particles.¹³ Due to the small optical depths, most of the light scattered from the beam suffered only a single scatter. Hence, we did not expect to see circular polarization in the signal. The fact that a small circular polarization was observed probably was the result of the polarization produced by the incandescent light. (It is a general rule to expect a few percent polarization for all artificial sources).

Real opportunities, however, appear to exist for a determination of the imaginary index of refraction using circular polarization measurements. Circular polarization of twilight, when multiple scattering of sunlight in the atmosphere is the greatest, has been measured by Angel, Illing and Martin.¹⁴ Hannemann and Raschke¹⁵ find significant circular polarization in the sunlit sky over the heavily industrialized Ruhr Valley where aerosol concentrations are heavy; generally, skylight shows a very low circular polarization.¹⁶

The short pathlengths preclude circular polarization because the multiple scattering contribution is small; however, this barrier may be overcome by linearly polarizing the beam so that single scatterings may produce a circularly polarized component. The wavelength and angular dependences of this component should reveal the imaginary index of refraction.

Several improvements have already been made to render the measurements more useful. Many possibilities yet remain. The observations can reasonably be extended to 15,000 to 20,000 ft, if need be, with the present light source and even farther with a more intense source. Nothing will be lost in scattering angle coverage because the mirror can be moved and tilted to insure this coverage.

The study has made us aware of a number of possibilities and problems, but a characterization of aerosols seems possible. Further theoretical analysis and more extensive experimental results should soon make these investigative possibilities a reality.

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STABILITY TESTS OF SEVERAL DIFFERENT SUNPHOTOMETERS

N. S. Laulainen

A discussion of stability tests of two different sunphotometers is presented. Attempts were made to determine the causes of apparent zero-point drift in these instruments. Variable temperature and atmospheric turbidity are identified as potential drift sources. Further tests are needed to unambiguously determine the nature of these uncertainties in instrument calibration.

INTRODUCTION

Measurement of solar radiation at the earth's surface can provide important information about aerosols and trace gases in the atmosphere. A large body of data relating to atmospheric turbidity--a measure of light attenuation due to suspended particulate material--has been collected for nearly 20 years by various networks.¹ Recent studies by the author^{2,3} have indicated that large apparent zero-point variations in the instruments typically used for these measurements, may seriously contaminate these data. To further identify causes and assess the consequences of this effect, a study is underway to evaluate current and new instrumentation and techniques for measuring insolation and atmospheric turbidity. This report compares measurements obtained with two different types of sunphotometers at the Hanford Meteorological Station since July 1974.

DEFINITIONS

The basic equation relating the measured light intensity to the attenuation in the atmosphere is the Beer-Lambert law:

$$J = J_0 \exp(-\tau m)/F, \quad (1)$$

where J is the actual photometer reading,

J_0 is the outside-the-atmosphere photometer reading (zero-point),

F is a correction factor depending on solar distance,

τ is the optical depth, and

m is the relative airmass.

The optical depth, τ , is the sum of attenuation contributions due to Rayleigh (molecular) scattering, ozone absorption, and aerosol scattering and absorption.

From the standpoint of data analysis, the Beer-Lambert law is more conveniently written as

$$\ln J = \ln J_0 - \ln F - \tau m. \quad (2)$$

This form clearly shows the relation between the dependent variable, $\ln J$, and the independent variable, m . Indeed, a standard procedure for simultaneously determining the zero-point intercept, $\ln J_0 - \ln F$, and the optical depth (slope), τ , is a least squares solution to Eq. (2), from measurements of $\ln J_0$ for various values of m . This procedure requires that $\ln J_0$ and τ be constant over the time interval during which the observations are made; clearly, if $\ln J_0$ and/or τ are variable, then the determination of both of these quantities will be in error. The following questions then arise: How might such variations be detected? How does one distinguish between the two types of variations? One anticipates that zero-point variations may arise from temperature-induced or intensity-induced (saturation) sensitivity changes during a sequence of measurements, or from aging of the photoelectric cell and color filter over longer periods of time. The data seem to indicate that the latter effect is,

in most cases, relatively unimportant. On the other hand, turbidity variations could also be temperature dependent; it is more reasonable to assume, however, that these variations also depend on other meteorological variables, and should therefore be distinguishable from the instrumental variations. Only careful experimental tests can reveal the true nature of these variations.

Several tests, then, can be performed to distinguish between true τ variations and true $\ln J_0$ variations. If $\ln J_0$ variations are random, then making a number of near-simultaneous observations with two or more photometers should reveal no correlations if $\ln J_0$ varies, and rather strong correlations if the turbidity varies. If $\ln J_0$ variations are temperature dependent, then the above procedure would result in definite correlations for either type of variation. Finally, with a substantial investment in additional equipment, these variations could be distinguished with the aid of a standard light source. Parenthetically, it might be pointed out that Volz-type sunphotometers are attractive because of their low cost, portability and ease of operation; the necessity of using a standard lamp to check instrument stability completely negates these features. On the other hand, confidence in data reliability is also essential.

EXPERIMENTAL PROCEDURES AND REDUCTIONS

Tests on two different types of sunphotometers were begun in July 1974 at the Hanford Meteorological Station. One instrument was a Climet Volz-type sunphotometer using a single broad band filter (Wratten 65) centered at 500 nm in wavelength; the other was a 6-color sunphotometer developed at the University of Washington (on indefinite loan to the author), including a 500 nm filter identical to the Climet model, as well as a narrow band interference filter centered at 500.7 nm. Interference filters at 448.2 nm, 557.0 nm, 600.0 nm and 616.0 nm were also available. The sensing unit for the Climet (hereafter V1) was a selenium photovoltaic cell; the 6-color photometer (hereafter P) used a vacuum photodiode. Both photosensors have an upper wavelength cutoff around 650 nm.

Toward the end of August 1974, a second Climet photometer was also used.

Data were obtained by pointing the instrument at the sun and recording both the intensity reading and the time of day; the latter, when used with the solar coordinates, allowed the calculation of the solar zenith angle and thus the airmass path traversed by the solar beam. Data for those days where the airmass base line was at least $\Delta m \geq 1.5$ were fitted to Eq. (2) and the corresponding zero-point, $\ln J_0 - \ln F$, and optical depth, τ , determined. The photometers were normally stored at room temperature between measurements.

On several days, when the outdoor temperature was 15-20°F above that indoors, temperature sensitivity tests were also carried out. Both photometers were observed to produce significantly different readings as the instruments warmed up from room temperature to ambient outdoor temperature. In the case of V1 a 4-5% decrease was noted for a temperature difference of about 20°F. Photometer P showed a similar decrease for the blue (448.2 nm) and green (500 nm) wavelengths, but showed essentially no change for the yellow (557.0 nm), and an increase for the red (600 nm, 616 nm) wavelengths. The behavior of P with temperature suggests that the vacuum photodiode causes most of the variation, although transmission characteristics of the filters themselves are also known to change with temperature. The behavior of V1 is also consistent with photocell temperature response. These observations would lead one to expect a rather good correlation between $\ln J_0$ and temperature. No definite correlations were apparent.

In order to test for the cause of apparent zero-point variation, correlations between the zero-point of V1 and the various zero-points of P were sought, as shown in Figures 1-4. Inspection of these graphs reveals that the day-to-day variation of $\ln J_0$ tends to be larger than the assigned least-squares uncertainty (as shown by the error bars), and that there are only weak to medium correlations. The strongest of these are for those filters (blue and green) which have the temperature effects in the same sense as V1; the weakest is

Neg 66340-5

FIGURE 1. V1 Zero-point Versus P-488.2 nm Zero-point;
Moderate Correlation.

Neg 66340-8

FIGURE 2. V1 Zero-point Versus P-500 nm Zero-point;
Moderate Correlation.

Neg 66340-2

FIGURE 3. V1 Zero-point Versus P-557 nm Zero-point;
Weak Correlation.

Neg 66340-4

FIGURE 4. V1 Zero-point Versus P-600 nm Zero-point;
Weak Correlation.

for the yellow filter, which shows no noticeable temperature effect. On this basis, one might expect a negative correlation in the case of the red (660 nm) filter, but one in fact finds a weak positive correlation. As a further test, the zero-point of V1 is compared to that of the second Climet (V2); these results are shown in Figure 5. In this case no correlation is found. Since V1 and V2 are supposedly identical, this finding is rather surprising. Unfortunately, there were only 3 days in which all three instruments were operated simultaneously, so that these latter results are somewhat tentative.

The results presented here still do not resolve the question of the origin of the apparent zero-point

variation, since temperature and/or turbidity variations were not unambiguously identified. Further tests will be required. It is planned to carry out observations at the Battelle Observatory on Rattlesnake Mountain with these three instruments as well as a new prototype photometer presently being developed. Because of its elevation (1080 m), turbidity variations above Rattlesnake should be reduced. In addition, obtaining aerosol vertical profiles using the aerosol equipment aboard the Battelle Cessna 411 aircraft will be of considerable value in assessing the effects of turbidity variations. Finally, regular calibration of the photometers against a standard light source is also planned.

Neg 66340-5

FIGURE 5. V1 Zero-point Versus V2 Zero-point; No Correlation.

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ATMOSPHERIC TURBIDITY OVER HANFORD

N. S. Laulainen

Multiwavelength turbidity data obtained by sunphotometry techniques at the Hanford Meteorological Station are presented. In addition to large observed day-to-day variations in turbidity, an anomalous red "hump" is observed in most aerosol optical depth spectra which cannot entirely be accounted for by ozone absorption.

INTRODUCTION

The aerosol component of the earth's atmosphere is highly variable, particularly within the lowest 1-2 km of the surface. The presence of this aerosol has important effects on the incident solar radiation, resulting in some cases in severely reduced insolation at the surface and/or in distortions of the solar energy spectrum. Measurements of direct solar intensity with a sunphotometer are useful for quantitative assessment of light attenuation due to aerosols over a wide range of wavelengths. In a previous report,¹ instrumental stability of two types of sunphotometers was discussed. The data used in those studies also apply to aerosol extinction or turbidity studies. This report summarizes the turbidity measurements at Hanford Meteorological Station from 12 July to 30 October 1974.

For completeness, the basic equation governing the extinction of light in the atmosphere (the Beer-Lambert law) is restated:

$$J = J_0 \exp (-\tau m) / F, \quad (1)$$

where

J is the actual photometer reading

J_0 is the outside-the-atmosphere photometer reading,

F is a correction factor depending on solar distance,

τ is the optical depth or extinction factor through the zenith, and

m is the relative airmass (thickness) traversed by the light beam.

The optical depth τ is the sum of attenuation or extinction contributions due to Rayleigh scattering, ozone absorption, and aerosol scattering and absorption; explicitly it is

$$\tau = \tau_{\text{Rayleigh}} + \tau_{\text{ozone}} + \tau_{\text{aerosol}} \quad (2)$$

The Rayleigh contribution is relatively constant and predictable. The ozone contribution comes mainly from stratospheric ozone and varies with time of year and latitude by as much as 30%. The aerosol contribution, however, is highly variable and is the emphasis of this report. For lack of more detailed information, the Rayleigh and ozone contributions are assumed to be the standard atmosphere values tabulated by Elterman.² While this procedure is not particularly satisfying, especially for the ozone component, it is reasonable at this time, until the question of instrument stability (whose uncertainties, incidentally, may contribute anywhere from 5 to 10% error in the determination of the total optical depth)--is resolved. Simultaneous and independent measurements of ozone, both stratospheric and tropospheric levels, would also be of considerable value in determining aerosol extinction values accurately.

RESULTS AND DISCUSSION

Determination of total optical depth can be done in several ways. If the photometer has a stable, known

zero-point (J_0), the optical depth can be determined simply by

$$\tau = [\ln (J_0/F) - \ln J]/m. \quad (3)$$

As pointed out in the previous report,¹ there are some unresolved questions of instrumental stability, so that the reliability of this procedure is unknown. Alternatively, by arranging Eq. (3) to

$$\ln J = \ln (J_0/F) - \tau m, \quad (4)$$

one can perform a least squares fit to Eq. (4) for many observations of J and m and obtain an apparent zero point and optical depth. This procedure still contains uncertainties due to potential zero-point and/or optical depth variation. However, by choosing observation runs of one to two hours, when the airmass is changing rapidly with time, (sun rising or setting) then both types of variations can be minimized in many cases. Most of the observations reported here were reduced according to this latter procedure; only a few days were analyzed according to Eq. (3), using a mean value of J_0 from several least square determinations of the zero-point.

Optical depth values for three different wavelengths are shown in Figure 1 for 20 days (12 July to 30 October 1974) at the Hanford Meteorological Station. The values identified by V1 were obtained with a single wavelength Volz-type Climet sunphotometer centered at 500 nm; those identified by P were obtained with a six-wavelength sunphotometer developed at the University of Washington (on indefinite loan to the author). Optical depths for only two of the six wavelengths are shown in Figure 1, namely at 448.2 nm and 600 nm, respectively. An interesting increase in optical depth seems to have occurred during this time interval; similar observations have been reported elsewhere.^{3,4} Some caution must be exercised in making such conclusions, since the observations are biased toward clear days. However, since late summer and early fall in this region are times of increased agricultural activity, and frequent meteorological inversion conditions tend to trap any dust and other suspended particulate material, such conclusions are not unreasonable either.

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FIGURE 1. Total Optical Depth in Three Colors (448.2 nm, 500 nm, and 600 nm) at the Hanford Meteorological Station from 12 July - 30 October 1974.

In order to determine the aerosol optical depth or turbidity, the Rayleigh and ozone contributions are subtracted from the total optical depth, as indicated in Eq. (2). This procedure is shown graphically in Figure 2. The uncertainties in the total optical depth for the case shown are of the order of the size of the open circles. Even assuming no error in the Rayleigh and ozone contributions, one notices immediately that the subtraction procedure results in rather large uncertainties in the aerosol optical depth, as shown by the error bars. In spite of these uncertainties, the aerosol optical depth spectrum appears to go through a minimum around 540-550 nm. The position of the point labelled P2 (photometer P, filter 2) at 500 nm is disturbing and needs to be examined closely, since the presence of this point could also lead to the conclusion of a gray aerosol optical depth spectrum (i.e., particles larger than $\sim 1 \mu$).

Both photometers V1 and P have identical broad band Wratten 65 filters centered at about 500 nm. In addition, photometer P has a narrow band interference filter centered at 500.7 nm (labelled P3 in Figure 2); the value P3 is in good agreement with V1. The difference between P2 and V1 may be due to the difference in photodetectors, since V1 has a selenium photovoltaic cell and P has a vacuum photodiode with S4 response. The red cutoff of these two detectors is somewhat different, with the vacuum photodiode cutoff slightly shorter than that of the selenium cell. A shift of the effective wavelength response of photometer P and the Wratten 65 filter by 10 nm toward the blue will lower the aerosol extinction value to P2', due to increased Rayleigh attenuation. Thus it is clearly necessary to carefully determine the effective wavelength response of filter P2. This result illustrates well one of the problems encountered with broad band filters. In subsequent analysis averages of three values for V1, P2, and P3 are taken to be representative of the turbidity at 500 nm.

Returning to the shape of the aerosol extinction spectrum, it appears that the red "hump" could be real. Insufficient ozone correction could also produce a red hump, but though ozone correction which was

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FIGURE 2. Graphical Technique for Obtaining Aerosol Optical Depth from Total Optical Depths.

used may be in error by 20-30%, subtracting larger corrections does not remove the hump entirely--even for unrealistically large ozone concentrations. A power law aerosol size distribution predicts an aerosol spectrum of the form

$$\tau_{\text{aerosol}} = \alpha \lambda^{-\beta}, \quad (5)$$

which is a single valued function of λ for all β except zero.⁵ If the anomalous behavior observed here due to aerosol, then a more complex aerosol size distribution than a simple power law is required. This problem will be addressed in detail at a later time.

Figure 3 presents wavelength dependent turbidity data for eleven different days. In each case the red hump is present. Also striking is the variability of the turbidity--as much as a factor of 3-5, depending on the wavelength, between minimum

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FIGURE 3. Wavelength Dependence of Aerosol Optical Depth for 11 Days at the Hanford Meteorological Station During the Last Half of 1974.

and maximum values. It is also interesting to note that the wavelength dependence changes from day to day, perhaps signifying changes in particle size distribution or aerosol complex index of refraction or both.

It is clear from this discussion that much valuable information about aerosol characteristics and variability are contained in multi-wavelength turbidity data. Continued testing of various sunphotometers and acquisition of additional aerosol optical depth data are planned. Of particular interest are techniques for determining vertical profiles of suspended particles, examining day-night variations in turbidity (by conducting nighttime stellar extinction in conjunction with daytime sunphotometry at the Rattlesnake Mountain Observatory), and continued examination of seasonal variations in atmospheric turbidity.

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A PRELIMINARY PREDICTION MODEL FOR ALE RESERVE PRECIPITATION

J. M. Thorp, W. T. Hinds* and Ethel S. Gilbert**

Monthly precipitation data from 23 Arid Land Ecology (ALE) Reserve stations have been recorded since 1968. Bioyear (October-May) precipitation averages for these stations have been recently calculated for the period 1969-1974. A simple model is proposed which incorporates a ratio estimate of the 6-year precipitation means of the 23 ALE stations and the 1969-1974 6-year precipitation mean at the Hanford Meteorology Station. This model attempts to predict bioyear precipitation at any ALE Reserve station from any year for which data are complete for the Hanford Meteorology Station. The model is optimal when the intercept, α , is zero, and the variances of the data are proportional to the means.

Precipitation measurements on the Arid Land Ecology (ALE) Reserve began in 1968. With the addition of May 1974 precipitation data, we are able to calculate 6-year averages of bioyear (October-May) precipitation for 23 of the ALE climatology stations.

Scientists conducting investigations on the ALE Reserve are handicapped at times by the lack of long term precipitation records for the Reserve and only through continuance of the microclimatological measurements will this problem be overcome.

An interim solution, however, may provide reasonably reliable estimates of precipitation for years prior to 1968. This approach makes use of the precipitation records from the Hanford Meteorology Station (HMS) which go back to 1947, and the old Hanford town site records which were made from 1913 through 1942. The HMS is located less than 10 km north of the ALE Reserve boundary. If it is possible to significantly relate the 6-year ALE Reserve precipitation averages with the corresponding 6-year average at the HMS then it should be possible to estimate, within certain

statistical limits, the bioyear precipitation at any ALE Reserve station for any year from 1913 through 1942 and from 1947 through 1968.

One approach to establishing a relationship between the means is by grouping the 23 stations into sets based on the average precipitation. Another grouping was based on the regression coefficient, β , after precipitation data from each station were regressed on corresponding Meteorology Tower data. However, the 1973-1974 bioyear precipitation varied from 147% to 198% of the 1969-1973 average, and including these data with those of the previous five years altered the slopes of all the stations.

Instead of regrouping the stations by the new slopes, the standard regression equation was replaced by a ratio estimate. Thus, to estimate Y_{Ki} , the precipitation at Station K in year i

$$\hat{Y}_{Ki} = \frac{\bar{y}_K}{\bar{x}} x_i, \quad (1)$$

where

x_i is the Hanford Meteorology Station value for year i

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$\bar{y}_K$ is the mean for Station K of 6 bioyears of data

$\bar{x}$ is the mean for HMS of the 6 bioyears of data.

This equation is optimal when the intercept, α , is zero, and when the variances of the data are proportional to the means. Table 1 lists the slopes, variances, intercepts and means for the ALE Stations arranged according to the original grouping which preceded the bioyear 1974 data.

The intercept estimates, α , are plotted in Figure 1; it appears that the largest deviations of α from zero are from stations clustered high on

the southeast end of Rattlesnake Mt. (sites 20, 25 and 26). When the 6 year means are plotted versus the variances, σ^2 , these same three stations also show dispersion (Figure 2), though proportionality at most sites is established.

Using the admittedly inadequate sample of only six bioyears of data, it appears feasible to statistically estimate bioyear precipitation at most of the ALE Reserve sites for years between 1947 and 1968 by using data from the Hanford Meteorological Station, and possibly for the years 1913-1942 when similar records were kept near the Hanford townsite.

TABLE 1. 1969-1974 Bioyear Precipitation Statistics ALE Reserve Microclimatological Network.

STATION	MEAN (cm)	SLOPE β	STANDARD DEV. σ	RATIO $\bar{y}_K/\bar{x}$	VARIANCE σ^2	INTERCEPT $\hat{\alpha}$
041	22.8	1.41	7.53	1.56	56.70	2.21
091	23.2	1.52	5.12	1.59	65.93	1.30
141	22.7	1.52	8.41	1.55	70.73	0.80
150	21.6	1.37	7.16	1.48	51.27	1.60
191	22.9	1.47	7.95	1.57	63.20	1.44
251	20.8	1.09	5.71	1.42	32.60	4.89
081	19.6	1.27	6.71	1.34	45.02	1.06
071	20.9	1.43	7.89	1.43	62.25	0.22
231	19.4	1.17	6.10	1.33	37.21	2.32
241	19.7	1.23	6.36	1.35	40.45	1.74
181	17.7	1.13	6.20	1.21	38.44	1.20
211	17.0	1.05	5.58	1.16	31.14	1.67
221	18.4	1.25	6.45	1.26	41.60	0.15
101	24.6	1.55	8.11	1.68	65.77	1.97
111	24.6	1.71	8.87	1.68	78.68	-0.37
201	21.5	1.22	6.32	1.47	39.94	3.69
121	20.0	1.36	7.04	1.37	49.56	1.44
260	19.8	1.15	5.94	1.36	35.28	3.01
011	14.4	0.91	4.76	0.99	22.66	1.11
021	15.2	1.07	5.58	1.04	31.14	-0.42
051	15.2	1.01	5.26	1.04	27.67	0.54
131	15.7	1.17	6.32	1.08	39.94	-1.38
061	16.8	1.10	5.94	1.15	35.28	0.74

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FIGURE 1. Values of the Intercept Estimate, α , Bioyear (October-May) Data for 1969-1974.

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FIGURE 2. Proportionality of Mean Bioyear (October-May) Precipitation to Variance, 1969-1974.

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