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Pacific Northwest Laboratory Annual Report for 1981 to the DOE Office of Energy Research

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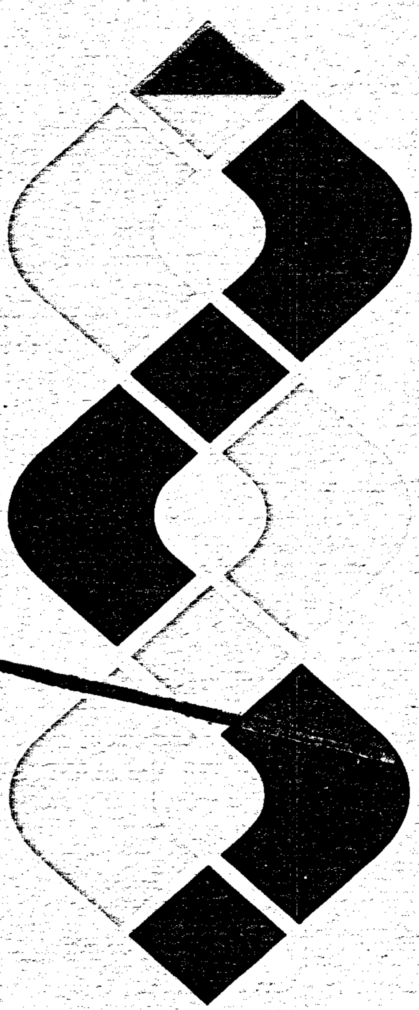
Part 3 Atmospheric Sciences February 1982



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Pacific Northwest Laboratory Annual Report for 1981 to the DOE Office of Energy Research

Part 3 Atmospheric Sciences

C. E. Elderkin and Staff Members
of Pacific Northwest Laboratory

February 1982

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

PREFACE

This 1981 annual report from Pacific Northwest Laboratory (PNL) to the Department of Energy (DOE) describes research in environment, health, and safety conducted during fiscal year 1981. The report again consists of five parts, each in a separate volume.

The five parts of the report are oriented to particular segments of our program. Parts 1 to 4 report on research performed for the DOE Office of Health and Environmental Research in the Office of Energy Research. Part 5 reports progress on all research performed for the Office of the Assistant Secretary for Environmental Protection, Safety and Emergency Preparedness. Each part consists of project reports authored by scientists from several PNL research departments, reflecting the interdisciplinary nature of the research effort. Project reports in Parts 1 to 4 are organized primarily by energy technology.

The parts of the 1981 Annual Report are:

Part 1: Biomedical Sciences Program Manager - H. Drucker	D. L. Felton, Editor
Part 2: Ecological Sciences Program Manager - B. E. Vaughan	B. E. Vaughan, Report Coordinator C. M. Novich, Editor
Part 3: Atmospheric Sciences Program Manager - C. E. Elderkin	R. L. Drake, Report Coordinator N. M. Burleigh, Editor
Part 4: Physical Sciences Program Manager - J. M. Nielsen	J. M. Nielsen, Report Coordinator I. D. Hays, Editor
Part 5: Environmental and Occupational Protection, Assessment, and Engineering Program Managers - D. L. Hessel S. Marks W. A. Glass	W. A. Glass, Report Coordinator R. W. Baalman, Editor

Activities of the scientists whose work is described in this annual report are broader in scope than the articles indicate. PNL staff have responded to numerous requests from DOE during the year for planning, for service on various task groups, and for special assistance.

The topic of the 21st Hanford Life Sciences Symposium, which was held this year, was "Biological Availability of Trace Metals: Chemical Estimation, Ecological and Health Implications."

A highlight of the year was completion of the third floor of the office wing of the Life Sciences Laboratory. This opens up substantial laboratory space by allowing investigators to move offices from the laboratory to the new area.

Credit for this annual report goes to many scientists who performed the research and wrote the individual project reports, to the program managers who directed the research and coordinated the technical progress reports, to the editors who edited the individual project reports and assembled the five parts, and to Ray Baalman and Irene D. Hays, editors in chief, who directed the total effort. The size of this year's report has been reduced as the result of an effort to report more fully in the open literature.

W. J. Bair, Manager
S. Marks, Associate Manager
Environment, Health and Safety Research
Program

Previous Reports in this series:

Annual Report for

1951	W-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; BNWL-361
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
1975	BNWL-2000, Pt. 1-4
1976	BNWL-2100, Pt. 1-5
1977	PNL-2500, Pt. 1-5
1978	PNL-2850, Pt. 1-5
1979	PNL-3300, Pt. 1-5
1980	PNL-3700, Pt. 1-5

FOREWORD

The goals of atmospheric research at Pacific Northwest Laboratory (PNL) are to assess, describe and predict the nature and fate of atmospheric pollution and to study the impacts of pollutants on local, regional and global climates. The pollutants being investigated are those resulting from the development and use of four energy resources: coal, gas, oil and nuclear power. In the course of this research, investigative tools are also being developed and atmospheric assessments are being made that will contribute to the development of environmentally acceptable oil shale, solar and fusion energy resources.

Coal, Gas and Oil Combustion

The behavior of air pollution resulting from fossil-fuel power plants is being examined. Involved in making this examination are these factors: the type of pollutants emitted, their transport and diffusion in the air, their physical and chemical transformations during transport, their removal by wet and dry deposition processes, and their impacts on climate, bodies of water and living species. Since a result of the current and projected coal and oil utilization is the release of large quantities of particulate matter and sulfur and nitrogen compounds, current research is being conducted primarily in the Atmospheric Studies in Complex Terrain (ASCOT) and DOE's Carbon Dioxide Program.

Fission and Fusion

Concern about long-lived particulates (i.e., plutonium and other radionuclides) released to the environment from fission and fusion plants indicates that the deposition and resuspension of these substances must be studied. For example, because the primary hazard from plutonium is inhalation, its residence in the atmosphere must be clearly defined. Current research is evaluating the removal of particulates from the atmosphere by deposition (which limits initial exposure) and any future resuspension from the surface (which continues the potential for inhalation).

Oil Shale

The mountainous oil shale regions of Colorado, Utah and Wyoming present a particularly difficult air pollution problem because air may be trapped in the valleys of these regions for extended periods of time under certain meteorological conditions. Therefore, especially stringent siting requirements must be fulfilled to meet state and federal air quality standards. Adequate models and field measurements of the complex airflow and dispersion conditions in this complex area are not available; they must be developed to assure acceptable siting of oil shale facilities. PNL has undertaken the assessment of the requirements for proper modeling activities and field measurement programs that will contribute to this important area of research, and to the development of solar and geothermal energy.

The description of atmospheric research at PNL is organized in terms of energy technologies:

- Coal, Gas and Oil
- Fission and Fusion
- Oil Shale

This report describes the progress in FY 1981 for each of these technologies. A divider page summarizes the goals of each area and lists, as bulleted items, project titles that fund research in each technology.

R. L. Drake
Program Coordinator

CONTENTS

COAL, GAS AND OIL

FISSION AND FUSION

OIL SHALE

Coal, Gas and Oil

COAL, GAS AND OIL

- **Atmospheric Diffusion in Complex Terrain**
- **Atmospheric Boundary Layer Studies**
- **Pollutant Transformation in the Atmosphere**
- **Atmospheric Carbon Dioxide—An Archival Study of Spectroscopic Data**
- **Meterological Effects of Thermal Energy Releases (METER)**
- **Theoretical Studies and Applications**

As the use of fossil fuels as an energy source increases, so too will air pollutants, such as sulfur, nitrogen compounds and trace metals, produced by the combustion and conversion of these fuels. The analysis of the fate of these pollutants from source to receptor is most urgent so that the nation's energy plan can proceed efficiently and be environmentally sound.

The research activities at PNL are basically related to three interlaboratory programs: the ASCOT, METER and Carbon Dioxide Programs. The ASCOT Program is currently concerned with the Geysers Geothermal Site in California, but future studies will be centered at other complex terrain sites in the west. The METER Program is studying the effects of cooling tower and stack plumes on local climate and precipitation quality while the Carbon Dioxide Program concentrates on the effects of fossil fuel burning on global climate.

The field activities in the ASCOT Program include the micrometeorological description of drainage winds, radiation measurements, and tracer experiments. Field activities in the METER Program include air chemistry, meteorological and scavenging measurements in and around cooling tower and pollutant plumes from a very large coal-fired power plant.

Atmospheric Diffusion in Complex Terrain

Objectives of this study are:

Defining and developing a long-range technical plan for studying the transport and diffusion of pollutant particles and gases over complex landforms.

Reviewing past theoretical, numerical and field studies pertinent to diffusion in complex terrain.

Assisting a multilaboratory program studying diffusion in complex terrain by providing methodologies and techniques of analyses for transport and diffusion over a variety of complex landforms.

PROJECT ASCOT - PACIFIC NORTHWEST LABORATORY'S CONTRIBUTION TO ASCOT COOLING TOWER EXPERIMENTS AT THE GEYSERS GEOTHERMAL RESOURCE AREA, AUGUST 1981

M. M. Orgill, T. W. Horst, D. W. Glover, J. M. Thorp, R. I. Schreck, P. W. Nickola, J. S. Wetzel and R. N. Lee.

This report gives a general overview of Pacific Northwest Laboratory's (PNL's) contributions to DOE's Atmospheric Studies in Complex Terrain (ASCOT) Cooling Tower Field Study, August 10-26, 1981. This was the third relatively intensive, multilaboratory effort to collect field data on drainage winds and from tracer experiments in The Geysers-Calistoga Geothermal Resource Area (GCGRA). The purpose of these experiments was to acquire a data base to evaluate existing cooling tower plume rise and transport-dispersion models for improving techniques in predicting the impact of H_2S or other pollutant emissions from future geothermal power plants or other energy production facilities located in complex terrain. These experiments were conducted jointly by DOE's ASCOT program and the California Energy Commission.

The ASCOT experiment for August 1981 differed from the previous two ASCOT experiments in two ways. First, the 1981 nighttime experiments were located in the Big Sulphur Creek area of the GCGRA, which is on the west side of the Mayacmas Mountains; the previous experiments were on the east side. Second, the field effort this year was divided between nocturnal drainage flow experiments (principally at Big Sulphur Creek) and daytime limited mixing operations experiments which included the Big Sulphur, Anderson-Putah and Cobb valleys and adjoining ridges. This is in contrast to experiments of the last two years, which were conducted only at night.

PNL's field effort in August 1981 consisted of the following tasks:

- Assisting with the establishment and documentation of a perfluorocarbon tracer sampling network in the Big Sulphur Creek area.
- Assisting with the establishment of a radio-repeater communications network in the area of operations.
- Establishing single and dual time-lapse and still photographic baselines and conducting photographic monitoring of cooling tower plumes during nighttime and daytime experiments.
- Collecting temperature, humidity, pressure, and wind data up to about 1000 m with a Tethersonde®. The PNL Tethersonde was located at two different locations on different nights in the Big Sulphur Creek area during the nighttime experiments and at Sawmill Flats in Cobb Valley during the daytime experiments.
- Collecting data on the height and structure of temperature inversions and thermals by a monostatic acoustic sounder at Sawmill Flats.
- Taking rawinsonde ascents in the Sawmill Flats area in order to obtain upper-level data on temperature, humidity, and winds.
- Conducting aircraft sampling of trace metals, scattering extinction coefficient, ozone, hydrogen sulfide and sulfur hexafluoride upwind and downwind of the GCGRA.

The location of these various field operations is shown in Figure 1. PNL's Tethersonde was part of a five-Tethersonde network in the GCGRA, and PNL's acoustic

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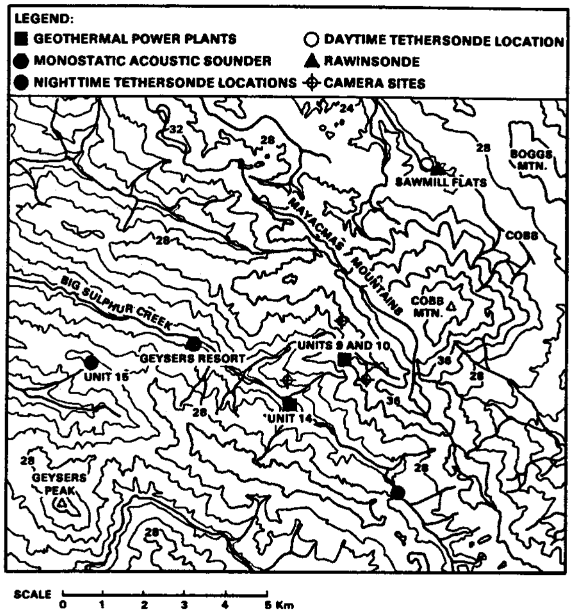

FIGURE 1. Topography of The Geysers Area and the Location of PNL Field Operations During The ASCOT Cooling Tower Experiments, August 10-26, 1981

sounder was part of a six-sounder network. A PNL rawinsonde provided the project with upper-level meteorological data. PNL also contributed to a three-aircraft sampling operation and our photography supplemented an airborne Lidar system in monitoring the cooling tower plumes. Table 1 lists the dates in which meteorological, tracer release and aircraft sampling operations were conducted in The Geysers area.

PNL's contributions to the ASCOT field effort through photography of cooling tower plumes, the establishment of a perfluorocarbon tracer network and aircraft sampling are presented in the following articles in this Annual Report, as well as a report on continuing studies of the ASCOT data base of 1979 and 1980.

TABLE 1. Dates of ASCOT Field Operations and Experiments in the GCGRA During August 1981

Experiment	Date	Time of Day	Operations
—	Aug. 12-13	Nighttime	Meteorology
1	Aug. 15-16	Nighttime	Tracer releases Meteorology Photography
2	Aug. 16-17	Nighttime	Tracer releases Meteorology Photography
—	Aug. 20	Daytime	Aircraft sampling (PNL)
—	Aug. 21	Daytime	Aircraft sampling (PNL)
3	Aug. 21-22	Nighttime	Tracer releases Meteorology Photography
4	Aug. 23	Daytime	Meteorology SF_6 tracer release Aircraft sampling Photography
5	Aug. 24	Daytime	Meteorology SF_6 tracer release Aircraft sampling Photography

POWER PLANT PLUME PHOTOGRAPHY AT THE GEYSERS GEOTHERMAL AREA--ASCOT 1981 EXPERIMENTS

J. M. Thorp and D. W. Glover

The Atmospheric Studies in Complex Terrain (ASCOT) cooling tower experiments were conducted in August 1981 at The Geysers, California geothermal area. To better predict the impact of H_2S emission from geothermal plants or gaseous emissions from cooling towers operating at energy production facilities, data is needed to evaluate existing models of cooling tower plume rise.

Five experiments were conducted: three at night during periods when valley drainage flows were established, and two during the afternoon during periods of limited vertical mixing. The Geysers Pacific Gas and Electric Co. (PG&E) plant, Unit 14, located near the bottom of the valley of Big Sulphur Creek, was chosen for the nighttime experiments. The afternoon experiments were conducted at Units 9 and 10, perched 1300 feet higher on a ridge spur.

Tasks

Perfluorocarbon or sulfur hexafluoride tracers were released into the steam plume for one hour at the beginning of each experiment. Our task was to provide a continuous photographic record of the visible plume behavior prior to, during, and following the tracer release into the steam plume. This record consisted of sequential, long exposure (300 second), black-and-white and color photographs for a period of several hours during each experiment. Additionally, during the daytime experiments, the plume from Units 9 and 10 was photographed with time-lapse 16-mm movies. The photographic record of the five experiments is summarized in Table 1.

Preliminary Results

A brief description of visual observation of plume variation and behavior during tests follows.

- Nighttime Experiments

Experiment 1, night of August 15-16--The plume appeared to rise vertically throughout the night. It resembled a lamp flame in shape, with the center portion rising much higher than the outer portions. Before midnight, detached puffs of steam sometimes reached our skyline, which was the high ridge south of Big Sulphur Creek. Plume height decreased with time during the night; the detached puffs were not observed after 0100 Pacific Daylight Time (PDT).

Experiment 2, night of August 16-17--All color photos between 2100 and 2337 PDT were lost due to a camera loading error. The black-and-white photos and subsequent color slides for this period show a short vertical plume throughout the experiment.

Experiment 3, night of August 21-22--The plume, though somewhat higher than observed during Experiment 2, was still quite low. Again, it was triangular in shape, but without the elongated apex observed in Experiment 1. After midnight some detached puffs reached and went above the southern skyline. There was no moon until after midnight; it was in the last quarter phase and did not illuminate the plume. A searchlight was used between 2300 and 0315 PDT to illuminate rising segments of the plume that otherwise we could not see by eye and which, no doubt, failed to register on the film except when the light was on them.

TABLE 1. Photographic Record of Five ASCOT Experiments Conducted in August 1981

Experiment No.	Date (1981)	Time Period	Photo Period (PDT)	No. Color Slides	No. B & W Slides	Movies (ft)
1	Aug. 15-16	Night	2100-0402	63	75	
2	Aug. 16-17	Night	2100-0530	69	94	
3	Aug. 21-22	Night	2100-0528	98	99	
4	Aug. 23	Day	1205-2025	97	97	~80
				93	90	~80
5	Aug. 24	Day	1200-2025	98	98	~90
				95	91	~75
			Total	613	644	

The Unit 14 plume was relatively unaffected by the drainage wind down Big Sulphur Creek in the three nighttime tests. This is probably because Unit 14 is located in a slight cove formed by a tributary of the creek.

Figure 1 is a photo of the steam plume from Unit 14, taken at night at an exposure of 300 seconds.

- Daytime Experiments

Experiment 4, afternoon of August 23--The plume was influenced by a light southwest wind throughout the experiment. At the start of the photographic record the plume had a high rise. Within an hour the plume grew shorter, with some detached puffs moving beyond the photo field. By 1400 PDT the plume was shorter yet and was contained in the photographs from Site 1. A shorter plume with detached puffs continued for the rest of the photo period until near sunset, when the plume became longer (ca. 2000 PDT).

Experiment 5, afternoon of August 24--Stronger wind from the southwest caused a lower-angle plume than with any other test. Blowdown and curl of the downwind side of the plume against the cooling tower was observed during much of the period. From Site 1, north of Units 9 and 10, it also appeared that the plume from the stacks at the west end of the cooling tower was subjected to somewhat higher wind speeds than the portion from the east stacks. The plume was quite low and short all afternoon, but showed some increase in length after 1900 PDT. It became more vertical near the end of the experiment, indicating a reduction in wind speed.

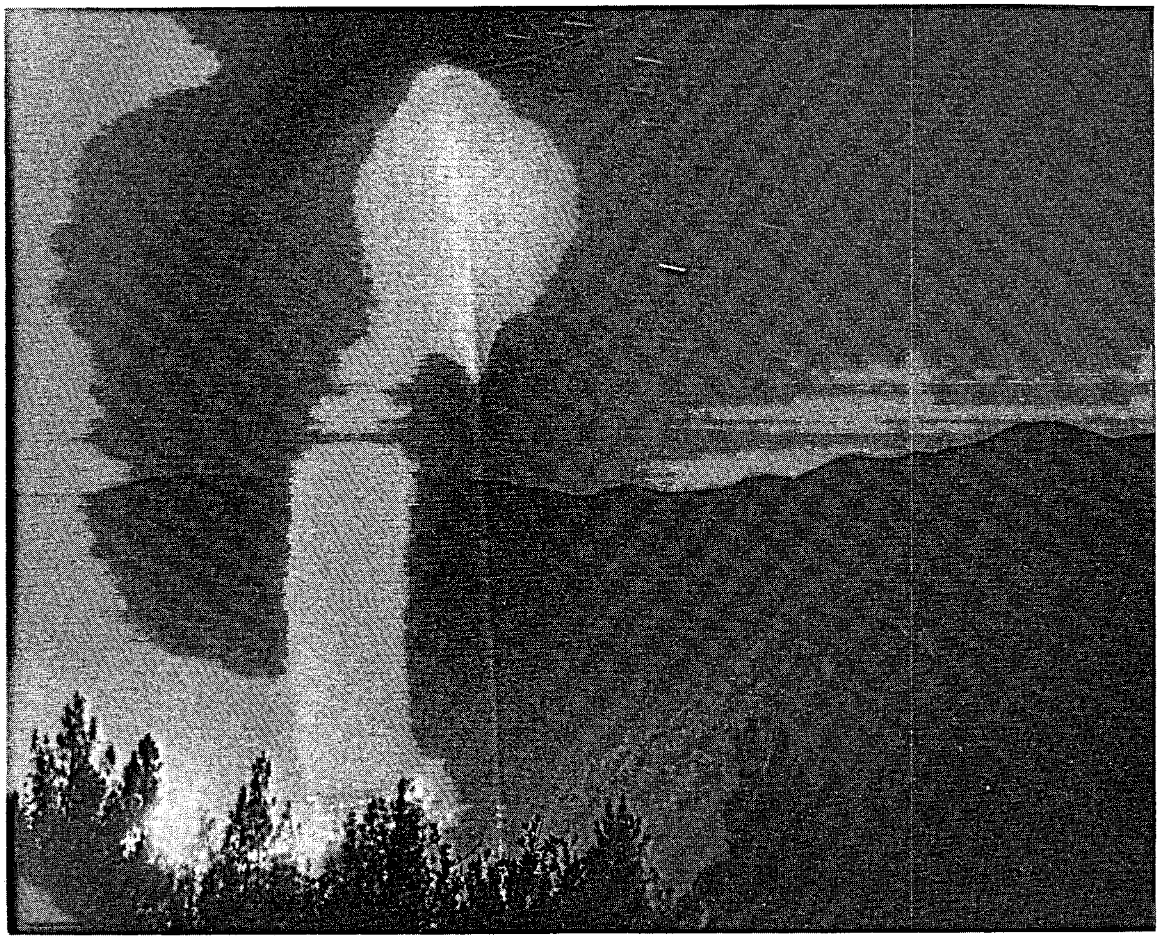

FIGURE 1. Steam Plume from Unit 14 Illuminated at Night by Moonlight and Searchlight

Future Work

Variation of plume rise with time during each of the experiments will be calculated by geometric analyses of the photographs.

SELECTION AND DOCUMENTATION OF TRACER SAMPLING LOCATIONS IN BIG SULPHUR CREEK VALLEY - PROJECT ASCOT, AUGUST 1981

P. W. Nickola

The 1979 and 1980 series of ASCOT field experiments in The Geysers geothermal area emphasized nocturnal transport and diffusion in the easterly downvalley drainage of Anderson Creek. An extensive array of ground-level tracer sampling locations was established in the Anderson Creek area in August 1980 (Nickola 1981). PNL was heavily involved in the selection and documentation of these sampling sites.

Big Sulphur Creek is a westerly flowing creek whose headwaters are within a few hundred meters of the headwaters of Anderson Creek. The Mayacmas Mountain Range divides the two watersheds. Since the ASCOT 1981 field program entailed perfluorocarbon tracer releases into cooling tower plumes in Big Sulphur Creek Valley, a new array of field sampling locations was necessary in that valley. Once again PNL was called upon to design a field sampling grid, to physically mark sampling locations in the field, and to document those locations.

Both the design and the in-field deployment of samplers in this complex terrain were compromises between ideal locations and practical locations. Departures from the ideal were dictated by such uncontrollable local factors as terrain, road access, legal access and vegetation. The 55-station network, as staked out in May 1981, is illustrated in Figure 1.

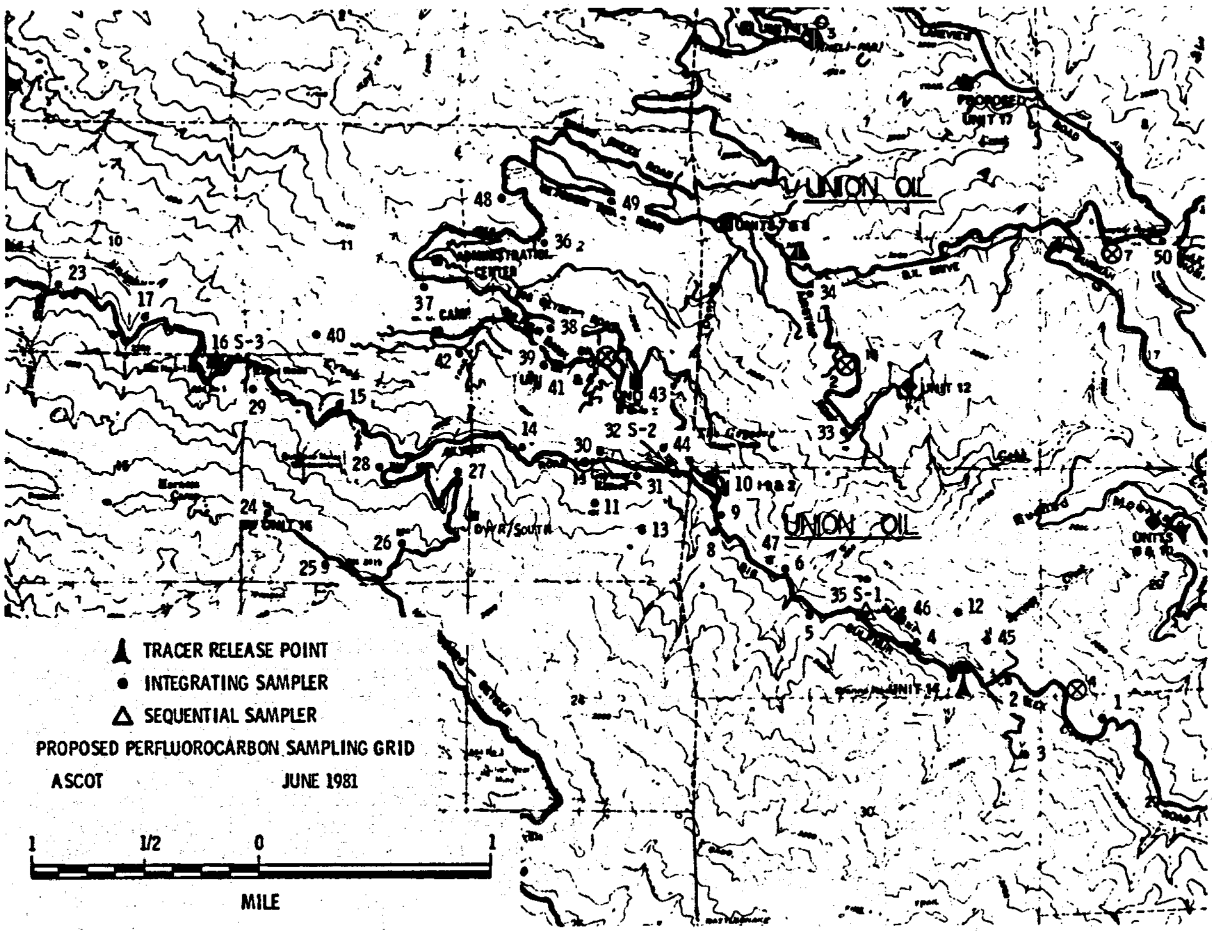

FIGURE 1. ASCOT 1981 Perfluorocarbon Release and Sampling Sites

During site selection, sampling site coordinates and elevations were documented as carefully as possible from a variety of topographic maps. These maps were obtained from the U.S. Geological Survey and from organizations involved in the industrial activity in the area.

Reference

Nickola, P. W. 1981. "Selection and Documentation of Tracer Releases and Sampling Locations - Project ASCOT, September 1980." In Pacific Northwest Laboratory Annual Report for 1980 to the DOE Assistant Secretary for Environment, Part 3, Atmospheric Sciences. PNL-3700, PT3, pp. 7-8, Pacific Northwest Laboratory, Richland, Washington.

AIRCRAFT SAMPLING OF SF_6, H_2S AND TRACE ELEMENTS DURING THE ASCOT FIELD OPERATIONS, AUGUST 1981

M. M. Orgill, R. N. Lee and R. V. Hannigan

This report briefly describes the results of the PNL aircraft sampling efforts during the ASCOT Cooling Tower experiments in August 1981. There were two purposes of these sampling flights: 1) to define the types and quantities of airborne materials that are being emitted from The Geysers geothermal operations and 2) to provide data on the locations, heights and concentrations of SF_6 tracer released from cooling towers in The Geysers area.

The aircraft sampling was conducted with the PNL Cessna 411 aircraft, which was equipped with the following measurement systems: a real-time SF_6 analyzer, a real-time sulfur analyzer, an ozone detector, an integrating nephelometer, and a high-volume filter sampler. Data from the tracer analyzer, sulfur analyzer, ozone detector, nephelometer, temperature-dewpoint probes, and a global navigation system (GNS-500A) were recorded on a DAS 32 Data Acquisition System. The aircraft sampling was supplemented with the operation of six ground-based, high-volume filter samplers located at sites generally west and east of The Geysers geothermal operations. Locations of the ground-based samplers and an example of the aircraft sampling flight patterns are shown in Figure 1.

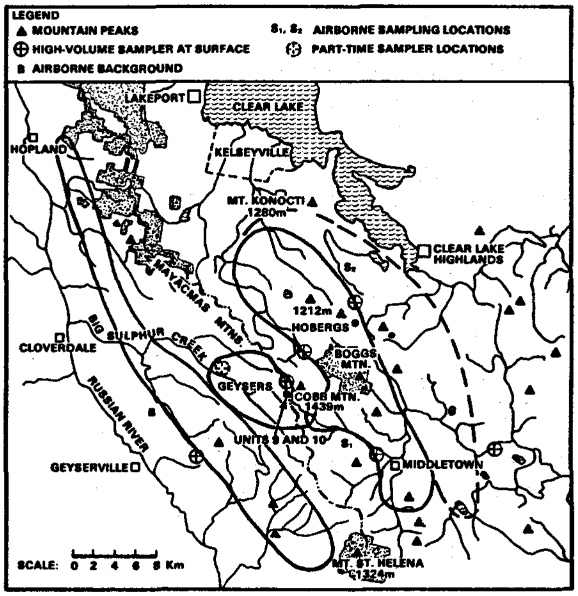

FIGURE 1. Generalized Topography of The Geysers-Calistoga Geothermal Resource Area (GCGRA), Location of The Geysers Geothermal Development Area, and Locations of Airborne Sampling Flights and Surface High-Volume Filter Samplers

The airborne sampling was conducted during the daytime hours of August 20, 21, 23 and 24. On August 20 and 21, sampling flights were conducted primarily for trace materials and H_2S. During the last two days, SF_6 tracers were released from Units 9 and 10 cooling towers, and airborne SF_6 sampling was included with the other sampling.

In general, the flight procedure was to obtain background measurements in the morning hours just west of The Geysers at approximately 1067 m and 1524 m mean sea level (MSL). In the afternoon, SF_6 tracer, H_2S and other sampling was conducted east of The Geysers between 1067 m MSL and 1829 m MSL. However, on the first sampling flight August 20, the procedure was reversed and background measurements were conducted in the afternoon. The ground-based, high-volume filters were operating during the seven hours of aircraft sampling.

The daytime sampling was conducted during light to moderate, westerly to southwesterly winds near the surface under sunny, clear skies. Hydrogen sulfide and SF_6 were detected to the northeast, east and southeast of The Geysers; H_2S was detected as far as 18 km from The Geysers. Preliminary concentration estimates for H_2S varied from 2.7 ppb to 41 ppb, with the highest concentration detected northwest (Sawmill Flats) and just east of Cobb Mountain. On August 20 and 21, H_2S was detected at 1829 m MSL at concentrations between 5 and 13 ppb. Sulfur hexafluoride concentrations were detected up to 1524 m

MSL at concentrations of approximately 200 ppt, and 230 to 240 ppt at 1067 m and 1280 m MSL.

The results presented above were based on a preliminary analysis of the strip chart record for H_2S and SF_6. A more detailed analysis of these data and the other data will modify and amplify these very tentative results.

DATA ANALYSIS OF THE JULY 1979 AND SEPTEMBER 1980 ASCOT DATA BASE

M. M. Orgill

During July 1979, September 1980 and August 1981, DOE sponsored multilaboratory field programs in The Geysers-Calistoga Geothermal Resource Area (GCGRA) to study the development and characteristics of nocturnal katabatic (drainage) winds and their effects on atmospheric transport and diffusion. The purpose of this report is to briefly discuss some of the analysis that is being performed on the July 1979 and September 1980 data base (also see reports by T. W. Horst and J. C. Doran in this Annual Report).

Analysis of July 1979 Data Base

Part of the July 1979 ASCOT data base has been utilized to provide initial analysis of the problem of how larger-scale forcing mechanisms (synoptic and mesoscale) may affect the development and persistence of nocturnal katabatic flow in The Geysers area.

The principal data bases utilized to date have been the ASCOT and Pacific Gas and Electric Co. wind and temperature data from 10-m towers, PNL 30-m tower data at Diamond D, and PNL and Wave Propagation Laboratory (WPL) Tethersonde® data. In addition to these data, surface (sea level) pressure data were obtained from the National Climatic Center (NCC) to assist in examining synoptic variations throughout the experimental period.

The preliminary analysis of these data have identified some possible synoptic to local-scale interactions that could have an important influence on the development and persistence of nocturnal drainage winds in The Geysers area. Some of these interactions are briefly summarized below.

- Pacific Coast Marine Air Invasion

The mean sea-level pressure differential between the coast and inland areas and the observed winds at The Geysers undergo long-term (several-day) and diurnal oscillations. Surges of modified marine air invade The Geysers area every afternoon. Although the westerly surges of modified marine air normally decrease in the evening, at times they may persist through the evening. In these cases, the resultant wind has a pronounced effect on plume trajectories and dispersion (Orgill, Schreck and Nickola 1980).

- Warming Events

Temperature data from several 10-m tower locations and the two Tethersondes have revealed the occurrence of warming periods ($\Delta T \sim 0^\circ$ to $2.0^\circ C$) during the normal nocturnal cooling period. These warming periods decrease the nighttime cooling rates and may interfere with the development of strong drainage winds.

- Elevated Temperature Inversions

An elevated temperature inversion was usually observed at or below the height of the Mayacmas Ridge. Vertical oscillations of the inversion have been observed over the valley and correlate with some of the warming events on the valley slopes.

A summary of these results, entitled "Synoptically Driven Down-Slope Winds and Their Effects on Local Nocturnal Drainage Airflow in the Geysers Geothermal Resource Area," was presented at the Second Conference on Mountain Meteorology, November 9-12, 1981, in Steamboat Springs, Colorado, by M. M. Orgill, R. I. Schreck and C. D. Whiteman.

Analysis of September 1980 Data Base

During the ASCOT field program in September 1980, seven Tethersondes were used to obtain wind direction and speed, temperature, humidity and pressure data up to approximately 1000 m above the surface (Schreck 1981). The PNL ASCOT group was selected by Lawrence Livermore Laboratory (LLL) to edit and plot this Tethersonde data base. In addition, PNL was requested to provide a brief narrative and statistical summary of the data for each evening. The parameters that were considered for this summary were approximate average and range in 1) depth of westerlies (including drainage winds), 2) maximum speed in the

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first 100 m, 3) maximum speed above 100 m, 4) mixing ratio, 5) relative humidity, 6) depth of the surface temperature inversions, 7) surface temperature, and 8) surface nocturnal cooling rate.

On the basis of the Tethersonde data and the narrative and statistical summaries, some tentative conclusions can be made regarding the effects of synoptic mechanisms in the development and persistence of drainage winds.

- Marine Air Invasion and Other Synoptic Disturbances

Surges of modified marine air invaded The Geysers area on some afternoons, but were usually replaced by descending easterly winds during the late evening or early morning hours. A synoptic disturbance September 17-19 postponed experiments on the night of September 18 and terminated experiments early the following night.

- Descending Easterly Winds

Descending easterly winds over The Geysers area were a problem on four evenings. On two of these evenings (September 21-22 and September 22-23) the easterly winds invaded the lower slope of Anderson Creek and terminated operations early in the evening. The origin of these easterly winds has not been determined.

- Warming Events

Near-surface temperature data from the Tethersondes indicated the occurrence of warming periods during the normal nocturnal cooling period. Some of these warming periods appeared to be associated with descending easterly winds.

Further analysis of the July 1979 and September 1980 ASCOT data base will proceed and be reported in the literature.

References

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• Atmospheric Boundary Layer Studies

Objectives of these studies are:

Investigating the meteorological characteristics of the planetary boundary layer that pertain to pollutant transport and dry removal.

Analyzing the transport and diffusion of pollutants in complex terrain, particularly in the Geysers Geothermal Area of California.

Constructing simple models for the prediction of the depth, speed and direction of gravity-controlled drainage flows and pollutant dispersion within these flows.

COMPARISONS OF MODELS AND OBSERVATIONS OF NOCTURNAL SLOPE WINDS

J. C. Doran and T. W. Horst

The systematic investigation of the behavior of nocturnal slope winds is complicated by a large number of factors that may influence their behavior. Comparisons of models and measurements may be difficult since data are rarely collected from an ideal site or in circumstances that conform to the assumed boundary conditions of the model.

In order to address the problem, we have compared some observations of nocturnal slope winds with the predictions of two models. The observations were made at a topographically simple location, Rattlesnake Mountain, near Richland, Washington, and at a more complex site in The Geysers area, California. The first model is a one-dimensional hydraulic approach proposed by Manins and Sawford (1979), and the second is a two-dimensional, modified K-theory approach. Emphasis is placed on the measurements taken at Rattlesnake Mountain.

The sites and instrumentation used for the measurements are described elsewhere (Horst and Doran 1981). For the present paper it is sufficient to summarize these descriptions by noting that the Rattlesnake Mountain site is a nearly ideal, relatively smooth, two-dimensional tilted plain. The Geysers site is in considerably more complex terrain, covered by an extensive canopy of trees and shrubs. The data from these two sites allow sensitive tests of the models to be made. In particular, the simplicity of the Rattlesnake Mountain site should permit the testing of many of the basic assumptions of one- and two-dimensional models, while the relative complexity of The Geysers area provides a challenge to the flexibility and generality of such models.

One-Dimensional Model

The structure of the one-dimensional model and a comparison of the model to actual observations are discussed below.

- Model Structure

This model is essentially the hydraulic one proposed by Manins and Sawford (1979). The basic equations for momentum (parallel to a tilted plane), thermal energy and continuity are integrated, in a direction normal to the slope, to a height H sufficiently far from the surface that the measured flow is unaffected by katabatic winds. Scales of velocity U, thickness h and buoyancy deficit Δ are defined by

$$Uh = \int_0^H u \, dn \qquad (1)$$

$$U^2 h = \int_0^H u^2 \, dn \qquad (2)$$

$$U\Delta h = \int_0^H \frac{u g d}{\theta_r} \, dn \qquad (3)$$

where u = the component of velocity parallel to the slope
n = the normal coordinate
g = the acceleration of gravity
θ_r = the ambient potential temperature
d = the deficit of potential temperature in the katabatic flow.

Analytic solutions for U, Δ and h were derived for conditions in which the adiabatic warming due to downslope advection may be neglected. When this was not the case, numerical solutions were obtained. We have extended the work of Manins and Sawford to take into account the possibility of changes in slope angle as well.

- Comparison of Model Predictions with Measurements

Values of U, Δ, and h were obtained from measurements made on Rattlesnake Mountain on July 1, 1980, and at The Geysers on September 16-17, 1980. Results from Rattlesnake Mountain (Tower B) and The Geysers are shown in Table 1. The table also contains the results of the model calculations. It is evident that there are a number of differences between the observed and modeled scales.

In Table 1, the values of U, Δ and h in the first row were obtained by assuming that the ambient wind velocity was zero and extrapolating the velocity profile found on the tower to this value. The results in the second row were obtained by assuming the ambient velocity was $\sim$2 m/s. It is surprisingly difficult to determine which is the more appropriate value.

The model calculations also cause some difficulties since there are a number of parameters that must be estimated which affect the derived values. The results shown in Row 3 were chosen as a reasonable compromise to a "best" fit. In the model, U is underestimated, h is of the same order as the experimental results, and Δ shows especially poor agreement.

The ambient winds during the measurement period at The Geysers were quite light, and the calculated and theoretical velocity and thickness scales (Rows 4 and 5) agree much better than those for the Rattlesnake Mountain data. However, the buoyancy deficit again shows very poor agreement. Once again, parameters have been selected to produce a "best" fit. The assumed heat flux and entrainment are the same as those used in the Rattlesnake Mountain calculations, but the drag coefficient is ten times higher to account for the presence of dense vegetation at The Geysers site.

TABLE 1. Scale Parameters U, h and Δ Determined from Measurements and the Hydraulic Model of Manins and Sawford

	Scale of Velocity, U (m/s)	Thickness, h (m)	Buoyancy Deficit, Δ (m/s^2)
Rattlesnake Mtn., Tower B			
Measured			
$U_{amb} = 0$	2.1	18.0	0.046
$U_{amb} = 2$ m/s	2.7	7.8	0.08
Model	0.9	14.0	0.01
The Geysers			
Measured	1.0	30.0	0.07
Model	1.0	27.0	0.024

It is clear that the failure of the hydraulic model to account for the effects of ambient winds is a serious difficulty. In the presence of such winds, the height H to which the intergrations (1), (2) and (3) must be carried out is ambiguous. Moreover, it is not evident how to separate the katabatic flow from the ambient flow.

Despite the problems with this model, it does retain some attractive properties. One is the ability to provide useful, working estimates for the depths in which the velocity maximum and most of the temperature changes occur. We may assume that h is a measure of this depth. The model predicts that h varies linearly with slope length; for the cases listed in Table 1, the proportionality constant was estimated to be 0.033.

The predicted values of h at Towers A and B are 6.6 m and 14 m, respectively. These depths seem to encompass essentially all the temperature changes associated with the katabatic layer; in the case of Tower B, where velocity data were available, the jet is also well defined within this distance.

Two-Dimensional Model

The two-dimensional model structure is given below, as well as a comparison of this model to actual measurements.

- Model Structure

This model is an adaptation of a number of earlier ones, beginning with Delage's (1974) and used in various forms by a number of other investigators (Nieuwstadt and Driedonks 1979; Rao and Snodgrass 1981). It is a gradient diffusion approach in which the K parameters are determined by the local turbulent kinetic energy, E,

$$K = \ell(cE)^{1/2} \qquad (4)$$

where ℓ is a mixing length and the proportionality constant c = 0.16.

The governing equations for the model specify the behavior of the velocity components U and V, potential temperature θ and turbulent kinetic energy E; U is the wind component parallel to the slope in the downslope direction and V is perpendicular to U and also parallel to the slope. The initial conditions are given by specifying values of the geostrophic wind components U_G and V_G obtained at some upper level (~1 km). The ambient lapse rate is also specified. Advection terms are discarded, and the equations for U, V and E are integrated until an approximate steady state is obtained. The steady-state solutions are then used as a starting point for further computations with the full set of equations.

- Comparison of Model Predictions with Measurements

Figure 1 shows the calculated development of the temperature velocity component for slope distances corresponding to the positions of Towers A and B. Although there are qualitative similarities between the observed and calculated behavior, there are several differences. For example, the actual inversion of Tower A is more sharply defined than in the model. At Tower B, the observed inversion also appears to be a bit sharper, although the discrepancy is not as large. At both sites, the model results show smaller temperature changes between ~1 m and the highest measurement positions on the towers than was observed. The height of the velocity maximum is also greater in the model than in the observations, although this may be partly attributed to the somewhat coarser spacing of the instruments relative to the computational grid. The calculated peak velocities are also slightly less than the observed ones.

The results are generally encouraging. They show the development of the drainage flows with slope distance and reproduce the principal observed features reasonably well. Additional measurements carried out during the summer of 1981 will provide a basis for further development and comparisons.

Conclusions

Both the one- and two-dimensional models discussed here have applications in the study of

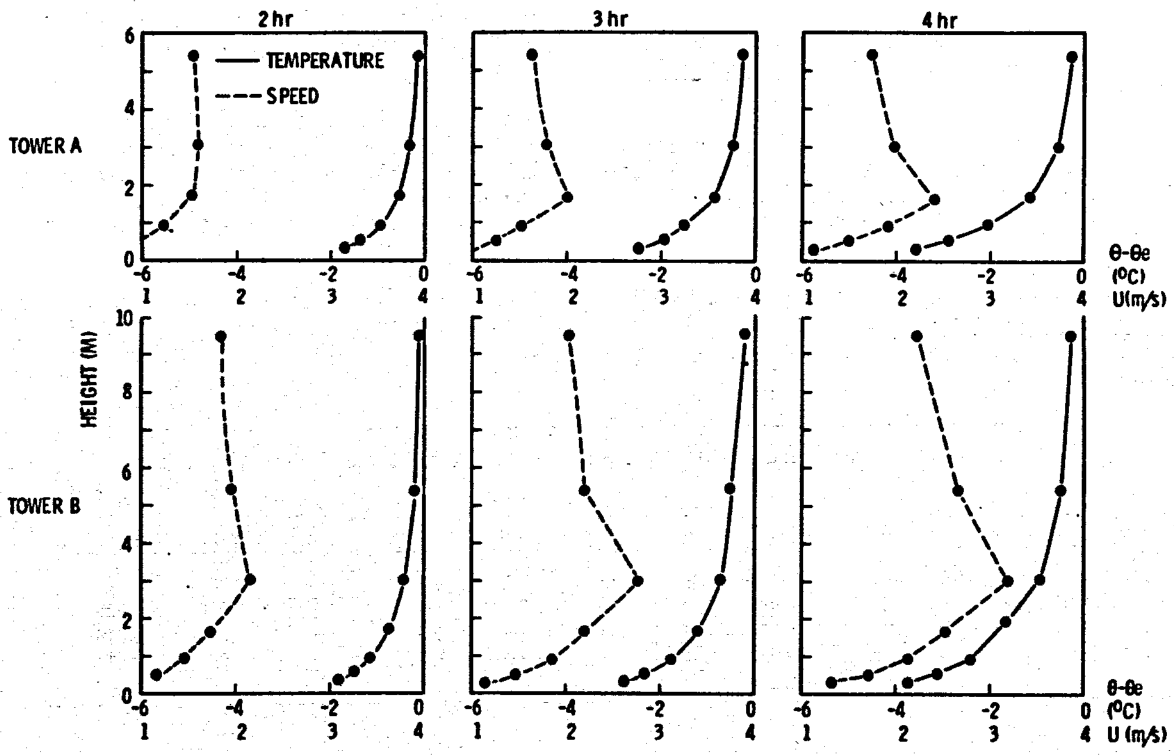

FIGURE 1. Calculated Temperature and Velocity Profiles for Rattlesnake Mountain. Times indicated are after onset of surface cooling at a rate of 1.4°C/hr.

drainage winds. The hydraulic approach suffers from an inability to incorporate the effects of ambient winds and an apparent lack of similarity between atmospheric behavior and the tank experiments on which it is based. Nevertheless, it provides useful estimates for the depths to which significant temperature inversions are observed and in which velocity maxima will be found. The two-dimensional model is more complicated to apply, but is more flexible and provides a more accurate description of the observations. Its relatively good success, without the necessity of incorporating a large number of undetermined parameters, suggests that it adequately describes the principal physical mechanisms reponsible for drainage winds. It therefore serves as a basis for the study of additional, complicating factors such as cloud cover and canopy effects.

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OBSERVATIONS OF NOCTURNAL SLOPE WINDS

T. W. Horst, J. C. Doran and O. B. Abbey

Nocturnal drainage winds begin when air adjacent to an inclined surface flows down the slope because it is cooled more than the free air at some distance from the surface. These slope winds commonly merge and are channeled by the topography to form larger-scale drainage or mountain winds. This report describes observations of these nocturnal slope flows at two quite different sites.

The two sites are located on Rattlesnake Mountain in eastern Washington and on Cobb Mountain in northern California. The Rattlesnake site approximates a simple tilted plane covered with bunchgrass and scattered sagebrush. Wind and temperature profiles are being measured with three towers located at different distances down the slope (Table 1). The Cobb site is a broad, bowl-shaped valley with several convergent drainage systems and with vegetation alternating between areas of tall pine trees and low brush. Wind and temperature measurements were made at eight levels on a single 60-m tower located amid 10- to 15-m trees. The two sites are described in detail by Horst, Doran and Abbey (1981). At both sites, tower measurements were supplemented by regular Tethersonde® profiles of wind and temperature to 300-500 m above the surface.

Data were collected at the Rattlesnake site on favorable nights during the summers of 1980 and 1981, and on Cobb Mountain during the ASCOT Geysers Field Program, September 11-25, 1980. At this time the Cobb data

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TABLE 1. Comparison of Slope Flow Depth Z to Slope Length S and Vertical Drop ΔZ

Site	ΔZ (m)	S (m)	Z_T (m)	Z_u (m)	Z_s (m)	Z_u/Z_T	Z_s/Z_T	$Z_T/\Delta Z$	Z_T/S
Cobb Mountain	670	2590	45	23	32	0.5	0.67	0.067	0.017
Rattlesnake A	71	198	4	—	—	—	—	0.056	0.020
B	153	427	7	2	—	0.3	—	0.046	0.016
C	229	898	10	3.5	—	0.35	—	0.044	0.011

have been analyzed more thoroughly than the Rattlesnake data, and highlights of that analysis are presented below. A more detailed discussion can be found in work by Horst and Doran (1981).

The observations made at Cobb Mountain suggest three indicators of slope flow. Two of these may be seen in Figure 1: a surface-based temperature inversion and a low-level maximum in the down-slope wind. The third and best indicator of drainage flow occurrence, however, is wind direction. During the best slope flows, the wind direction near the center of the katabatic layer is steadily downslope.

Using wind direction to classify the nocturnal winds on Cobb Mountain, good slope flows were observed when the wind velocity at ridge level U_O was less than 6 m/s and there was an inversion ΔT of at least 5°C above 5 m. These parameters were combined with temperature T, gravitational acceleration g, and depth of the inversion Z_T to form a bulk Richardson's number

$$Ri_B = \frac{\Delta T}{U_o^2} \frac{g}{T} Z_T.$$

Most of the good drainage wind occurs when $Ri_B > 0.7$; good drainage wind never occurs when $Ri_B < 0.2$ and no drainage wind occurs when $Ri_B \leq 0.15$.

Although the occurrence of slope flow is a function of the ambient wind speed, its structure depends only weakly on the ambient wind because the stable thermal stratification decouples the katabatic flow from that above.

The Cobb Mountain data have also been used to investigate the development of slope flows. The surface cools rapidly after local sunset, but establishment of fully-developed slope flow is delayed until the surface-based inversion is built. Once established, the katabatic layer cools at a less rapid rate and the strength of the inversion increases more slowly for the remainder of the night. During this time, the depth of turbulent mixing, the wind speed maximum, and the "thickness" of the nocturnal jet decrease. Disruption of the slope flow is nearly coincident with local sunrise.

The depth of the slope flow can be determined by using three parameters: depth Z_T of the surface-based inversion, height Z_u of the wind maximum, and depth Z_s of turbulent

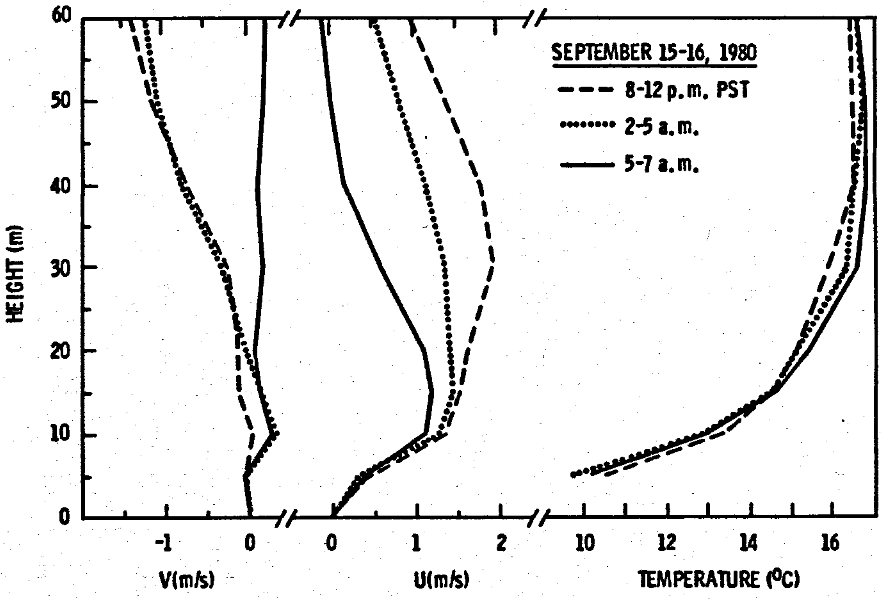

FIGURE 1. Cross-Slope Wind V, Down-Slope Wind U, and Temperature Profiles from Tower on Cobb Mountain

mixing as recorded by an acoustic sounder at Cobb Mountain. Table 1 lists these parameters for both observation sites and also compares Z_T to the vertical drop from the top of the slope ΔZ and to the slope length S. The Rattlesnake data, particularly $Z_T/\Delta Z$, support the supposition that the katabatic layer grows with distance down the slope.

Calculations have also been made of turbulent energy as a function of height at Cobb Mountain. The most notable feature is that the turbulence in the center of the katabatic layer is only 20% of that found above the layer. This is due to the small shear and correspondingly large stability near the wind maximum. Also, in this region the turbulent energies in the down-slope and cross-slope wind components are equal. The turbulent energy within the canopy is comparable to that near Z_u, but is contained almost entirely in the down-slope component.

Reduction of the Rattlesnake data and a thorough analysis of data from both sites will be completed in FY 1982.

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• Pollutant Transformation in the Atmosphere

Objectives of this study are:

Measuring chemical transformation rates of polycyclic aromatic hydrocarbons and other organic compounds emitted by fossil-fueled power plants.

Correlating transformation rates with distance from the source, ambient temperature, light intensity and concentrations of sulfur dioxide, nitrogen oxides, and ozone.

Evaluating the results of laboratory studies on discrete reaction steps in the overall transformation processes.

TRANSFORMATION OF ENERGY-RELATED POLLUTANTS

D. R. Kalkwarf

The purpose of this study is to measure the chemical transformation rates of polycyclic aromatic hydrocarbons (PAH) and other undesirable organic compounds emitted by fuel-fired power plants as these compounds are transported through the atmosphere. This information is needed to more accurately assess the potential environmental impact of such plants. Research this year focused on characterizing the products and rates of reactions between NO_2 gas and PAH deposited on coal fly ash. Laboratory studies showed that several representative PAH were converted to more-mutagenic nitro-derivatives at rates that were proportional to the first power of the NO_2 concentration. The initial products were identical to those obtained by nitration of PAH with NO_2^+ ions in liquid solution, but a comparison of the relative rates of nitration indicated that a different mechanism was involved in the gas-solid reactions. Extrapolation of the rate data to conditions in stack plumes indicated that reactions of NO_2 with PAH would only contribute significantly to the overall rate of PAH transformation at distances within 1 km of the stack.

Concern about reactions between NO_2 and PAH stems from recent reports on the mutagenicities of their possible products. The compound 1-nitropyrene was reported to be mutagenic, in contrast with non-mutagenic pyrene; and 1,8-dinitropyrene was found to be one of the most mutagenic compounds ever tested in assays with Salmonella bacteria (Rosenkranz et al. 1980). Similarly, 4-nitroperylene, 1-, 3- and 6- nitrobenzo[a]pyrene were reported to be more mutagenic than their parent PAH and were shown to be formed on PAH-coated glass filters exposed to gas mixtures containing NO_2 (Pitts et al. 1978).

In the present work, the products and rates of reaction between NO_2 gas and dry surfaces of pyrene, perylene and benzo[a]pyrene were examined. These PAH were supported either on Pyrex® glass or on coal fly ash collected with an electrostatic precipitator. Glass-supported PAH films only a few molecules thick were prepared by evaporating solutions of PAH in organic solvents so that the resultant film uniformly coated the interior walls of round-bottom flasks. Fly-ash-supported PAH films were prepared from fly-ash samples which had been coated previously with the equivalent of a monomolecular layer of PAH. Aqueous slurries of these samples were then coated on the interior walls of round-bottom flasks and dried to form adherent films of PAH-coated ash. A reaction was initiated by introducing a measured amount of purified NO_2 gas into a coated flask held at 25°C in a water bath. The flask was then sealed with Teflon®, and the NO_2-air mixture inside was stirred with a Teflon paddle so that diffusion would not limit the rate of NO_2 reaction with the PAH surface. After a measured time period, the reaction was stopped rapidly by drawing pure air through the flask, and the reaction products were analyzed.

Reaction products were identified by comparison of their chromatographic retention times to those of pure samples of possible products synthesized by procedures described in the literature. Both gas chromatography, using a siloxane-bonded quartz capillary column,* and high-performance liquid chromatography, using a mobile phase of 30:70 water:methanol

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*Durabond DB-5, manufactured by J W Scientific, Inc.

on a reverse-phase column,* were utilized, although the latter was found to be more versatile. The initial products formed with the three PAH were 1- nitropyrene, 4-nitroperylene and 6-nitrobenzo[a]pyrene. These compounds are also the main products formed by the nitration of the parent PAH with NO_2^+ ions in liquid solutions (Dewar, Mole and Warford 1956).

The reaction rates were determined by measuring the residual amount of PAH as a function of time. In a six-fold molar excess of NO_2, this residue decreased exponentially with time, as illustrated by the examples shown in Figure 1. Both glass-supported and fly-ash-supported PAH showed this behavior, indicating that the rates were proportional to the amount of PAH remaining. The use of reaction vessels of different volumes yielded rates proportional to the concentration of NO_2. A general rate equation was formulated as:

$$\frac{d(PAH)}{dt} = -kA(PAH)[NO_2]$$

where k = the rate constant in $\ell m^{-2}s^{-1}$
A = the surface area of PAH in m^2/mole
(PAH) = the residual moles of PAH
$[NO_2]$ = the residual concentration of NO_2 in moles/ℓ.

For reactions on glass, the value of A was the internal surface area of the flask, whereas for reactions on fly ash, A was calculated from the measured specific surface area of the fly ash and the amount of PAH coating it. Table 1 lists values of k that were evaluated when $[NO_2]$ was 2×10^{-6} to 3×10^{-5} M (40 to 700 ppm).

The rate constants for reactions on fly ash were found to be only about one-tenth as large as those on glass, suggesting that the PAH are inherently less reactive on fly ash. Also, the relative rates of these heterogeneous reactions, k(pyrene):k(benzo[a]pyrene):k(perylene) = 2:1:1, are quite different from the corresponding rates for reactions in solution, 0.2:1:0.7 (Dewar, Mole and Warford 1956), indicating that the reaction mechanisms are different. Since the dinitropyrenes were reported to be particularly mutagenic, the nitration rate of 1-nitropyrene was also measured and found to be only 7×10^{-4} ℓ $m^{-2}s^{-1}$ on glass at 25°C.

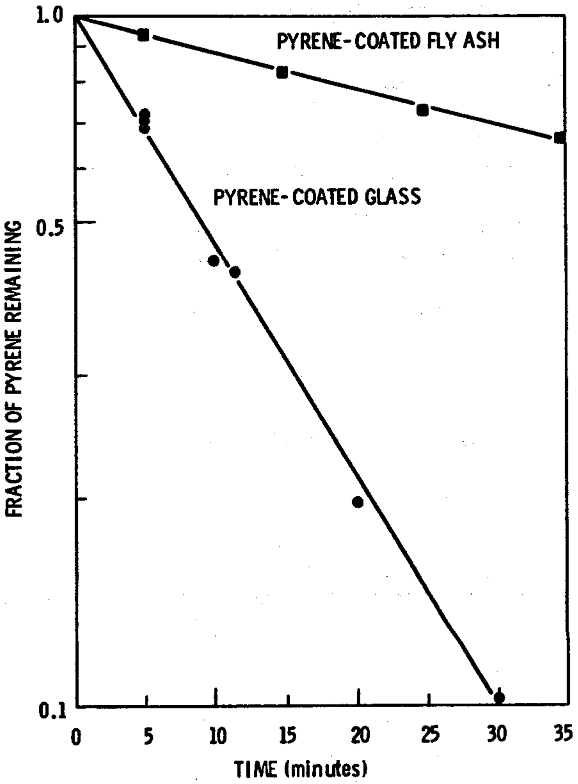

FIGURE 1. Residual Pyrene on Glass and Fly-Ash Supports After Exposure to 60 ppm NO_2 Gas for Various Periods of Time

TABLE 1. Rate Constants for Reactions of NO_2 with PAH Supported on Glass and Fly Ash

PAH	Support	k (25°C), ℓ $m^{-2}s^{-1}$
Pyrene	Glass	4×10^{-3}
	Fly ash	2×10^{-4}
Benzo[a]pyrene	Glass	3×10^{-3}
	Fly ash	1×10^{-4}
Perylene	Glass	6×10^{-4}
	Fly ash	1×10^{-4}

The half-times for PAH nitration in a hypothetical but representative stack plume were calculated and compared with the overall PAH transformation rates at the Colstrip, Montana power plant. Calculations were based on rate constants listed in Table 1 and on

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*Supelcosil LC-PAH, manufactured by Supelco, Inc.

reports that the NO_2 concentration was 500 ppm (2×10^{-5} M) in the flue of a large power plant (Hobbs, Hegg, Eltgrowth et al. 1979) but only a few hundred ppb ($\sim 2 \times 10^{-8}$ M) at 2 km downwind (Hegg and Hobbs 1979). Assuming that these values represent steady-state concentrations of NO_2 and that any PAH exist in the plume as monomolecular layers on fly ash ($A = 10^5$ m^2/mole), the reaction half-times were calculated from the equation $\tau = 0.693\ kA[NO_2]$ and are listed in Table 2.

Data previously reported on samples of the Colstrip stack plume indicated transformation half-distances of 8 and 12 km for pyrene and benzo[a]pyrene, respectively (Kalkwarf and Garcia 1979). Since those samples were collected when the wind velocity was 2 km/hr, the corresponding transformation half-times were 4 to 6 hr. Comparisons of these values with those listed in Table 2 indicate that reaction with NO_2 is not likely to contribute noticeably to the overall transformation rates of PAH in the plume beyond the first km from the stack. However, within that first km, nitration could be a significant mechanism for PAH transformation.

TABLE 2. Estimated Half-Times for Reactions of PAH with NO_2 in a Stack Plume

PAH	NO_2 (ppm)	Half-Time (hr)
Pyrene	500	0.5
Benzo[a]pyrene	500	1
Perylene	500	1
Pyrene	0.5	500
Benzo[a]pyrene	0.5	1000
Perylene	0.5	1000

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• Atmospheric Carbon Dioxide Abundance—An Archival Study of Spectroscopic Data

Objectives of this study are:

Determining the current and past abundance of the carbon dioxide in the earth's atmosphere by spectroscopic means.

Providing independent corroboration of the CO_2 increase that has been observed by other in-situ techniques.

Extending the knowledge of the total atmospheric CO_2 burden back to a time for which no information now exists.

ATMOSPHERIC CO_2 ABUNDANCE - AN ARCHIVAL STUDY OF SPECTROSCOPIC DATA

G. M. Stokes

Abstract

Progress in a program to determine the atmospheric abundance of carbon dioxide (CO_2) is described. Efforts are directed toward the measurement of the abundance of CO_2 back to the 1890s, using absorption spectra of the sun as the source of data.

Introduction

The relationship between the atmospheric abundance of carbon dioxide (CO_2) and various anthropogenic factors, most notably fossil fuel burning, has become a matter of great concern. Carbon dioxide has a significant effect on the average temperature of the earth's surface and lower atmospheric layers. This is primarily a result of the gas's large infrared opacity, which produces a "greenhouse effect." Because of the importance of CO_2 to the energy budget of the atmosphere, changes in its abundance could have a major effect on the earth's climate.

In order to understand how man's consumption of fossil fuels could affect climate through altering the atmospheric CO_2 abundance, it is necessary to understand the factors that control its atmospheric abundance. The atmosphere represents a relatively small reservoir of carbon compared to other reservoirs such as the deep oceans or sedimentary deposits. As a result, relatively modest changes in these much larger reservoirs can have an overwhelming effect on the amount of CO_2 in the atmosphere. It is therefore not surprising that there continues to be intense debate as to which processes affecting which sources and sinks of carbon control the atmospheric CO_2 abundance.

This interest in changes in atmospheric CO_2 abundance has led to a critical evaluation of the existing data that may relate to the history of that abundance. As a result, an attempt is being made to extract data accumulated since the 1890s giving the abundance of CO_2 from spectra of the sun.

The mechanism by which sun spectra yield CO_2 abundance data follows. As sunlight passes through the earth's atmosphere, the molecules in the atmosphere absorb and scatter the radiation. A substantial fraction of this absorption is caused by the excitation of various energy levels in atmospheric molecules. Absorption of this kind occurs at specific wavelengths and results in what are called absorption lines. The strength of these lines is primarily a function of the properties of the molecule and the number of molecules between the observer and the sun. It should therefore be possible to determine the amount of CO_2 in the atmosphere from spectra of the sun. Such spectroscopic data exist for a period extending back to at least the mid-1890s, when Samuel P. Langley began observing and recording the spectrum of the sun with a prism spectrobolograph.

Phases in the Determination of Atmospheric CO_2 Abundance

There are several important phases to the determination of the amount of CO_2 in the atmosphere from sun spectra data.

First, the existence of old spectral data needs to be established and the actual data recovered from storage. This process may be as simple as a review of published literature or as difficult as searching the archives of major observatories. For example, one collection of data that will be analyzed in the current program was recovered from a cellar at a remote observing site, where it had been stored and forgotten for more than 50 years.

Second, once a data set has been located, its potential value to the program needs to be assessed. While the utility of a particular data set may not actually be known until a final CO_2 abundance is determined from the data, there are preliminary analyses which can be used to determine if further analysis is warranted. In the current program our approach has been to develop a reduction technique for each collection of spectral data as a basis for assessing utility of the data. The development of this preliminary reduction plan has the important advantage of forcing an evaluation of the complete data reduction process of a particular data set as early as possible. For example, much of the data cannot be analyzed without supplementary data such as molecular constants or special information relating to the circumstances of the observations. The preliminary analysis plan identifies the need for supplementary data and either the existence of the required data or the possibility of obtaining new data to meet program requirements.

The final phase of the program is the actual analysis and intercomparison of the individual data sets to determine the changes in the amount of atmospheric CO_2. We will now describe the current status of the program in terms of the three phases of archival investigation, preliminary reduction plan and analysis.

Current Status of the Study of Atmospheric CO_2 Abundance

We have been disappointed by our failure to locate additional data from the critical period from 1895 to 1920. We have looked for spectrobolograms taken during expeditions to Mt. Wilson by members of the staff of the Smithsonian Astrophysical Observatory, but this search, conducted at the plate archives of the Mt. Wilson Observatory, yielded nothing. In the course of the search, reports were received of the existence of copies of plates taken at Mt. Montezuma, Chile, during the Smithsonian Solar Constant Program. These copies could double the existing data base for the period from 1920 to 1950. In September 1981, at least some of the copies were discovered in the archives of the Smithsonian Institution. This coming year we will determine the extent of these copies and their suitability for analysis. We also expect to expand our search for further data to European observatories.

The development of reduction methods for the individual data sets has occupied most of the program resources, with particular emphasis placed on the Smithsonian spectrobolograms. This data set consists of approximately 2000 8-in. x 24-in. glass plates on which spectra have been recorded in a fashion similar to a chart recorder. Each plate contains two to six solar spectra covering the 0.3- to 2.5-µm wavelength region. Each of the spectra on the plates was taken through a different amount of the atmosphere. This amount generally varies between two and five times that which would be encountered if the sun were observed while it was directly overhead.

The reduction plan for this data, which was developed in conjunction with Drs. R. Roosen and R. Angione of San Diego State University, is now being implemented. It addresses four problems: 1) the extraction of the data in a computer-readable form from the plates, 2) the characterization of each day's observations on the basis of spectral and other data, 3) the identification of the source of absorption at each wavelength in the spectra, and 4) the determination of CO_2 abundances from the spectra. The entire process of turning the Smithsonian spectrobolograms into useful data will continue to involve the cooperation of many institutions and government agencies, including National Aeronautics and Space Administration's (NASA's) Goddard Space Flight Center, the Smithsonian archives, the United States Naval Observatory and the National Oceanic and Atmospheric Administration (NOAA).

The mechanism for putting the spectrobolograms in computer readable form consists of several important steps. The large plates are first photographically reduced in size at NOAA to permit more rapid analysis. The resulting copies are then scanned with Goddard Space Flight Center's microdensitometer to record plate density as a function of position. The overall accuracy of the above process is continually reviewed using a special standard plate that is measured at the United States Naval Observatory. The tapes of digital data are then sent to PNL, where they are transformed into conventional spectral representatives for subsequent analysis. By the end of FY 1981 more than 1000 plates had been copied and almost 100 had been digitized.

At present, we are analyzing the 1.57-μm band of CO_2 and the 1.27-μm band of O_2 for both the spectrobolograms and other spectroscopic data. The analysis of high dispersion data taken at the Kitt Peak National Observatory (KPNO) shows that the contamination of these bands by other gases, particularly water, is minimal. The current reduction plan is to use these two bands to determine the change in the ratio of the atmospheric abundances of CO_2 to O_2. We will then determine a constant multiplication factor using standard gases and the KPNO Fourier transform spectrograph; at this stage the CO_2:O_2 ratios can be combined with abundances of CO_2 determined by more conventional means to provide a history of the atmospheric burden of CO_2.

• Meteorological Effects of Thermal Energy Releases (METER)

Objectives of this study are:

Developing technical data bases for assessing the potential effects of waste heat sources on weather, climate and ecosystems.

Determining the importance of waste heat releases on the spacing and location of groups of thermal power plants.

Conducting field studies on surface shadowing by cooling tower plumes, surface deposition of chemical drift from towers, augmented precipitation as the result of plume scavenging, increased incidence of fogging and icing from plumes, and effects on local precipitation chemistry.

WINTERTIME PRECIPITATION CHEMISTRY IN NORTH GEORGIA*

M. Terry Dana and A. A. N. Patrinos**

During a ten-week period at the beginning of 1981, precipitation samples were collected over a 300-km^2 network in northern Georgia for chemical analysis. The major objective of this operation was an examination of the effects on local precipitation quality of a large coal-fired power plant located at the center of the network (as part of the U.S. DOE-sponsored METER program). Despite the presence of this large source of sulfur and nitrogen oxides, data from many of the 31 sampling sites showed that precipitation was not affected by the plant during most of the storms sampled. The chemical information obtained from these sites is valuable because it provides a detailed and quality-controlled measure of the wintertime precipitation chemistry in a region perhaps experiencing a trend toward more acidic precipitation. In addition, the background data provide for an examination of the relatively small-scale (sampler separation 2.5 km) variabilities in precipitation chemistry from frontal precipitation in moderately rugged terrain.

Automatic wet-only rain collectors and weekly recording rain gauges were located at 15 network sites. Four of these sites also included daily recording rain gauges. Sixteen additional sites employed bulk precipitation collectors, which were deployed just before it rained. Supporting instrumentation included a Doppler acoustic sounder for fair-weather temperature and wind structure, an instrumented meteorology tower, and at two sites of the network, high volume filter samplers for particulate chemistry and bubblers for SO_2 air concentration.

A total of 11 separate precipitation events (all rain) were sampled. Analyses of the rain samples from most of the storms have been completed for the following species: pH, conductivity, dissolved SO_2, $SO_4^=$, NO_2^-, NO_3^-, Cl^-, PO_4^{-3}, NH_4^+, Na^+, K^+, Ca^{++} and Mg^{++}. This paper is concerned with the four storms most completely analyzed.

The sampling sites not affected by the power plant were objectively determined by considering the average ionic concentrations in each of the four compass quadrant sections of the network. In three of the four storms considered here, three of the quadrants had average concentrations (background) which were nearly the same for each species, while the fourth quadrant had higher concentrations for certain species. Subsequent examination of wind records indicated that the outstanding quadrant ("target") was downwind of the power plant. The fourth event showed no outstanding quadrant, and was considered all background.

Averages and standard deviations for rainfall and concentrations of the major ionic species for the background sites are listed in Tables 1 and 2. While the rainfall amounts vary considerably from storm to storm, variation over the network is small; this uniformity justifies the simple averaging of ionic concentrations for demonstration of chemical uniformity. The concentration standard deviations are generally less than one-tenth the means for the major ions. The ions not listed in the table make up

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*To be presented before the Division of Environmental Chemistry, American Chemical Society, Las Vegas, Nevada, March 28-April 2, 1982.

**Brookhaven National Laboratory, Upton, NY 11973; work was conducted while employed by Oak Ridge National Laboratory.

TABLE 1. Mean and Standard Deviation ( ) of Rainfall for Four Storms. For gauges, N=15 (see Table 2 for Collector N).

Storm	Dates (1981)	Rainfall (cm) Gauges	Collectors
1	Jan 20-21	1.1(0.10)	1.2(0.17)
2	Feb 01-02	3.6(0.19)	3.9(0.35)
5	Feb 10-11	7.6(0.22)	7.9(0.68)
11	Mar 04-05	2.2(0.14)	2.3(0.25)

less than 15% of the total equivalents in all of these storms.

For the four sites with daily recording rain gauges, ion concentrations can be plotted as a function of average rainfall rate. Figure 1 is an example for sulfate in several complete storms and for sequential portions of one of the storms. The all-storm trend is similar to that from earlier measurements in other regions; the 4B sequential, however, shows fine structure in this case an extremely rapid depletion of $SO_4^=$ in the rain at a relatively constant rain rate.

Some initial air concentration results from filters collected at the air chemistry sites allow calculation of dimensionless scavenging or "washout" ratios (ratio of rain concentration to air concentration).

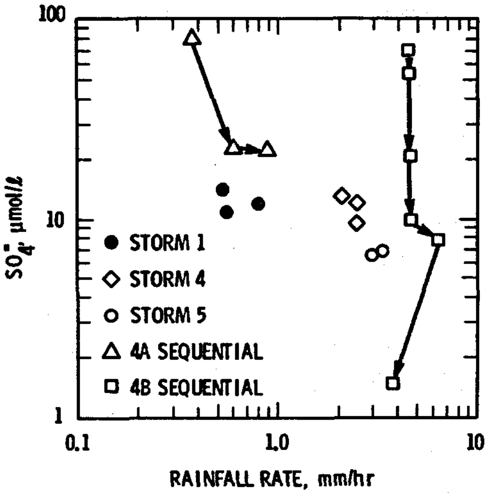

FIGURE 1. Concentrations of Sulfate vs. Rainfall Rates at Daily Rain-Gauge Sites. 4A and 4B refer to two portions of Storm 4. The arrows denote sequence.

The ratio for sulfate during the light rainfall portion of Storm 4 (see Figure 1) is 8.7 x 10^5, while that for the entire Storm 5 (rainfall rate about 4 mm/hr) is 3.5 x 10^5. These values are compatible with recent model calculations. Forthcoming open-literature publications will describe in detail the above results and the results of the power-plant plume effects portion of the field study.

TABLE 2. Mean and Standard Deviation ( ) of Ionic Concentrations

		Concentrations, µmol/ℓ						
Storm	N	SO_2(a)	SO_4	NO_3	Cl	H	NH_4	Na
1	11(b)	5.7(1.3)	11(1.1)	27(2.2)	11(3.6)	61(5.5)	5.0(2.6)	2.1(0.76)
4	18	2.2(1.2)	13(1.5)	9.7(1.3)	12(2.2)	29(2.4)	7.9(1.0)	8.8(1.2)
5	22	.27(0.55)	7.0(0.64)	6.4(0.69)	3.6(0.69)	18(1.7)	1.3(0.45)	1.9(0.44)
11	34(c)	.25(0.61)	26(3.5)	13(1.6)	7.3(1.7)	45(5.5)	9.7(2.2)	3.9(1.0)

(a) Total of S^{IV} measured as $SO_3^=$.
(b) Automatic collectors only.
(c) Three sites had collectors of two types.

- **Theoretical Studies and Applications**

Objectives of this study are:

Developing models for describing the sources, characteristics, evolutions, and fates of energy-related air pollutants.

Translating theory and data into forms useful for evaluating and ameliorating consequences of these pollutants.

PREDICTIONS FOR PARTICLE DEPOSITION TO VEGETATIVE CANOPIES*

W. G. N. Slinn

A theoretical framework has been developed for the analysis of particle deposition to vegetation. This framework is similar to the one used for the analysis of momentum in a vegetative canopy. First, the case of momentum was re-examined, and it was suggested that the pressure-gradient, momentum source can be important in some canopies. An approximate solution to the equation describing the air concentration of particles was then found, using similar approximations found to be reasonable for the case of momentum. A specific, particle-size-dependent collection efficiency was then proposed, based on reported wind-tunnel data. The results suggest that dry deposition velocities for 0.1- to 1.0-μm particles are typically about an order-of-magnitude smaller than values currently used in many assessment models, e.g., those used in nuclear-accident and acid-rain assessments.

ESTIMATES FOR THE LONG-RANGE TRANSPORT OF AIR POLLUTION**

W. G. N. Slinn

It was demonstrated that different atmospheric, source, and surface conditions can result in substantially different transport ranges for airborne pollutants; for example, even for anthropogenic sulfur and nitrogen, the ranges can vary from about 10^1 to 10^5 km. The emphasis in this report is on indicating some of the causes for the great variability in these ranges. Thus, some of the complexities of dry deposition, atmospheric chemistry, and precipitation scavenging are described, and it is demonstrated how synoptic-scale meteorologic conditions can control both dry and wet deposition. On the other hand, it is suggested that the mean, tropospheric-residence time of particulate sulfur and nitrogen from fossil-fuel combustion in temperate latitudes is about a week; but the amount of this material remaining airborne after a week can be large, since it is demonstrated that the amount remaining airborne can be described approximately with a log-normal distribution for an ensemble of realizations. Applications of the results to the U.S./Canadian acid-rain issue, to episodic events, and to global-scale atmospheric pollution are indicated.

WET REMOVAL OF ATMOSPHERIC PARTICLES*

W. G. N. Slinn and J. M. Hales

This report describes interrelations between the precipitation scavenging of particles and of water vapor. Therefore, the report contains a review of water vapor condensation on particles, cloud formation, storms, and precipitation efficiency. The report then demonstrates how scavenging rates, ratios, and efficiencies are related to the corresponding ways to describe the fate of water vapor in the atmosphere. Applications of the results, providing estimates of the tropospheric residence times for anthropogenic particles, are included.

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*To appear as a chapter in the book, Fine Particles in the Atmosphere, (A. P. Altshuller, ed.) to be published by the U.S. Environmental Protection Agency, 1982.

SURFACE FLUXES OF SMALL PARTICLES*

B. B. Hicks** and W. G. N. Slinn

This comprehensive report surveys theoretical analyses, and experimental results and techniques, developed to increase understanding of particle dry deposition from the atmosphere. Preprints of this 130-page report are available.

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*To appear as a chapter in the book, Fine Particles in the Atmosphere, (A. P. Altshuller, ed.) to be published by the U.S. Environmental Protection Agency, 1982.

**Atmospheric Turbulence and Diffusion Laboratory, National Oceanic and Atmospheric Administration, Oak Ridge, Tennessee.

Fission and Fusion

FISSION AND FUSION

- **Air Pollution Dry Deposition: Radioisotopes as Particles and Volatiles**
- **Particle Resuspension and Translocation**

The major pollutants of concern from the nuclear energy industry are long-lived particles (i.e., plutonium and other radionuclides) and waste heat and water vapor from power plant cooling systems. Since these pollutants may affect living species and local and regional climate in an adverse manner, field, laboratory and theoretical investigations of their fate are important to human welfare.

Research activities at PNL include studies of atmospheric diffusion and deposition of material on the earth's surface, resuspension of particulate matter, and regional and global transport of material. Transport studies include the modeling of vertical diffusion and deposition, the determination of the effect of wind speed on resuspension rates and the study of the vertical temperature structure.

• Air Pollution Dry Deposition: Radioisotopes as Particles and Volatiles

Objectives of this study are:

Developing an experimental basis for models predicting removal of airborne particles and gases by dry deposition onto outdoor surfaces.

Developing predictive models accounting for the influence of particle size, gas characteristics, wind velocity, deposition surface and other variables on deposition rates.

Determining deposition rates using dual tracer techniques in the field and in a wind tunnel in the laboratory.

RAIN EFFECTS AND A THRESHOLD WIND SPEED FOR AIRBORNE CONCENTRATIONS FROM AN AREA WIND RESUSPENSION SOURCE OF VOLCANIC ASH

G. A. Sehmel

Airborne concentrations of wind resuspended substances are expected to decrease with time. Airborne concentration decreases may reflect resuspension surface-source changes, surface-source fixation to soil particles or dilution by source penetration to greater soil depths. Although decreases in airborne concentrations are expected for similar meteorological conditions, the half-life at which airborne concentrations from resuspension sources decrease with time is inconsistent and ill-defined (Sehmel 1980). To investigate this decrease, volcanic fallout ash was used as a tracer-of-opportunity for the half-life of a near-infinite source-area of freshly deposited respirable-size particulates. The purpose of this investigation (Sehmel 1981) was to determine the decay of airborne concentrations from an area resuspension source as the source weathered and became less available to be resuspended with time.

Experiment

Two sampling sites at Hanford were activated to collect airborne volcanic ash. Sampling started the day after the major Mount St. Helens eruption, May 19, 1980, and continued through December 18, 1980. Both sites were distant from urban activity. Thus, advected particulates were sampled from area resuspension sources rather than from local-resuspension surface disturbances. The Horn Rapids Dam (HRD) sampling site is near the southwestern boundary of the Hanford area, and the Hanford Meteorological Station (HMS) sampling site is 24 km northwest of the HRD site. Both sites received relatively low ash deposits, which were about 1- to 2-mm and 3- to 4-mm in depth, respectively.

Sampling Equipment

Filters were used to collect airborne solids by:

- sampling continuously for all wind directions and speeds at the HRD and HMS sampling sites
- sampling only during 200° to 290° wind directions at the HMS sampling site.

The sampling time for each filter was a day, several days, or a week.

Airborne Solid Concentrations

Airborne solid concentrations, $\mu g/m^3$, are shown as a function of time in Figures 1 and 2. Data for May 19 to July 13 are shown in Figure 1. Figure 2 shows data from July 12 to December 18. Each figure shows airborne concentrations, airborne solids sampling times and meteorological data. The airborne concentrations are shown at the top of the figure. The horizontal bar through each datum point illustrates the airborne solids sampling time—the day of starting airborne solids sampling to the day of ending sampling. Meteorological data measured at the HMS are shown in the lower parts of each figure.

Effects of Rain

The effects of rain reducing the resuspendability of volcanic ash differed for unaged

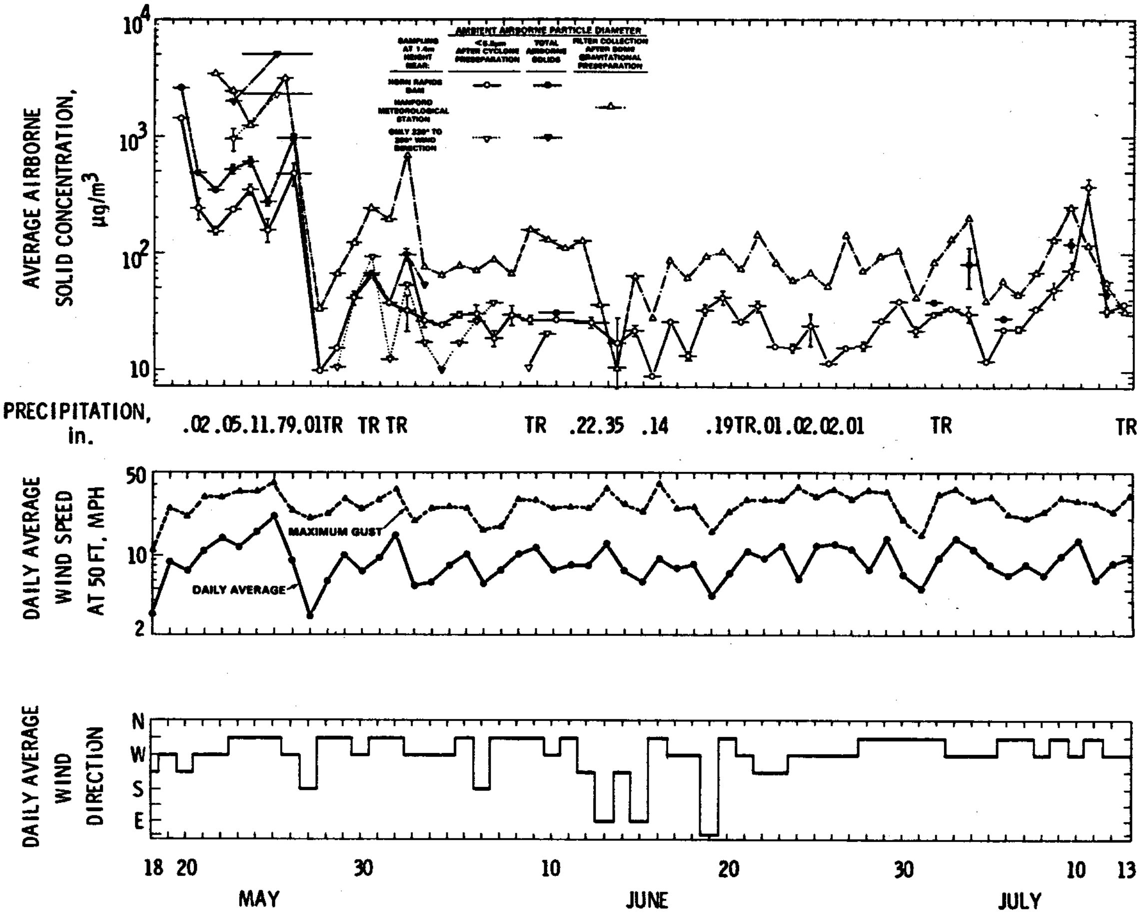

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FIGURE 1. Airborne Solid Concentrations Measured Through July 13 at Two Hanford Area Sampling Sites After the Mount St. Helens Eruption on May 18, 1980

and aged surface sources. Comparable rains were more effective in reducing resuspension rates and, hence, average airborne concentrations for the unaged surface source. However, this apparent increased effectiveness may only reflect that monthly average wind speeds were above a "threshold" wind speed required for increased resuspension rates. The threshold wind speed will be discussed further in the next subsection.

Although resuspension and, hence, airborne solid concentrations are a function of both surface and meteorological parameters, an estimate is made for the amount of rain that significantly reduced resuspension rates from this unaged surface source. There were only three significant rain periods in the months after the eruption through June 20:

- 2.0 cm (0.79 in.) on May 26
- 0.6 and 0.9 cm, 1.5 cm total (0.22 and 0.35 in., 0.57 in. total) on June 12 and 13
- 0.5 cm (0.19 in.) on June 20.

Significant (i.e., an order of magnitude) reductions in airborne solid concentrations occurred only during the first two rain periods (2.0 and 1.5 cm) and not during the last period (0.5 cm). Thus, a tentative conclusion is that 0.5 to 1.5 cm of rain is required to significantly reduce resuspension rates at unaged resuspension sources. However, the duration of reductions may be as short as a day. Airborne solid concentrations increased as ground surfaces dried and resuspension occurred.

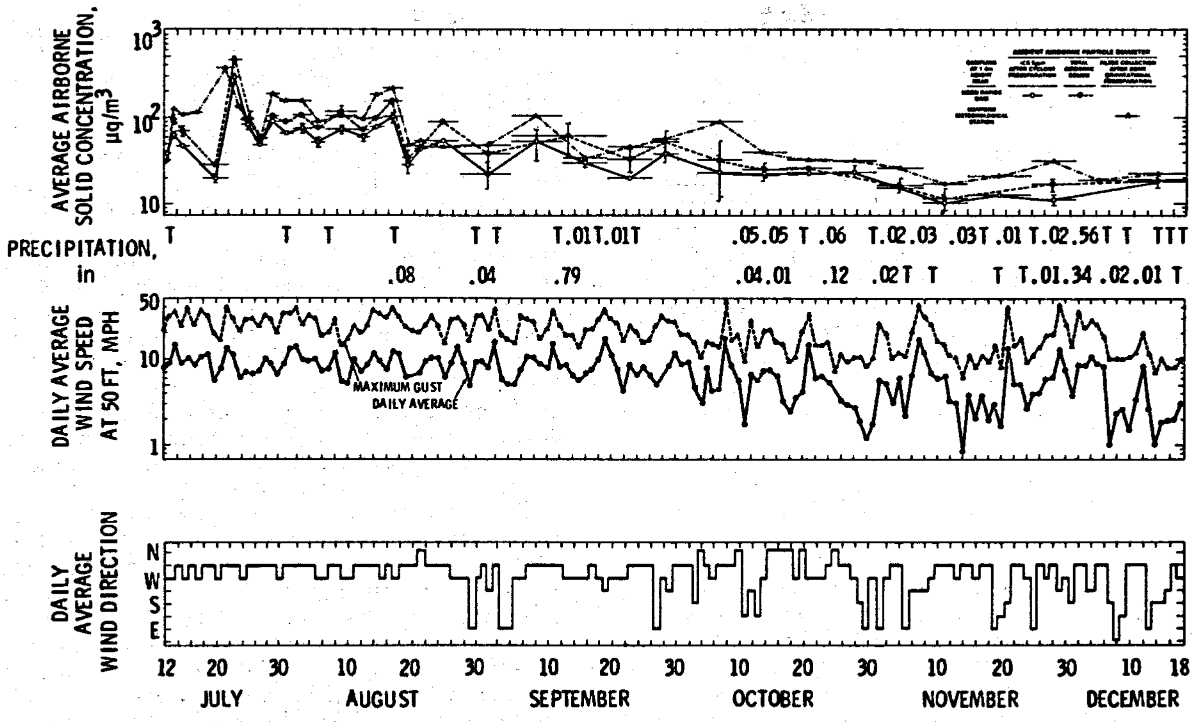

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FIGURE 2. Airborne Solid Concentrations Measured from July 12 Through December 18 at Two Hanford Area Sampling Sites After the Mount St. Helens Eruption on May 18, 1980

The effects of rain on reducing the resuspendability of surface sources may be different for aged sources. In this difference, however, effects on resuspension rates of differing wind speeds are assumed unimportant.

As shown in Figure 2, the precipitation on September 12 was 2.0 cm (0.79 in.), above the 0.5 to 1.5 cm of rain which significantly reduced resuspension rates for the unaged source. For the aged source, however, airborne concentrations did not decrease as significantly as the decrease during the 2.0-cm (0.79-in.) precipitation on May 26.

Resuspension Threshold-Wind Speed

Average airborne solid concentrations fluctuated throughout the time period investigated. On a monthly basis, airborne concentrations tended to be uniform when the average wind speed was above a 3.6-m/sec (8-mph) threshold wind speed. Monthly average concentrations were approximately uniform from June to September. Thus, the half-life for any decrease in airborne concentrations

$$t_{1/2} = \frac{\ln(C/C_0)}{-\lambda} = \frac{\ln 0.5}{-\lambda} = \frac{0.693}{\lambda}$$

was infinity for a four-month time period. The weathering rate, λ, was zero. For monthly-average wind speeds less than the threshold wind speed, monthly-average airborne concentrations tended to decrease with time.

Beginning in September, 3-1/2 months after the May 18th eruption, concentrations exhibited a relatively persistent decrease on a monthly time scale. Some of the decrease may reflect:

- more persistent precipitation, though small, decreasing resuspension rates
- a persistent decrease in average wind speeds starting in about September.

For this four-month time period, the half-life was on the order of 50 days, corresponding to a weathering rate, λ, of 5.1 yr^{-1}. The airborne concentration half-lives of infinity and 50 days are only for monthly-average concentrations and wind speeds.

Monthly average wind speeds would not reflect the detailed resuspension physics from isolated small sources, but may be suitable to parameterize resuspension from wide area sources. The monthly average wind speeds (15-m level) are shown in Table 1. If September were a suspension transition month, the monthly average wind speeds and concentration data could reflect a resuspension threshold wind speed above which resuspension is more pronounced. The threshold wind speed was interpreted from monthly average concentrations which tended to:

- remain relatively high for monthly average wind speeds greater than 3.6 m/sec (8 mph)
- decrease for monthly average wind speeds less than 2.4 m/sec (5.4 mph).

Although a tentative value of 3.6 m/sec (8 mph) is suggested as a threshold wind speed for resuspension, resuspension events are spatially dependent and episodic; i.e., resuspension rates increase with wind speed raised to a power, u^n, where n is often 3 or greater (Sehmel 1980). Although resuspension is episodic, the volcanic ash fallout was a resuspension source of large area extent, which might have been nearly impossible to investigate otherwise.

TABLE 1. Monthly Average Wind Speeds (15-m level)

Month	m/sec	mph
May	3.8	8.5
June	3.7	8.3
July	4.0	9.0
August	4.0	9.0
September	3.8	8.4
October	2.4	5.3
November	2.4	5.4
December	2.2	5.0

Conclusions

In addition to recording information on volcanic ash concentrations, airborne concentrations were used to estimate a threshold wind speed for increased resuspension rates and the amount of rain to significantly reduce resuspension rates for an unaged source. A significant attribute of this source was that the volcanic ash contained a large fraction of respirable particle sizes, a near ideal condition for investigating airborne concentrations of potential inhalation concern.

The amount of rain required to significantly reduce resuspension rates depended, in part, on the age of the resuspension source. For a nearly unaged source, the 2.0-cm (0.79-in.) of rain on May 26 significantly reduced airborne concentrations. Afterward, airborne solid concentrations increased as surfaces dried and resuspension occurred. Changes in airborne solid concentrations after subsequent rains through July indicate 0.5 to 1.5 cm (0.2 to 0.6 in.) of rain was required to significantly reduce, at least for a day, airborne solid concentrations from an unaged wide-area resuspension source. In contrast, the amount of rain required to significantly reduce airborne concentrations appears different for an aged source. There was also a rain of 2.0 cm (0.79 in.) on September 13. In this case, airborne concentrations did not decrease significantly.

A monthly-average threshold wind speed for resuspension was defined as 3.6 m/sec (8 mph). A significant weakness of this threshold velocity is that most collected solids were resuspended upwind at locations of differing wind speeds, an obvious conclusion since wind speeds show significant spatial variations. Nevertheless, resuspension data are needed for wide-area sources.

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• Particle Resuspension and Translocation

Objectives of this study are:

Investigating particle resuspension rates and air concentrations as functions of atmospheric stresses, local soil-working stresses, particle variables and surface variables.

Developing an experimental resuspension data base to aid in establishing surface contamination limits to assure adequate public protection from hazardous materials.

Factoring time-varying resuspension rates into models describing delivery of surface contaminants to man.

A RELATIONSHIP BETWEEN PLUTONIUM ACTIVITY DENSITIES ON AIRBORNE AND SURFACE SOILS

G. A. Sehmel

Resuspension is defined as the insertion of surface contamination into the air by wind or mechanical stresses.

Airborne concentrations from resuspension have been predicted based on considerations of airborne mass loading and pollutant concentrations on the airborne soil (Anspaugh, Shinn and Wilson 1974; Anspaugh et al. 1975). The mass loading approach was developed based on an assumption of equality; i.e., the pollutant concentration per unit mass of airborne soil (A_{AIR}) is identically equal to the pollutant concentration per unit mass on the local soil surface $A_{SURFACE}$. In this case, airborne pollutant concentrations are predicted from the equation

$$C = (ML)(A_{AIR})$$

where ML = airborne soil mass loading per unit volume of air
A_{AIR} = pollutant concentration per unit mass of airborne soil.

The method was based upon data for plutonium. In this case, the pollutant concentrations on airborne and surface soils are activity densities, A_{AIR} and $A_{SURFACE}$, fCi/g. In developing this method, predicted airborne concentrations, μCi/cm^3, were based on an assumed airborne mass loading of 100 μg/m^3 and an equality of A_{AIR} and $A_{SURFACE}$. The activity density on airborne solids was not determined.

The purpose here (Sehmel 1981) is to summarize data for plutonium on airborne and surface soils, if both are available at study sites, and to examine the data for relationships between plutonium concentrations on airborne soils and on surface soils near the airborne particulate sampling sites. Data will be summarized only for sites where plutonium concentrations on both airborne and surface soils have been investigated. These sites include the Bikini Atoll (Shinn, Homan and Robinson 1980), the Hanford site (Sehmel 1980b, 1981), and Rocky Flats (Krey et al. 1976; Sehmel 1980b). The plutonium concentrations are reported as activity density ranges, fCi/g of collected soil, i.e., 10^{-15} Ci/g. All plutonium activity density data on surface soils were determined for total soil samples rather than as a function of host soil particle diameters to which the plutonium was attached. Thus, the reported plutonium activity densities are for both respirable and nonrespirable size host soil particles. In contrast, most plutonium activity densities for airborne solids were determined for particles which were respirable, or near respirable.

Activity Density Relationship

Plutonium activity densities on airborne, A_{AIR}, and surface soils, $A_{SURFACE}$, are summarized (Sehmel 1981) as ranges measured at Bikini Atoll, Hanford, and Rocky Flats. Activity densities on airborne soils range from 10^0 to 10^5 fCi/g, while activity densities on surface soils range from 10^0 to 10^6 fCi/g.

The maximum activity densities on airborne soils as a function of activity densities on surface soils were represented by two equations (Sehmel 1982). The maximum activity density equations reflect the presence in airborne samples of plutonium and soil particles from both upwind and local resuspension. The equations intersect at an activity for surface soils of 2 x 10^3 fCi/g. For surface soil activity densities "less" than at the intersection, the equation is

$$A_{AIR} = 3.3 \times 10^3 (A_{SURFACE})^{0.35}. \qquad (1)$$

For surface soil activity densities "higher" than at the intersection, the equation is

$$A_{AIR} = 1.3 \times 10^4 \; (A_{SURFACE})^{0.17}. \qquad (2)$$

Conclusions

An improved but conservative, high-value, relationship was developed for using the mass-loading calculational approach for predicting airborne plutonium concentrations from wind resuspension. The method does not address changes in resuspension as a function of wind speed, surface cover, or contaminant parameters. The improvement was to include the observed activity densities on airborne soils over regions of different surface contamination levels. The relationship correlates the maximum observed plutonium activity densities on airborne soils as a function of the minimum plutonium activity densities on surface soils. The activity density on airborne soil is not simply equal to the activity density on surface soils for sites investigated. The inequality in activity densities is caused, in part, by the relative contributions to the airborne mass loading from soil transported from upwind, as compared to soil resuspended locally near the airborne particulate sampling site.

The activity density relationship can be used for improved predictions of airborne concentrations from wind resuspension: 1) to decrease the ultraconservatism in estimating airborne plutonium concentrations above more "highly" contaminated surface areas, and 2) to more realistically predict airborne concentrations over surfaces of "lesser" surface contamination, i.e., much closer to surface contamination levels attributed solely to weapons testing fallout. The relationship is useful for predicting airborne concentrations above a contaminated area, but has less utility for predicting airborne concentrations downwind. Resuspension rates (Sehmel 1980b) are needed for predicting airborne concentrations downwind.

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RESUSPENSION FACTORS FOR AGED PLUTONIUM SOURCES

G. A. Sehmel

Wind and mechanical stresses on contaminated soils can resuspend the contaminant; thus airborne contaminant concentrations can be an inhalation source for years. Accurately predicting airborne concentrations is important in evaluating airborne effects from resuspension. During some resuspension investigations, airborne contaminant concentrations per unit volume of air, X, have been related to the contaminant surface concentration per unit area on the ground, G. The resuspension factor, RF, is defined as

$$\text{RF (units of } m^{-1}) = \frac{\text{X, airborne concentration } (m^{-3})}{\text{G, surface contamination } (m^{-2})}$$

The resuspension factor (Sehmel 1980) has ranged in experimental studies from about 10^{-10} to 10^{-2} m^{-1} (9×10^{-11} to 3×10^{-4} m^{-1} for wind resuspension and 1×10^{-10} to 4×10^{-2} m^{-1} for mechanical stresses). Often in using these data an average resuspension factor is assumed. Although assumed, there has been limited guidance for selection except by personal judgment.

The purpose of this investigation was to develop guidance for selecting conservative resuspension factors, conservative by predicting maximum airborne plutonium concentrations. The guidance was to be developed using plutonium activity densities for both airborne and surface soils.

Data Base

Plutonium activity densities for both airborne and surface soils have been reported only for the Bikini Atoll, the Hanford area and the Rocky Flats area. Shown in Figure 1 are data from several sources summarized by Sehmel (1981). Reported activity densities for surface soils, $A_{SURFACE}$, ranged over six orders of magnitude from 2 to 3×10^6 fCi/g while activity densities on airborne solids, A_{AIR}, ranged five orders of magnitude from 2 (also to less than analytical limits) to 1.3×10^5 fCi/g.

The maximum activity densities on airborne solids were a function of the activity

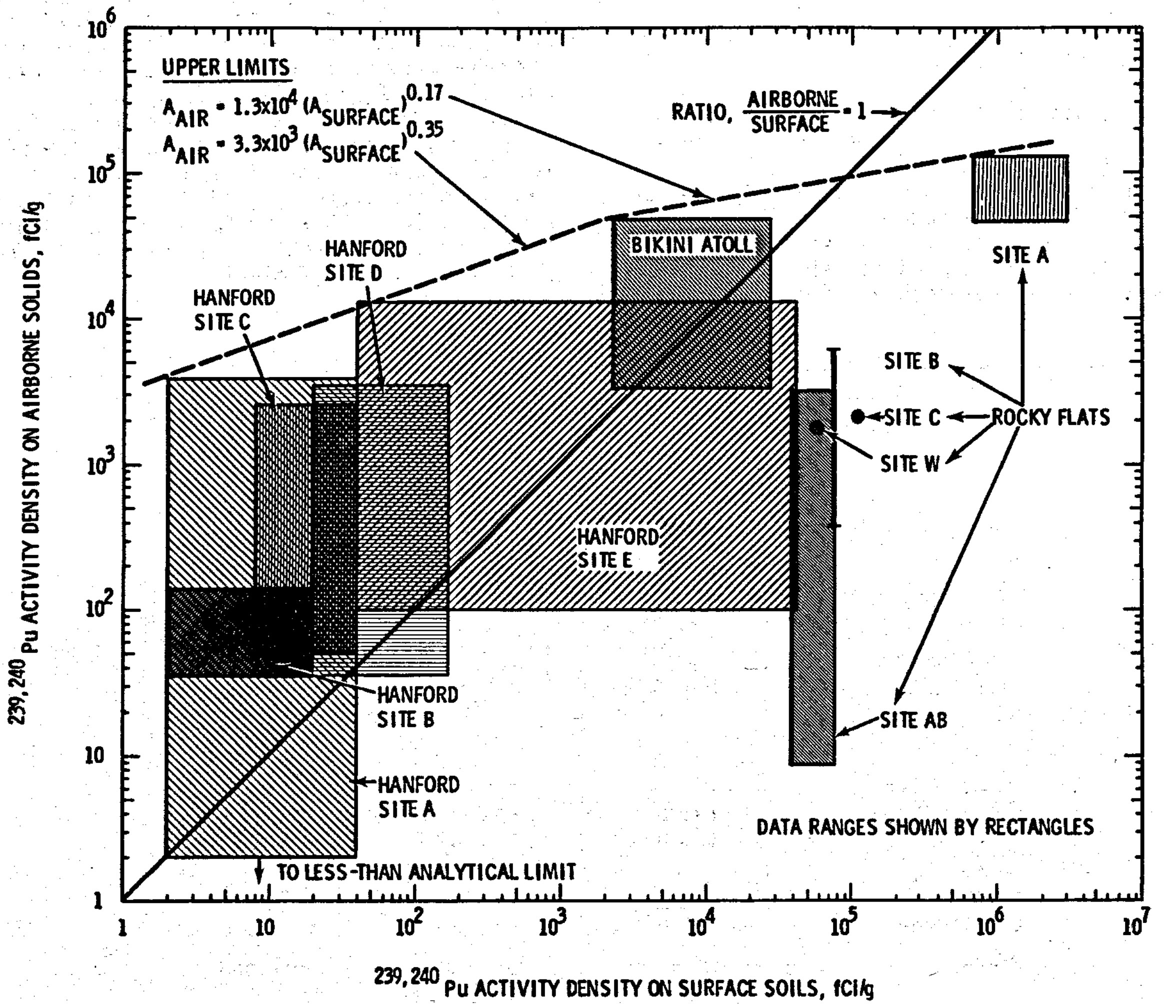

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FIGURE 1. Plutonium-239, 240 Activity Densities on Airborne and Surface Soils

densities on surface soils. For surface soil activity densities less than 9 x 10^4 fCi/g, plutonium was enriched on the airborne solids. For surface soil activity densities greater than 9 x 10^4 fCi/g, plutonium was depleted on the airborne solids. These enrichment and depletion relationships are represented by two equations. The equation intersects at a surface soil activity density of 2.0 x 10^3 fCi/g. For surface soil activity densities less than the intersection,

$$A_{AIR} = 3.3 \times 10^3 \left(A_{SURFACE}\right)^{0.35} \qquad (1)$$

and for surface soil activity densities greater than the intersection,

$$A_{AIR} = 1.3 \times 10^4 \left(A_{SURFACE}\right)^{0.17}. \qquad (2)$$

Resuspension Factors

Resuspension factor ranges were calculated from the upper limits represented by Equations 1 and 2. For these calculations, ranges of airborne mass loadings, C, and resuspension surface source depths, Δz, were assumed. Next, activity densities were applied to the corresponding calculated soil masses. Thus, resuspension factors were calculated from

$$RF = \frac{A_{AIR}\left(C, \frac{\mu g}{m^3}\right)\left(\frac{g}{10^6\ \mu g}\right)}{A_{SURFACE}\ (\Delta z, cm)\left(10^4\ \frac{cm^2}{m^2}\right)\left(1.5\ \frac{g}{cm^3}\right)}$$

$$= 6.7 \times 10^{-11}\ \frac{\left(A_{AIR}\right)}{\left(A_{SURFACE}\right)}\ \frac{(C)}{(\Delta z)}$$

where a soil density of 1.5 g/cm^3 was assumed. Calculations were made for resuspension source depths of 0.1 and 1.0 cm and for airborne mass loadings of 10 and 260 µg/m^3. These ranges are considered realistic. The resuspension source depth actually resuspended is unknown, but probably is on the order of 0.1 cm for non-eposodic resuspension events. The 10-µg/m^3 airborne mass loading corresponds to clean air while the 260-µg/m^3 airborne mass corresponds to the 24-hr maximum National Ambient Air Quality Standard (NAAQA). Calculations were made for surface soil activity densities of 2 and 3 x 10^6 fCi/g (corresponding to the minimum and maximum values shown in Figure 1), and also at an intermediate value of 9 x 10^4 fCi/g for which airborne and surface activities are equal. Results are shown in Table 1.

TABLE 1. Resuspension Factors for Aged Plutonium Sources

	Surface Contamination		
	Low	Intermediate(a)	High
Activity density, $A_{surface}$ (fCi/g)	2	9 x 10^4	3 x 10^6
Activity density, A_{air} (fCi/g)	4.2 x 10^3	9 x 10^4	1.6 x 10^5
Enrichment factor ($A_{air}/A_{surface}$)	2.1 x 10^3	1.0	5.3 x 10^{-2}
Resuspension factor (m^{-1})			
for Δz = 0.1 cm			
C = 10 µg/m^3	1.4 x 10^{-5}	6.7 x 10^{-9}	3.6 x 10^{-10}
C = 260 µg/m^3	3.7 x 10^{-4}	1.7 x 10^{-7}	9.2 x 10^{-9}
for Δz = 1.0 cm			
C = 10 µg/m^3	1.4 x 10^{-6}	6.7 x 10^{-10}	3.6 x 10^{-11}
C = 260 µg/m^3	3.7 x 10^{-5}	1.7 x 10^{-8}	9.2 x 10^{-10}
Range	10^{-6} to 10^{-4}	10^{-9} to 10^{-7}	10^{-11} to 10^{-8}

(a) Intermediate corresponding to an enrichment factor of unity.

Conclusions

Calculated resuspension factors range six orders of magnitude from 10^{-11} to 10^{-4} m^{-1}, a range almost comparable to the range of experimentally measured resuspension factors. The similarity is encouraging and lends credence to the resuspension factor predictions. An important predictive improvement is that resuspension factor ranges can now be predicted as a function of the resuspension-source activity density. Resuspension factors range only two orders of magnitude from 10^{-6} to 10^{-4} m^{-1} for low surface contamination, contamination levels which might be applicable for the general population. In comparison, resuspension factors range three orders of magnitude from 10^{-11} to 10^{-8} m^{-1} for the highest reported surface contamination, contamination levels for which controls might be applicable.

The activity density data are for aged plutonium resuspension sources, rather than for resuspension from freshly deposited sources. Hence, the predicted resuspension factors could be used in long-term assessments. In comparison, resuspension from a freshly deposited source is expected to be more rapid. For the freshly deposited source, less confidence should be placed in the resuspension factor predictions.

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PLUTONIUM AND AMERICIUM ACTIVITY DENSITIES ON AIRBORNE SOLIDS

G. A. Sehmel

Evidence is increasing that resuspended plutonium is distributed nonuniformly as a function of host particle size (Sehmel 1981). During resuspension the plutonium is enriched on the smaller particles; i.e., the plutonium activity density, µCi/g, is usually larger for respirable compared to nonrespirable size particles. In other words, the activity density on resuspended respirable particles is larger than on bulk surface-soil samples. Although evidence is mounting to support these enrichment concepts, little is known about enrichment processes, enrichments for different radionuclides, or predictable enrichments as a function of particle size. Enrichment must be predictable if satisfactory methods are to be developed for relating resuspension surface-source characteristics to resulting airborne concentrations from resuspension.

To investigate enrichment, radionuclide enrichment was sought under field conditions. Enrichment could be investigated with samples collected from either resuspension sources or from airborne solids. However, existing resuspension sources are nonuniform, requiring many surface samples before source strengths can be accurately estimated by integrating concentrations over those samples. In contrast, integration over source variations has already occurred for samples of airborne solids. These latter integrations can also reflect activity density differences as a function of sampling height because of the relative source distances, the differences for each radionuclide, the meteorology and the transport of uncontaminated soil from upwind. The purpose of this investigation was to simultaneously investigate activity densities as a function of host particle diameter and sampling height for both airborne plutonium and americium.

Experiment

Airborne samples were collected as a function of height from the Hanford Meteorological Station (HMS) tower. Samples were collected at four sampling heights of 1.4, 17, 63, and 124 m during all wind speeds, but only for a selected wind direction segment of 225° ±35°. This wind direction segment encompasses the times of most high wind speeds, resuspension increases with increasing wind speed. Hence, this wind-direction sampling segment was selected to minimize sample variations caused by any resuspension source differences. Another source was also investigated by sampling only during wind directions of 45° ±145°, at three sampling heights of 1.4, 62, and 124 m. Sampling was between April 12 to July 25, 1979. Samplers were turned off during precipitation.

Samples were collected with three-part sampling systems, each system consisted of a cyclone-preseparator, a cascade-particle-impactor and back-up filter. Samples were analyzed separately for each part of the collection system. The 50% cut-off diameter for the cyclone-preseparator is 5.5 µm and for the last stage of the cascade impactor is 0.5 µm. Thus, radiochemical results were obtained for airborne particle diameters greater than 5.5 μm, for particle diameters from 0.5 to 5.5 μm, and for particle diameters less than 0.5 μm.

Results

Activity densities, μCi/g, for plutonium-239, -240 and americium-241 are shown in Figures 1 and 2, respectively, for sampling during the 225° ±35° wind direction segment. Also shown for completeness are airborne concentrations, μCi/cm^3. The error bars at each data point correspond to the one-sigma radiochemical counting limits. If limits include zero, the data point is shown at the upper limit with an arrow pointing toward zero. In contrast for the 45° ±145° wind direction segment, results are tabulated in Table 1. Tabulation is used to de-emphasize the possibility of concluding changes with height for the 45° ±145° wind direction segment, de-emphasized because these samples were collected during significantly different time periods because of equipment malfunctions.

Activity densities and airborne concentrations were a function of both sampling height and host particle diameter. For both plutonium and americium, activity densities were maximum near the 17-m sampling height. These maximum activity densities may reflect either upwind radionuclide resuspension with particle dry deposition occurring between the resuspension source and the HMS tower sampling site or vertical diffusion from an elevated source. For plutonium, activity densities were maximum for the intermediate size range, >0.5 to <5.5 μm. The particle size range for maximum americium activity densities is less certain since americium analyses were only made for two particle diameter ranges, <0.5- and >5.5-μm. Similar to plutonium for these two size ranges, the maximum americium activity density was, with one exception, on the <0.5-μm size. At the 62-m sampling height, the americium activity density on the >5.5-μm size was greater than on the <0.5-μm size.

Conclusions

Plutonium-239, -240 on resuspended airborne host particles is enriched, μCi/g, in the respirable versus the nonrespirable host particle size range. For samples collected during southwest winds, enrichment was largest for the 0.5- to 5.5-μm particle diameter range.

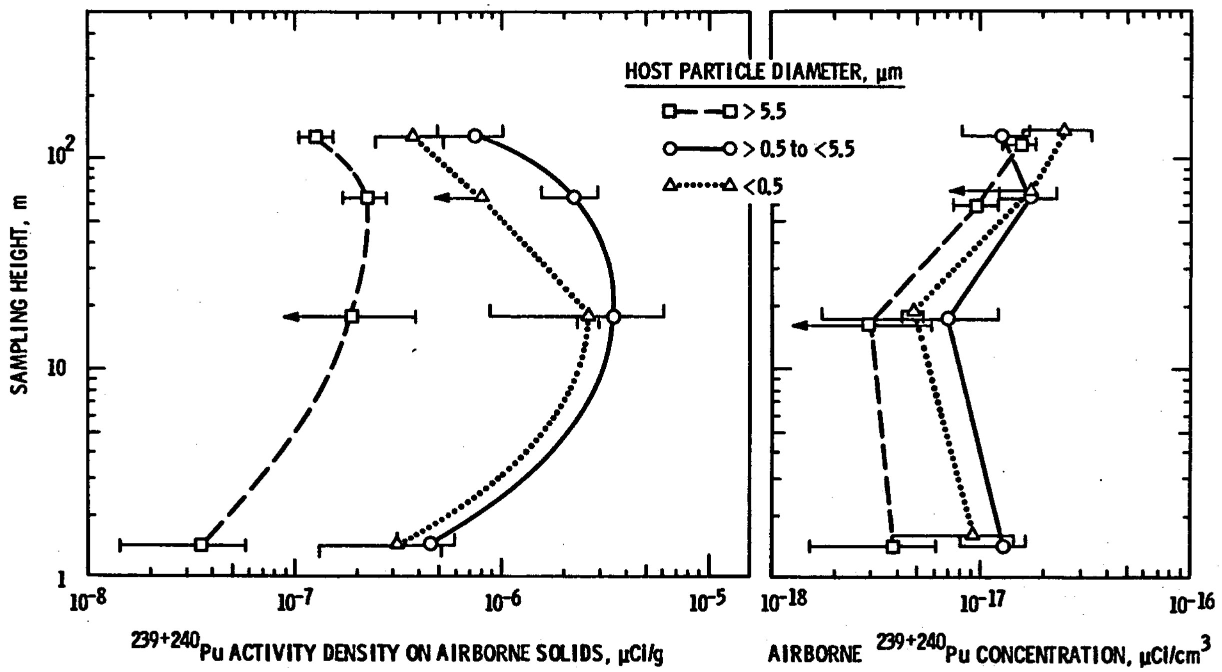

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FIGURE 1. Airborne ^{239,240}Pu Activity Densities and Concentrations as a Function of Height at the HMS Tower from April 12 to July 25, 1979, When Sampling During Winds from 225° ± 35°

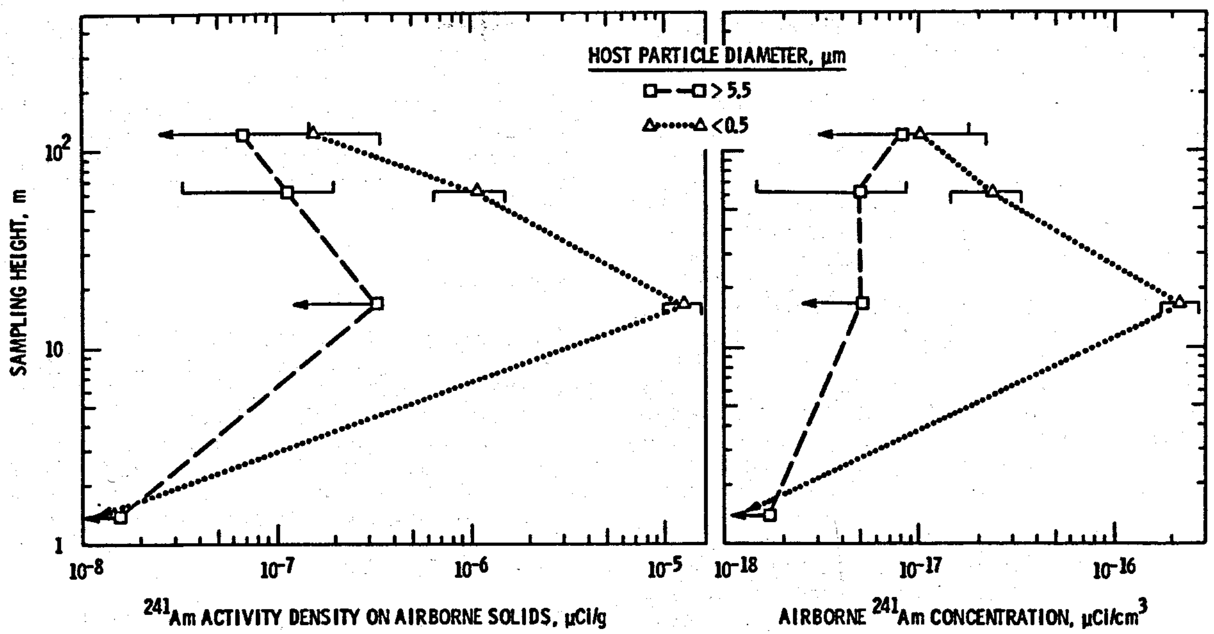

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FIGURE 2. Airborne ^{241}Am Activity Densities and Concentrations as a Function of Height at the HMS Tower from April 12 to July 25, 1979, When Sampling During Winds from 225° ± 35°

TABLE 1. Airborne $^{239+240}$Pu and ^{241}Am Activity Densities and Concentrations as a Function of Height at the HMS Tower from April 12 to July 25, 1979, When Sampling During Winds From 45° ± 145°

		$^{239+240}$Pu			^{241}Am		
Sampling Height (m)	Host Particle Diameter (µm)	Activity Density (µCi/g)	Airborne Concentration (µCi/cm³)	1σ Radiochemical Error (±%)	Activity Density, (µCi/g)	Airborne Concentration (µ/Ci/cm³)	1σ Radiochemical Error (±%)
124	>5.5	2.1×10^{-7}	3.7×10^{-16}	28	$<5.2 \times 10^{-8}$(a)	$<8.9 \times 10^{-19}$(a)	(a)
	>0.5 to <5.5	2.2×10^{-6}	8.5×10^{-16}	19	NA	NA	NA
	<0.5	6.7×10^{-7}	5.0×10^{-16}	33	1.6×10^{-6}	1.2×10^{-17}	42
62	>5.5	7.7×10^{-7}	3.9×10^{-16}	12	2.8×10^{-7}	1.1×10^{-16}	25
	>0.5 to <5.5	1.1×10^{-6}	5.9×10^{-16}	10	NA	NA	NA
	<0.5	7.6×10^{-7}	7.0×10^{-16}	12	7.9×10^{-8}	7.2×10^{-19}	75
1.4	>5.5	6.8×10^{-8}	3.1×10^{-16}	11	1.2×10^{-8}	5.4×10^{-19}	29
	>0.5 to <5.5	1.2×10^{-6}	7.4×10^{-16}	7	NA	NA	NA
	<0.5	3.5×10^{-7}	2.2×10^{-17}	8	4.2×10^{-8}	2.6×10^{-18}	17

(a) This is the radiochemical analysis error when the radiochemical result is zero.
NA = not analyzed for americium content.

The relative enrichment is illustrated by normalizing to the mass-average activity density for the total collected sample at each height. For the 0.5- to 5.5-μm particle diameter range, the activity density range is 2.1 to 6.0 times the average activity density for the total sample. Except for one sample with zero enrichment in the less than 0.5-μm particle diameter range, the activity density range is from 1.5 to 2.0 times the average activity density for the total sample. For samples collected during northeast winds, the enrichment range is larger, 0.9 to 4.2.

Americium-241 is also enriched on respirable particles. However, the relative enrichment compared to the total airborne sample is unknown, the >0.5- to <5.5-μm size fraction was not analyzed for americium-241 content.

For surface soil samples, enrichments on the respirable particle size range may be even greater than the 0.9- to 6.0-enrichment range measured for airborne particles. The potential enrichment increase reflects that the collected airborne solids did not contain the largest size soil particles, soil particles of lowest activity-density but largest mass-fraction in surface soils. Truncation in the airborne size distribution is caused by the largest size surface-soil particles, which tend to move by surface creep and saltation. Most of these particles are usually transported below 1.4 m, the lowest sampling height.

Radionuclide surface sources can be estimated from bulk soil samples. Activity densities for these bulk surface samples can be low, but significant resuspension can occur if all the surface contamination is enriched on small particles. Methods need to be developed to predict airborne concentrations from resuspension sources of differing enrichment.

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SAMPLING REPRODUCIBILITY FOR AIRBORNE SOLIDS

G. A. Sehmel

Airborne concentrations of ambient solids are often calculated from single samples at air pollution monitoring stations. Much effort is expended in accurately collecting these samples. Although these samples are collected with state-of-the-art sampling equipment, the concentration uncertainty may still be controlled by the true spatial variations in airborne concentrations. These variations could be a function of the airborne solid source characteristics and distances from the sources. The purpose of this investigation was to investigate sampling reproducibility from an area source, a source of resuspending volcanic ash.

Experiment

The sampling site (Sehmel 1981a) for investigating sampling reproducibility was within the Hanford area near the Horn Rapids Dam. This site was near the southern edge of the fallout plume from the May 18, 1980 Mount St. Helens eruption. At this site, the volcanic ash deposit was about 1 to 2 mm. Airborne solids were collected during high to low airborne concentrations, from May 19 to December 18, 1980.

Sampling reproducibility was investigated with two samplers located 2 m apart. The air-sampling flow rate was 1.1 m^3/min at a sampling height of 1.4 m. The diameter of the cylindrical inlet was 7.1 cm. During sampling the inlets were continuously oriented into the wind with tail fins. Several sampling times were used, a day, several days, or a week. Sampling was continuous, except for automatic shutoff during rain.

The samplers were cyclone preseparators with back-up filters. This sampler is designed to separate particles into size fractions, those above and those below 5.5-μm diameter. The 5.5-μm is the 50% cutoff diameter for the cyclone; i.e., 50% are collected in the cyclone and 50% pass through and are collected on a filter. Particles passing through the cyclone are collected on a 20- x 25-cm^2 glass-fiber filter. After sample weighing, airborne solid concentrations were calculated separately from mass collected on the filter and the total mass of filter-plus-cyclone-separator collections. To determine the mass of solids collected, filters were equilibrated with low-humidity laboratory air both before and after sample collection. Solid weights were determined after equilibration.

Sample Reproducibility

Statistical tests of the measured concentrations were made to investigate sampling reproducibility. Parameters used for each sampling time period were average concentration, $\overline{C}_i$, the concentration variance, s_i, and the coefficient of variation, $(s/\overline{C})_i$. Concentrations are shown by Sehmel (1981b). For statistical analysis, logarithmic transformations were used, i.e., $\ell n(s/\overline{C})_i$ and $\ell n(\overline{C}_i)$. The average concentration, $\overline{C}_i$, during each sampling period is the average of the two collected samples, $(C_{1i} + C_{2i})/2$. The standard deviation, s_i, is

$$s_i^{\,2} = \frac{(C_1 - C_2)_i^2}{2} .$$

Two statistical tests were used to analyze the transformed data. The results from a linear comparison of the data showed that $\ell n(s/\overline{C})_i$ was not a statistically significant function of $\ell n(\overline{C})_i$. Thus, the average and variance of $\ell n(s/\overline{C})$ were calculated. Subsequently, these calculated values were transformed back into the measured values.

For particles <5.5 μm diameter, the average sampling variance was

$$s_i = 0.11\ \overline{C}_i$$

with a one standard deviation range from $0.038\ \overline{C}_i$ to $0.31\ \overline{C}_i$. For total particle collection, the average sampling variance was

$$s_i = 0.099\ \overline{C}_i$$

with a one standard deviation range from $0.033\ \overline{C}_i$ to $0.30\ \overline{C}_i$.

Conclusions

The variance of the average sampling reproducibility was about 10 percent of the average airborne concentration, an acceptable $(s/\overline{C})_i$ for many applications. The variance range, however, is much larger. The sampling variances, though small, could have significant impact on compliance with and establishment of regulations. For instance, there are standards or proposed standards for fugitive emissions, prevention of significant deterioration (PSD) of ambient airborne particulate concentrations, and ambient airborne pollutant concentrations. One interpretation of the variance in sampling reproducibility is that regulations based upon sampling for compliance should also include an interpretation of sampling variance in those regulations. The suggestion is that regulations could be based in part upon sampling uncertainty, rather than solely upon absolute air pollution concentrations.

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ENRICHMENT OF AMERICIUM-241 ON AIRBORNE COMPARED TO SURFACE SOIL SAMPLES

G. A. Sehmel

Evidence is increasing that radionuclide activity densities, μCi/g, on airborne soils are greater than on bulk samples of surface soils. This enrichment reflects a preferential radionuclide particle attachment and/or transport on respirable-size host-soil-particles. In assessments of resuspension, enrichments on respirable particle sizes can impact the prediction accuracy of airborne concentrations. If airborne radionuclide activity-densities are only assumed equal to activity-densities for bulk soil surface samples, predictions may be less conservative than previously thought; i.e., predicted airborne concentrations may be low.

Although plutonium enrichments have been investigated, americium-241 enrichments have not been reported. There are now data from which americium-241 enrichments can be calculated. Americium-241 activity densities have been reported for both airborne samples (Sehmel 1981) and soil surface samples (Price, Gilbert and Gano 1981). The airborne samples were collected from the Hanford meteorological tower. The surface samples were collected at two sites, onsite about 500 m east of the PUREX gaseous effluent discharge stack and offsite approximately 60 km west from Hanford. Samples were collected either as a function of particle diameter, or for all particle diameters. The airborne particles were collected with cyclone preseparator cascade-impactor collection systems. Americium-241 activity densities were obtained for the greater than 5.5 μm diameter airborne size fractions. The soil surface samples were collected for total soil depths up to 2.5 cm.

Analysis

Reported activity-density ranges are summarized in Table 1. Activity densities range from less than radiochemical detection limits to 1.2×10^{-5} µCi/g. There are differences in the activity density ranges as a function of particle size. The upper-limit of each activity-density range is largest for the less-than 0.5-µm diameter airborne particles. An enrichment is indicated. The upper-limit for 0.5-µm particles, 1.2×10^{-5} µCi/g, is 1.2×10^{2} times the upper-limit for surface soils, 9.8×10^{-8} µCi/g. In contrast, the upper-limit for 5.5-µm particles, 1.1×10^{-7} µCi/g, is only 1.1 times the upper-limit for surface soils.

Conclusions

Resuspended americium-241 is enriched, µCi/g, on airborne soils less than 0.5 µm diameter. These enrichments, as estimated from activity-density ranges, can be as large as 10^{2}. Although enrichments do occur for americium-241 as well as other radionuclides, enrichments can only be imprecisely predicted. An increasing enrichment data base is needed for general predictions.

References

Price, K. R., R. O. Gilbert and K. A. Gano. 1981. Americium-241 in Surface Soil Associated with the Hanford Site and Vicinity. PNL-3731, Pacific Northwest Laboratory, Richland, Washington.

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TABLE 1. Americium-241 Activity Densities

Source	Particle Diameter (µm)	Activity Density Range (µCi/g)
Airborne	>5.5	LAL[(a)] to 1.1×10^{-7}
Airborne	<0.5	LAL[(a)] to 1.2×10^{-5}
Surface soils		
Onsite	All	LAL[(a)] to 9.5×10^{-8}
Offsite	All	LAL[(a)] to 9.8×10^{-8}

(a) LAL = Less than radiochemical analytical limits.

PLUTONIUM SOURCE IDENTIFICATION IN SURFACE SOILS

G. A. Sehmel

Even though satisfactory for surveillance purposes, the usual soil sampling procedures and radiochemical analyses only partially characterize resuspension sources. Characterization is incomplete since surface-depth samples are collected for all soil particle sizes, and total plutonium contents are determined.

Resuspension source strength characterization could be improved during both soil sampling and plutonium analysis. The source of resuspended particles is from the top layer of the soil surface, the usual layer of maximum activity density, µCi/g. This top surface layer needs characterization for plutonium content. When resuspension is considered, more complete source characterization of this top layer includes investigating activity densities as a function of particle size and plutonium isotopic content. Analyses as a function of particle size are needed since airborne respirable size soil particles are enriched in plutonium content relative to activity densities determined from total-soil, surface-depth collected samples.

Plutonium isotopic analyses are needed to investigate the relative proportions of fallout versus process-produced plutonium in the soil. The ^{240}Pu/^{239}Pu atom ratios are significantly greater for fallout compared to process-produced plutonium for the military.

Atom ratios for integrated nuclear-weapons testing fallout are reflected in soil samples taken at sites remote from production facilities. For soil-surface samples collected in 1970-71 with atom ratios corrected to January 1971, worldwide fallout ^{240}Pu/^{239}Pu atom ratios average 0.18 ± 0.01 (Krey et al. 1976, pp. 671-678). These results are given with one standard deviation precision. In comparison to fallout, the average process-produced plutonium might have a ^{240}Pu/^{239}Pu atom ratio between 0.051 (Krey et al. 1976, pp. 671-678) and 0.067 (ERDA 1977, p. 3.8). For subsequent calculation purposes, a ^{240}Pu/^{239}Pu atom ratio of 0.064 will be assumed for process-produced plutonium, a ratio corresponding to 6 atom percent ^{240}Pu and 93.5 atom percent ^{239}Pu.

Even though there is a significant difference between the ^{240}Pu/^{239}Pu atom ratio for fallout (0.18) and process-produced (0.064), the presence of the two sources in the soil might go undetected. Detection and characterization of plutonium in soil samples are needed to relate airborne concentrations and atom ratios to resuspension source characteristics. Characterization could be hindered because of the large depth used in most surface soil sampling.

The objective is to examine the sensitivity of soil surface-depth samples to yield $^{240}Pu/^{239}Pu$ atom ratios, which confirm the presence of process-produced plutonium in the soil. For the examination, a graphical representation is presented showing the relationships between assumed average activity densities for both fallout and process-produced plutonium and the average $^{240}Pu/^{239}Pu$ activity densities.

Calculations

The average $^{240}Pu/^{239}Pu$ atom ratios for mixtures of fallout and process-produced plutonium is dependent on the radioactive decay half-lives for each radionuclide. The half-lives are significantly different, 24,360 years for ^{239}Pu and 6,580 years for ^{240}Pu. For calculational convenience the equations for calculating the average atom ratio that follow are based on assumed activity densities for each radionuclide. The end product of these calculations is an average activity density for both radioisotopes and an average atom ratio. Ultimately the calculational process will be reversed for interpreting average surface sample data. The number of atoms, N, of each radionuclide is calculated from

$$N_i = -\frac{t_{1/2}(dN/dt)}{\ln 2} \qquad (1)$$

where $t_{1/2}$ = half-life, and dN/dt = decay rate for radionuclide i in sample. Thus, the average atom ratio in the sample is

$$^{240}Pu/^{239}Pu = \frac{N_{240}}{N_{240} + N_{239}}. \qquad (2)$$

Activity densities are calculated from the atoms present divided by the sample weight.

Results

Relationships between activity-densities and $^{240}Pu/^{239}Pu$ atom ratios are shown graphically in Figure 1. The ordinate is the activity-density for process-produced and the abscissa is the total of both process and fallout plutonium. Activity-densities for fallout are shown by dashed curves intersecting average $^{240}Pu/^{239}Pu$ atom ratios shown by solid lines. The solid lines also represent constant percentages of the total activity density, percentages corresponding to process-produced plutonium content.

Samples with $^{240}Pu/^{239}Pu$ atom ratios less than 0.18 for fallout contain a large fraction of process-produced plutonium. The percentage increases as atom ratios decrease. For instance, consider a sample of 17.8 fCi/g total activity density and a 0.104 $^{240}Pu/^{239}Pu$ atom ratio (Price and Dirkes 1981). From the detailed calculations for Figure 1, the sample contains 10.45 fCi/g of process-produced and 7.25 fCi/g of fallout plutonium. The process-produced activity density is 59 percent of the total activity density.

Conclusions

Plutonium-240/plutonium-239 atom ratios are used to identify the relative contributions of plutonium from two sources, both sources contribute to resuspension. However, the relative resuspension characteristics for both sources is unknown. If any resuspendability difference exists, both components must be identified. To assist identification ease from sample atom ratio analyses, average atom ratios were calculated from mixtures of fallout and process-produced sources. The calculations were made with $^{240}Pu/^{239}Pu$ atom ratios of 0.18 for fallout and 0.064 for process-produced plutonium. Both ratios could have variations within local study areas near nuclear plants, but fallout variations are minor compared to variations in process-produced plutonium.

If several surface samples are analyzed, the presence of process-produced plutonium may be indicated if there is any significant change in the $^{240}Pu/^{239}Pu$ atom ratio, changes as small as 0.01. For the range of variables investigated, the contribution of process-produced plutonium to the total plutonium content ranged from 6.6 to 10 percent for each 0.01 variation in the $^{240}Pu/^{239}Pu$ atom ratio. Even if $^{240}Pu/^{239}Pu$ variations were absent, the $^{240}Pu/^{239}Pu$ atom ratios could still reflect a uniform resuspension source from both fallout and process plutonium. In this case, however, additional surface samples would be required to further define the fallout $^{240}Pu/^{239}Pu$ atom ratio. These additional samples would be collected at significant, but yet close, distances to the study area. Fallout atom ratios should be determined from collection sites at which the absence of process-produced plutonium can be ensured.

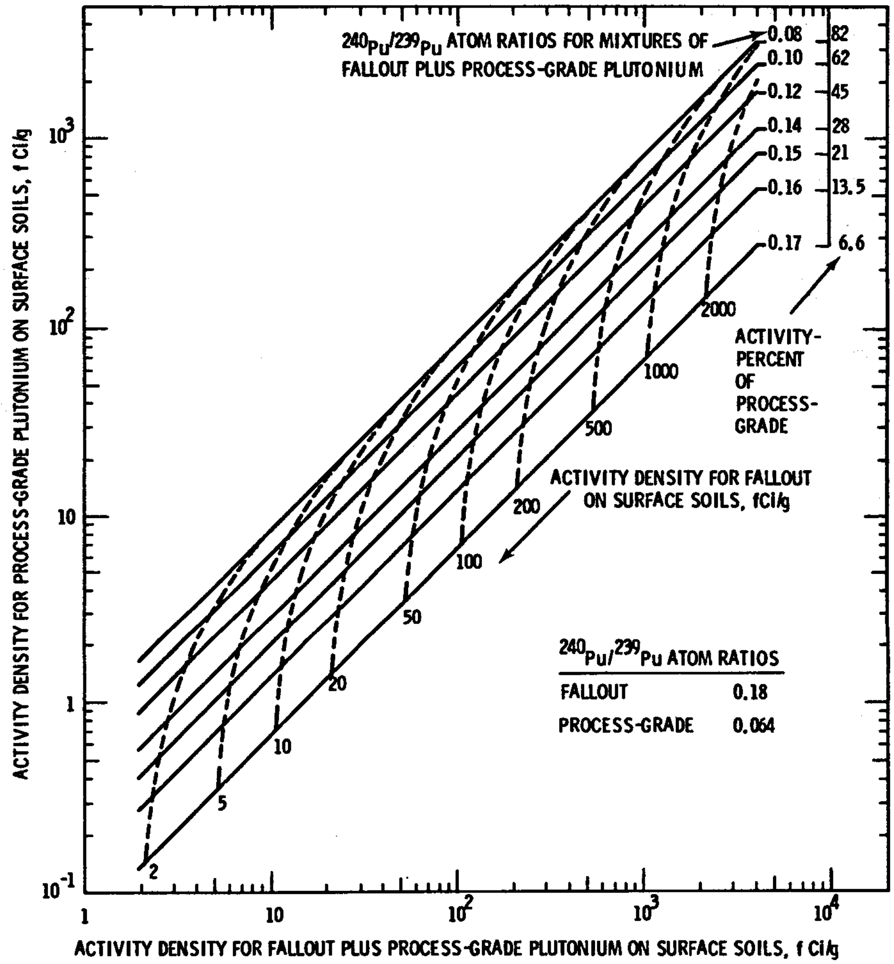

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FIGURE 1. Plutonium-240/Plutonium-239 Atom Ratios for Mixtures of Nuclear Weapons Fallout Plus Process-Grade Plutonium

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PLUME DEPLETION BY PARTICLE DRY DEPOSITION IN A FORESTED CANYON

G. A. Sehmel

Airborne particulate removal rates by dry deposition are a function of only particle size when other controlling parameters are held constant. Removal rates are minimum, and nearly constant, for a particle diameter range from about 0.5 to 1 µm. This diameter range can be estimated from either a summary of experimental field investigations (Sehmel 1980) in which polydispersed particle sizes were used or from model predictions (Sehmel and Hodgson 1980, Sehmel 1980) based upon wind tunnel experiments with monodispersed particles. Results from field experiments have not been generalized. In contrast, wind tunnel results have been generalized into a predictive dry deposition velocity model. Model predictions have greatly increased the ability to more accurately predict plume depletion. These model predictions are now being generally used to predict dry deposition removal rates, i.e., the particle dry deposition velocity. Nevertheless, continued improvement in model predictions are needed especially for nonuniform deposition fetches, fetches with a large vegetative canopy typical of environmental surface variations.

Investigators using particle dry deposition velocities are confronted with two major predictive qualifications in selecting particle dry deposition velocities. These are:

- The pollutant plume consists of an aerosol containing a particulate size distribution, rather than monodispersed particles. Conceptually, removal rates for size distributions are calculated by summing the deposition velocities for each size fraction.
- Even when the particulate size distributions, and changes in size distributions as a function of downwind distance are known or predictable, dry deposition predictions are often qualified because deposition fetches are nearly always nonuniform. These fetch variations are often ignored, or if considered, the investigator must still assume an average deposition velocity or apply integration techniques over those fetch variations.

The objective here is to develop an integrated plume depletion data-base for a large vegetative canopy, a forested canyon.

Experiment

Particle dry deposition was measured with dual-tracers during nocturnal drainage flows in a forested canyon. The canyon was along Icicle Creek near Leavenworth, Washington. Both the tracer release and sampling sites were adjacent to the creek. The tracers were non-depositing SF_6 gas and depositing lithium-traced particulates. Both tracers were released at a height of 2.6 m. The lithium-traced particles were generated from a water solution of Li_2CO_3 sprayed through nozzles. The SF_6 gas tracer was released from tanks in which the SF_6 was in the gas phase.

Airborne tracer concentrations were measured downwind at two sampling sites, site 1 closest to the tracer release and site 2 further downwind. At each site, airborne lithium-traced particle concentrations were measured in real-time with flame ionization detectors, and samples of airborne SF_6 concentrations were collected nearly instantaneously with syringes. These syringe samples were analyzed for SF_6 concentrations the following day.

Plume depletion at respirable height was measured between the two downwind sampling stations. Site locations were changed in three succeeding experiments on May 26, 27, and 28, 1981. The first downwind site was either 0.9 or 1.0 km from the tracer release location, a distance to allow spray droplet evaporation and to decrease electrical charge on the generated particles. The second site was 1.2 to 1.7 km further downwind. All distances are those measured along the road.

There are two lithium particle detectors at each site. Each detector response is a function of particle diameter. The detector placements (instrument numbers 31 and 94 at site 1 and 93 and 95 at site 2) were selected for the optimum data comparison between sites 1 and 2. Instrument response for number 31 at site 1 was compared to response for number 93 at site 2. Similarly, number 94 at site 1 was compared to number 95 at site 2.

The lithium-particle detectors were calibrated in a wind tunnel with both the poly-dispersed particles generated with the spray nozzles and monodispersed test aerosols. The calibration characteristics of each lithium-particle detector are shown in Figure 1 as a function of monodispersed tracer-particle diameter (the detectors were calibrated by

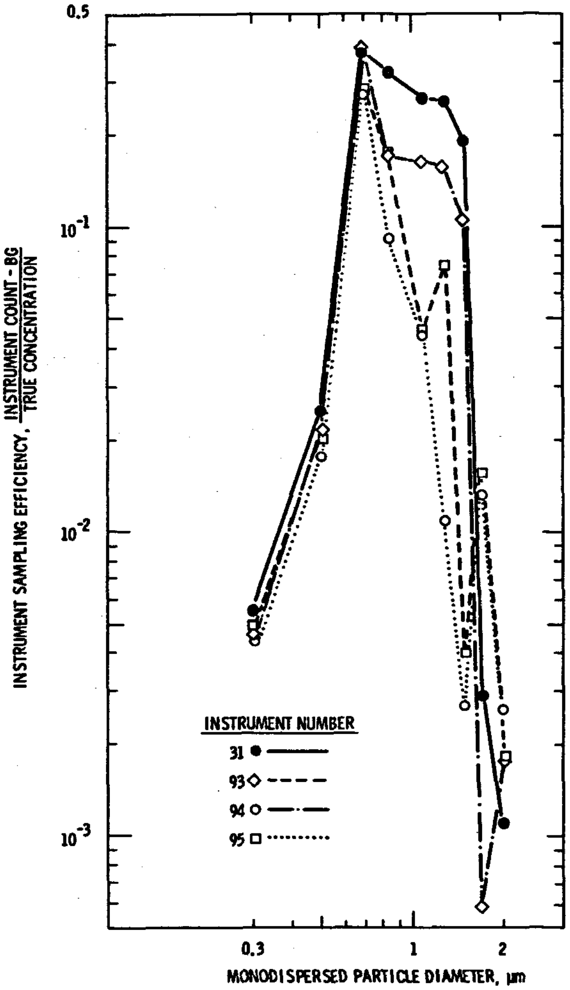

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FIGURE 1. Lithium Detector Calibrations with Monodispersed Lithium Carbonate-Uranine Particles

Sehmel and Hodgson (1981) in a wind tunnel with the true concentration determined by isokinetic sampling). Instrument responses are maximum for a particle diameter of about 0.7 µm. Responses decrease two orders of magnitude outside the range of about 0.3 to 1.5 µm.

Pairs of instruments have similar calibrations; i.e., instruments 94 and 95 are similar, likewise numbers 31 and 93. Instrument numbers 31 and 93 have a significantly greater response for particle diameters in the range from about 1 to 1.5 µm. To emphasize these differences, particle diameter response ranges are desirable that are less than the 0.3 to 2 µm calibration range. Diameter ranges of 0.6 to 1.0 and 0.6 to 1.5 µm were selected. These correspond to greater than eight percent sampling efficiency for instruments 94 and 95 and for instruments 31 and 93, respectively.

Calculations

Particle plume depletion at respirable height was calculated. Depletion is calculated from concentration ratios at each site,

$$\text{Ratio}_i = \frac{p_i}{g_i} \qquad (1)$$

where p is the particle concentration, g is the gas concentration, and the subscript i is the sampling location. This ratio is constant with distance near the plume centerline if there is no particle deposition. In this case, the conserved particle concentration at sampling site number 2 is

$$p_{2c} = p_1\,(g_2/g_1). \qquad (2)$$

When dry deposition occurs, the measured particle concentration at the second site, p_2, is less than the calculated conserved-concentration, p_{2c}.

Thus, the fractional plume depletion is:

$$\frac{p_{2c} - p_2}{p_{2c}} = 1 - \left(\frac{p_2 g_1}{g_2}\right)\left(\frac{1}{p_1}\right). \qquad (3)$$

Results

Of the three experimental time periods, the results of the May 28 experiment were most ideal for calculating plume depletion by dry deposition. Concentrations for May 28 are shown in Figure 2 for site 1 (before dry deposition on the right portion of the figure) and site 2 (after dry deposition on the left side of the figure). Average concentrations were calculated for the indicated integration intervals. These averages were used in Equation 3 for calculating plume depletion. Plume depletion at respirable height was 42 percent for the 0.6 to 1.0 µm diameter range and 74 percent for the 0.6 to 1.5 µm diameter range.

To be noted are the elevated SF_6 concentrations on the order of 1 and 10^4 pptv measured before the tracer release. These elevated concentrations may have been caused by a leaking valve at the SF_6 release site. These elevated concentrations should have a negligible

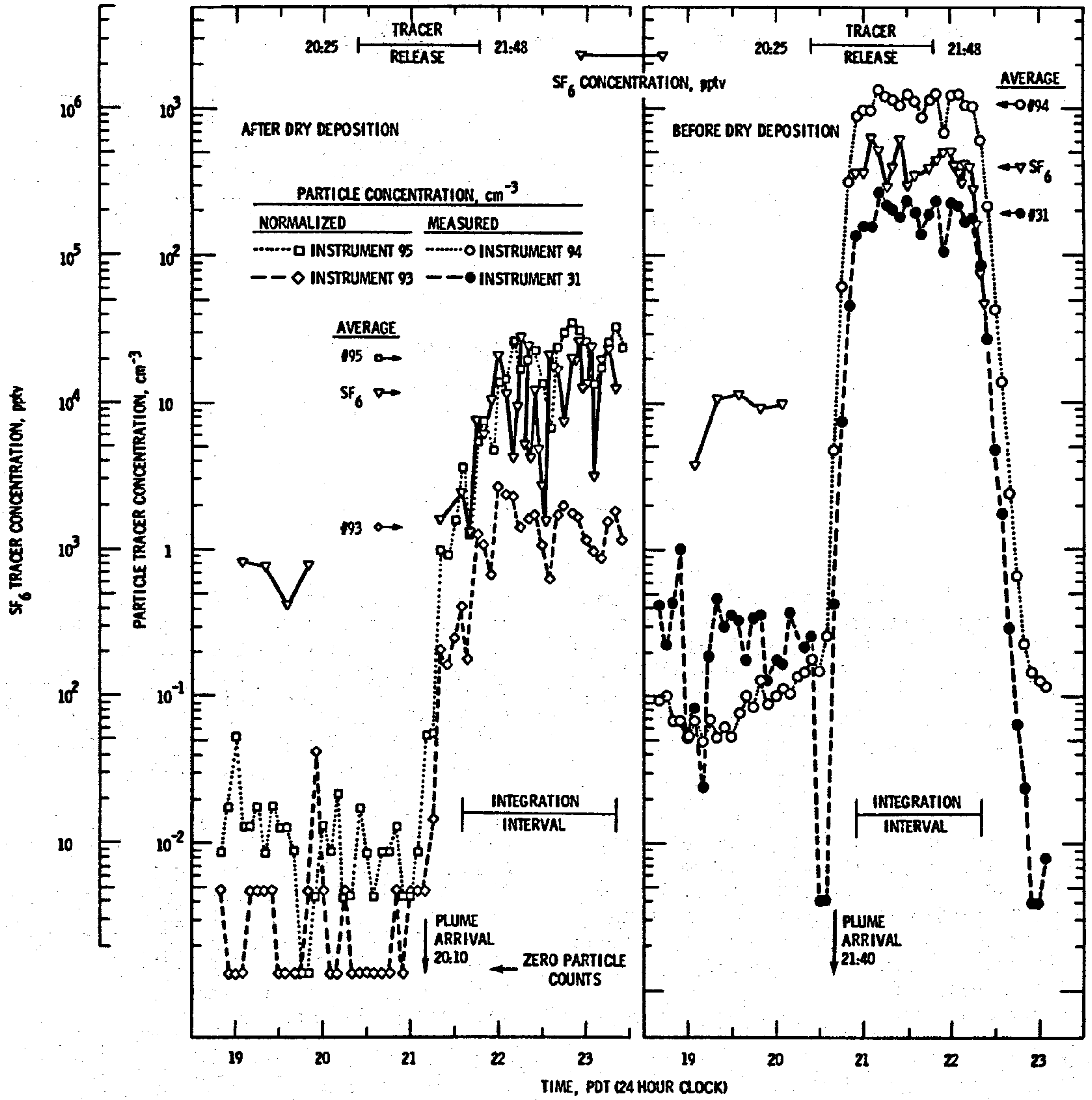

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FIGURE 2. Airborne Dual-Tracer Concentrations in a Forested Canyon, May 28, 1981

effect on plume depletion calculations since SF$_6$ concentrations were over one order of magnitude greater during the integration intervals.

Tracer concentrations were lower on the prior two nights, May 26 and 27. These lower and less steady tracer concentrations nevertheless resulted in comparable plume depletion. Results for all three nights are summarized in Table 1.

Conclusions

Particulate plume depletion at respirable height ranged from 42 to 74 percent across distances of 1.2 and 1.7 km in a forested canyon. These plume depletions are significantly greater than would be predicted using the state-of-the-art technology on particle dry deposition velocities as a boundary condition for predictive meteorological transport, dispersion and deposition removal

TABLE 1. Particle Dry Deposition Removal at Respirable Height in a Forested Canyon, 1981

Experimental Time Periods	Distance from Tracer Release Site (km)		Distance for Dry Deposition Between Sites 1 and 2 (km)	Tracer Release Time (PDT)	SF_6 Release (kg/hr)	Average SF_6 Concentration During "Steady" Plume Passage (PPTV)		Particle Plume Depletion at Responsible Height Between Sites 1 and 2(%)	
	Site 1	Site 2				Site 1	Site 2	0.6 to 1.5 μm Diameter Particles	0.6 to 1.0 μm Diameter Particles
Evening of May 26	0.9	2.2	1.2	20:01 to 21:22	10.8	4.6×10^3	2.7×10^2	65	44
Evening of May 27	1.0	2.6	1.6	19:15 to 20:33	10.8	6.9×10^2	4.2×10^2	(a)	73
Evening of May 28	0.9	2.6	1.7	20:25 to 21:48	10.5	4.0×10^5	1.2×10^4	42	74

(a) Sensor Problems

models. The dual-tracer plume-depletion data are sufficiently reproducible to now warrant more intensive investigations of removal processes in vegetative canopies, investigations considering integrated-deposition across the entire vegetative canopy and variations in the vegetative canopy. Admittedly, the experimental technique could be improved to also include measuring concentrations as a function of height, measuring residual electrical charge on the particles and extensive measurements of meteorological parameters. Although improvements could be made, the important conclusion is that plume depletion ranged from 42 to 74 percent for the particle diameter range of minimum expected dry deposition removal.

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Oil Shale

OIL SHALE

- **Oil Shale Fugitive Air Emissions**
- **DOE/RL Special Studies**

The primary oil shale regions of the country are in the Rocky Mountain West where any airborne pollutants will be emitted, transported, transformed and deposited over very complex terrain under meteorological conditions that are, at best, extremely complicated. This complexity dictates the development of a set of complex models and innovative field techniques.

PNL has assessed the future research needs in this area and has formulated an experimental design for atmospheric transport studies over the oil shale region. Other activities include:

development of a tracer technique to measure dry deposition of pollutants over complex areas

deposition measurements of ambient airborne soil

pollutant mixing in valleys

simulation of fugitive emissions from a proposed spent shale disposal site

aerosol and visibility measurements over complex terrain.

• Oil Shale Fugitive Air Emissions

Objectives of this study are:

Evaluating fugitive airborne particulate and gaseous emissions from the present and future oil shale activities.

Investigating emissions from mining, transportation, material handling, crushing, retorting, and spent shale disposal sites.

Identifying and classifying pollutants such as carbon monoxide, hydrogen sulfide, oxides of nitrogen, ozone, sulfur dioxide, hydrocarbons, and suspended particles.

A DUAL-TRACER EXPERIMENT TO INVESTIGATE POLLUTANT TRANSPORT, DISPERSION, AND PARTICLE DRY DEPOSITION AT AN OIL SHALE SITE

G. A. Sehmel

Atmospheric transport and plume depletion investigations were conducted in August, 1980, with the use of dual-tracers at the Federal Oil Shale Lease Tract C-a operated by Rio Blanco in Colorado. The objectives of the experiments were to simulate pollutant transport, dispersion, and plume depletion by particle dry deposition in valley nocturnal-drainage flows. The tracers were nondepositing SF_6 gas and depositing lithium-traced particles, i.e., Li_2CO_3.

This dual-tracer, real-time measurement technique was also directed toward measuring the minimum decrease in respirable airborne particle concentrations with distance. The minimum was measured because of the particle diameter range used, about 0.5 to 1.5 μm, a size range in which the dry deposition velocity is minimum (Sehmel 1980) and nearly independent of particle diameter. Particle dry deposition removal or plume depletion is calculated from the relatively more rapid downwind decrease of airborne tracer particle concentration referenced to the decrease of the tracer gas concentration.

These field experiments were conducted jointly with other investigators at the Pacific Northwest Laboratory and Los Alamos National Laboratory (LANL). Data volumes for both tracer and meteorological data for all investigations were published, Whiteman et al. (1981) at PNL and Clements et al. (1981) at LANL. The purpose here is to abstract the PNL tracer results (Sehmel 1981).

Experiments

The tracer experiments were in a valley (Corral Gulch) during four nocturnal drainage flows and one daytime release. Dual tracers were used during three nights, while only SF_6 was released during one night and the daytime release. Insufficient data for analysis were obtained for the daytime release since air bag samplers were not operated and airflow only occasionally delivered SF_6 tracer to the real-time sampling sites.

Tracers were released for an hour so downwind transport within the drainage flow would allow the tracer to be carried through the length of the sampling station grid. Sampling stations were located along multiple lines perpendicular to the valley axis at various downwind distances from the source. The lines extended across the valley floor and up the valley sidewalls.

Airborne tracer concentrations downwind of the tracer release point were measured using one of three methods:

- Samples of SF_6 and lithium-traced particles were collected and detected in real-time, called site PNL-1 at 1.6 km and site PNL-2 at 2.7 km from the tracer release site. The sampling height was 1.5 m.
- After tracer generation had ended, syringe samples of ambient air were collected from a vehicle driven along roads that crossed the gulch approximately 6.8 km below the tracer release site.
- Cross-gulch samples of SF_6 were collected using bag samplers that were distributed

along four sampling lines oriented perpendicular to the valley axis at distances of 2.0, 2.9, 4.8, and 5.9 km downgulch from the tracer release site. Each sampling line was operated by radio control to actuate selected sampling pumps. The sampling time was usually 10 minutes. Each of the lines consisted of 7 or 8 cross-gulch spaced sampling stations, 3 or 4 on the gulch floor and 2 on each sidewall. Each station was set up to fill three air sampling bags, each bag was attached to a separate air pump. At each sampling station, airborne SF_6 concentrations were investigated as a function of either succeeding sampling times or as a function of height for a single sampling time interval.

Results

Airborne tracer concentrations were summarized (Sehmel 1981). Concentrations for SF_6 tracer are reported as parts by volume; i.e., 10^{-12} parts by volume equals 1 ppt, parts per trillion. Concentrations for lithium-traced particles are reported as airborne particles/cm^3. Ratios of particle to gas concentrations were calculated to determine plume depletion by particle dry deposition.

Tracer concentrations as a function of time were different than might be predicted from limited meteorological measurements, i.e., wind speed measured at the tracer release site. In comparison, the tracer arrival speed at respirable height is defined as the distance between the release and tracer measurement site divided by the time between tracer release and first sensing the tracer at the real-time tracer sampling sites, distances at PNL-1 and PNL-2 of 1.6 and 2.7 km respectively. As shown in Table 1, tracer-arrival speeds were always greater than the wind speed at the release site and always greater at site PNL-2, compared to site PNL-1. Tracer arrival speeds being greater than the wind speed reflect:

TABLE 1. Tracer-Arrival Speeds

Night of August, 1980	Wind Speed, Release Site at 1.5 m height, m/s	Tracer-Arrival Speed at 1.5 m height, m/s Site PNL-1	Site PNL-2
5 - 6	1.3	2.7	3.2
7 - 8	0.9	2	2.8
9 - 10	1.1	2.7	2.8
11 - 12	1.1	3.0	3.0

- both horizontal transport and vertical mixing of the tracer. Tracer mixed to higher elevations is transported at greater wind speeds as compared to wind speeds at lower elevations.

- increased drainage flow with increasing downslope distance within the gulch.

Tracer concentrations as a function of the cross- and down-gulch areal extent reflect the following trends:

- Nonsteady-state nocturnal drainage flows can always be expected in complex terrain.

- Concentrations can be significantly different along a cross-gulch traverse.

- At gulch bends, the airflow inertia tends to cause air to flow up the gulch wall as compared to remaining at the bottom of the gulch.

- At the confluence of two "steep-walled" gulches, pollutants do not mix rapidly across the downwind gulch.

- Pollutants are not contained within the gulch, but spread out over the hills forming the gulch.

Plume depletion is calculated from the decrease in ratios, the tracer particle concentration/SF_6 concentration ratio, between the two real-time tracer measurement sites, i.e., between PNL-1 and PNL-2 sites.

Plume depletion at respirable height was calculated from the dual-tracer concentrations for the two nights for which both tracers were continuously measured. The average plume depletion was 68% for the August 5-6 experiment, with a maximum uncertainty range from 62% to 73% based upon the range of uncertainty in the lithium detector calibration factors. On August 11-12, the average plume depletion was 37% with a maximum uncertainty range from 13% to 62%.

Conclusions

The SF_6 tracer data yield further insight into predicting airborne pollutant concentrations during drainage flows in complex terrain. The accurate prediction of pollutant concentrations is the environmental concern, rather than average wind fields. Although there may never be enough meteorological measurement sites to completely define wind fields, tracer concentrations do reflect integrated wind field changes and diffusion.

Airborne tracer concentrations in local drainage flows are complex for the site investigated. The overall gulch drainage flow is a combination of multiplicity of drainage flows, flows from successively larger gullies forming the gulch tributaries. Each tributary can have a drainage wind which rapidly loses its identity when mixing with other drainages. However, when the tributary is large, a gulch, the mixing is much less rapid. For instance, mixing was usually incomplete at the confluence of Corral and Box Elder Gulches.

Predicting, or monitoring, air concentrations at valley confluences can be uncertain. For instance, is the sampling site dominated by air flow from only one of the two drainages? Possibly the effects of drainage dominance might be reduced by predicting, or monitoring air concentrations at a predictable down-gulch "mixing" distance. The mixing distance might be equal to several gulch widths prior to confluence. The mixing distance will be estimated. Sampling line 3 was about 2 km downwind of the confluence of Corral Gulch (the tracer release gulch) and Box Elder Gulch. Since the tracer plume appeared mixed in the cross-gulch direction at sampling line 3, this result is used to estimate the mixing distance. Upwind of the gulch confluence, the "widths" of Corral and Box Elder Gulches are about 0.3 and 0.1 km. Thus, the mixing-distance/gulch-width ratio ranges from 7 to 20, 2/0.3 to 2/0.1.

A tentative generalization is that cross-gulch interruptions of drainage flows in complex terrain are the norm, rather than the exception. Drainage flows were not uniform for the four drainage flows investigated. Significant cross-gulch winds always interrupted the drainage flows, even for average drainage wind speeds less than 1.3 m/sec measured at 1.5 m at the release site and tracer arrival speeds less than 3 m/sec. The drainage flow interruptions, however, did not extend the length of the gulch. Although important in gulches, cross-valley winds may be of less significance for deep valleys.

Significant vertical mixing of the tracer occurred as reflected in measured wind speeds and calculated tracer-arrival speeds. The tracer-arrival speeds were more than twice the wind speed measured at the tracer release site, measured at 1.5 m. Obviously, the height at which wind speed is measured is important. Predictive meteorological transport model development should be based on winds measured at greater heights, or appropriate extrapolations must be made as a function of height.

Particle plume depletions at respirable height (1.5 m) were significant. Depletion ranged from 37% to 68% across a distance of only 1.2 km. These depletion results are especially significant since less than a few percent depletion would be predicted based upon meteorological transport models which incorporate a dry deposition velocity to describe particle removal.

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ATMOSPHERIC TRACER GAS INVESTIGATIONS IN THE COLORADO OIL SHALE REGION

G. A. Sehmel

Pacific Northwest Laboratory (PNL) conducted atmospheric transport and plume depletion investigations in August, 1980, in Corral Gulch, down gulch from the Rio Blanco modified in-situ oil shale retort at Federal Lease Tract C-a in Colorado (Sehmel 1981). Sulfurhexafluoride tracer gas field experiments were also conducted in July, 1981 but for an entirely different terrain. The terrain was selected to simulate fugitive-emissions transport from a proposed spent-shale disposal site. The terrain was a sloping, undulating area between the hilltop along Corral Gulch and Dead Horse Ridge. The objective of these experiments was to investigate atmospheric transport during both day and nighttime conditions.

Experiments

During each experiment, the SF_6 tracer gas was released at a constant release rate of 26 kg/hr (12 lb/hr). To ensure that the tracer would have had time to reach the most remote sampling locations, it was released for about one hour before tracer sampling was begun.

The total tracer release times ranged from 3 to 9 hours. There were four experimental time periods, two in the evening and two in the morning. The evening time periods were for tracer release from 1500 to 2100 MDT (July 15) and 1800 to 2100 MDT (July 17). The morning time periods for tracer release were from 0330 to 0930 MDT (July 19) and 0300 to 1217 MDT (July 21).

Total release times were selected to include sunset and sunrise. The evening experiments were conducted first. The results for the evening experiments were less successful than for the morning releases. On the first night, the radio control system malfunctioned for automatically collecting air samples at remote sites. A power lead in the battery supply had shorted. On the second night the system was operational, but the experiment was stopped due to the safety hazards from a severe local storm.

Large tracer concentration data sets were obtained during the two morning tracer releases. Airborne SF_6 tracer concentrations were sampled by both radio-controlled sampling stations (15-minute sampling times) and syringe-grab-samplers (near instantaneous) sampling times. There were 32 radio-controlled sampling stations which encompassed the release site. These sampling stations were located at sites with road access. Distances from the tracer release site to these sampling stations ranged from 0.6 to 6 km (0.4 to 4 miles). The syringe-grab-samples were also collected along roads. Most syringe samples were collected at distances of 3 to 13 km (2 to 8 miles) from the release site.

Conclusions

The tracer and meteorological data are yet to be analyzed. Expected results will include information on:

- site-specific cross-wind diffusion, σ_y, at respirable height
- a time-history of pollutant concentration changes in both gulches and other terrain before, during, and after sunrise
- identification of major transport flow paths in the evening and morning
- pollutant transport over a major ridge to an adjacent drainage valley, Dead Horse Ridge to Big Duck Creek
- pollutant mixing and channeling in valleys and downwind of valley confluences
- the transient behavior of pollutant flows
- nocturnal drainage flows acting like a dense gas layer containing the tracer and flowing along terrain depressions.

The inappropriate selection of air pollution monitoring station sites, heights and concentration averaging times could make assessment applications toward PSD regulations of doubtful accuracy.

Reference

Sehmel, G. A. 1981. "A Dual-Tracer Experiment to Investigate Pollutant Transport, Dispersion, and Particle Dry Deposition at the Rio Blanco Oil Shale Site in Colorado." PNL-SA-9327, pp. 137-146, Proceedings of the Second Conference on Mountain Meteorology, November 10-13, 1981, Steamboat Springs, Colorado. American Meteorological Society, Beacon Hill, Massachusetts.

AIRBORNE TRACER CONCENTRATIONS AS A FUNCTION OF TIME AND DISTANCE IN A GULCH

G. A. Sehmel

Regulatory standards for permissible atmospheric emissions and downwind pollutant concentrations are often qualified scientifically because of uncertainties in model predictions. Predictions are especially uncertain when pollutants are released in complex terrain. Experimental data were collected in August 1980, which reflect these uncertainties during nocturnal drainage flows within Corral Gulch in the oil shale region of Colorado. Airborne concentrations of SF_6 tracer gas were measured as a function of time and downwind distance. These data were obtained by Clements et al. (1981) and Sehmel (1981) in separately sponsored research. Some of the SF_6 concentration data were obtained as a function of time at four sampling sites along the gulch. Cross-comparisons of these data can be used to reflect air and pollutant flows. The purpose here is to make these comparisons between reports.

Experiments

A gaseous SF_6 tracer was released near the surface during four nocturnal drainage flows. The release rates were 5.4 kg/hr on the first three nights and 3.0 kg/hr on the fourth night. Airborne concentrations were measured down-gulch at four sampling sites, two operated by the Pacific Northwest Laboratory (PNL) and two by the Los Alamos National Laboratory (LANL). All four sites were along the bottom of the gulch, at distances from 1.6 to 6.0 km downwind from the tracer release point. The respective distances were 1.6 km at PNL-1 (near visitors center), 2.7 km at PNL-2 (near Met-3 station), 3.3 km at LANL-1, and 6.0 km at LANL-2.

Airborne SF_6 tracer concentrations were measured as a function of time at each of these four sampling sites. The sampling times were different, but similar enough for cross comparing concentration data. The PNL concentrations were nearly instantaneous and were measured in the field with portable gas chromatographs (GC). In comparison, concentrations reported by LANL were obtained from bag samples; each concentration was an integrated average reflecting concentrations during the 7.5 min of bag sample collection.

Results

Airborne SF_6 gas concentrations, parts by volume, are shown as a function of time (MDT) for the four measurement sites in Figures 1 through 4, the nights of August 5 to 6, 7 to 8, 9 to 10 and 11 to 12, respectively. Concentrations are shown before, during and after tracer release.

Concentrations seldom reflected steady air flow and usually exhibited transient behavior. During the main transient plume passages, rates of initial concentration increase were greater than rates of final concentration decrease. Significant concentration transients also occurred during times of maximum concentrations, times on the order of one hour. These transients often reflect cross-gulch air flow. Maximum effects of cross-gulch flow decreasing airborne concentrations include the following locations and times:

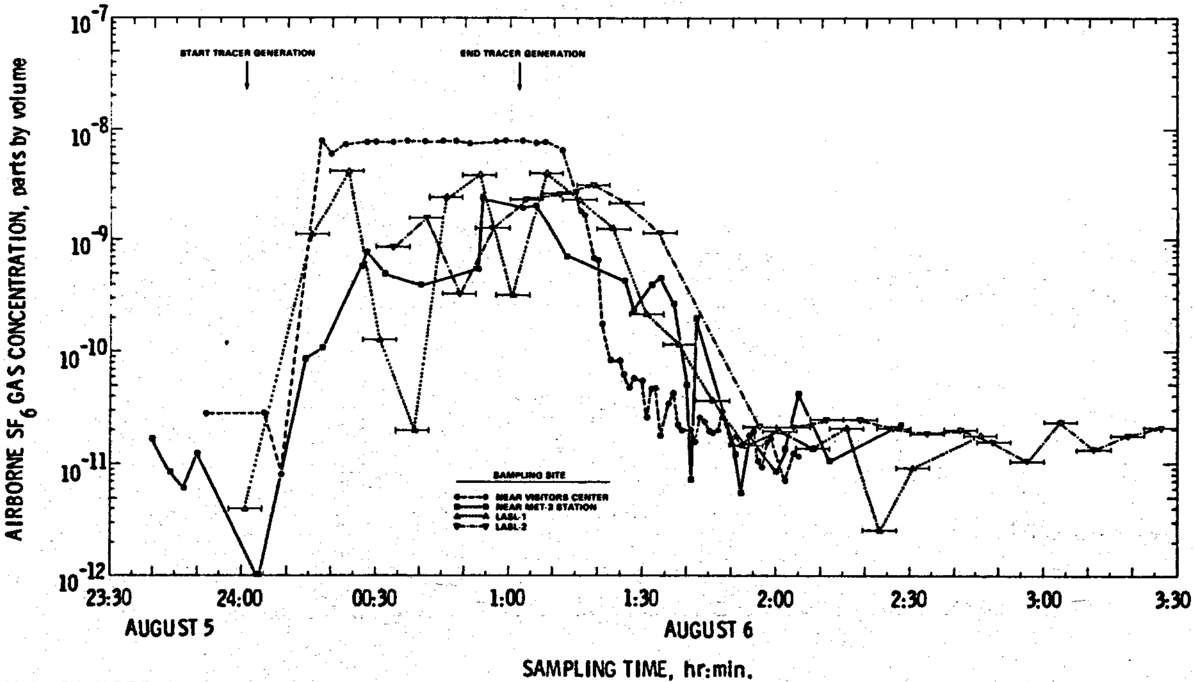

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FIGURE 1. Airborne SF_6 Tracer Gas Concentrations the Night of August 5 to 6, 1980

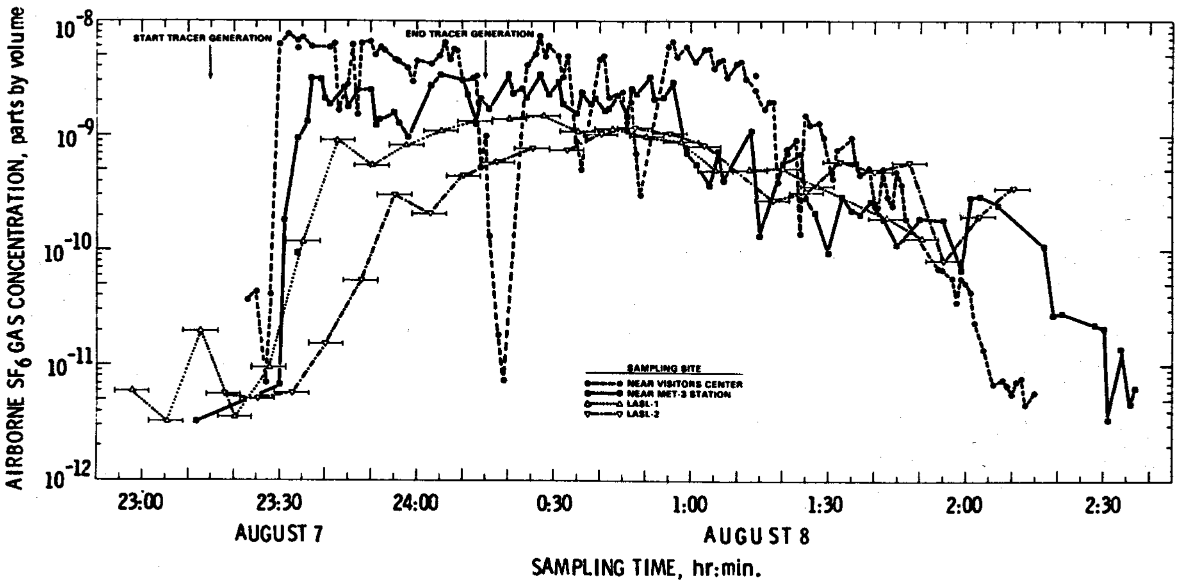

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FIGURE 2. Airborne SF$_6$ Tracer Gas Concentrations the Night of August 7 to 8, 1980

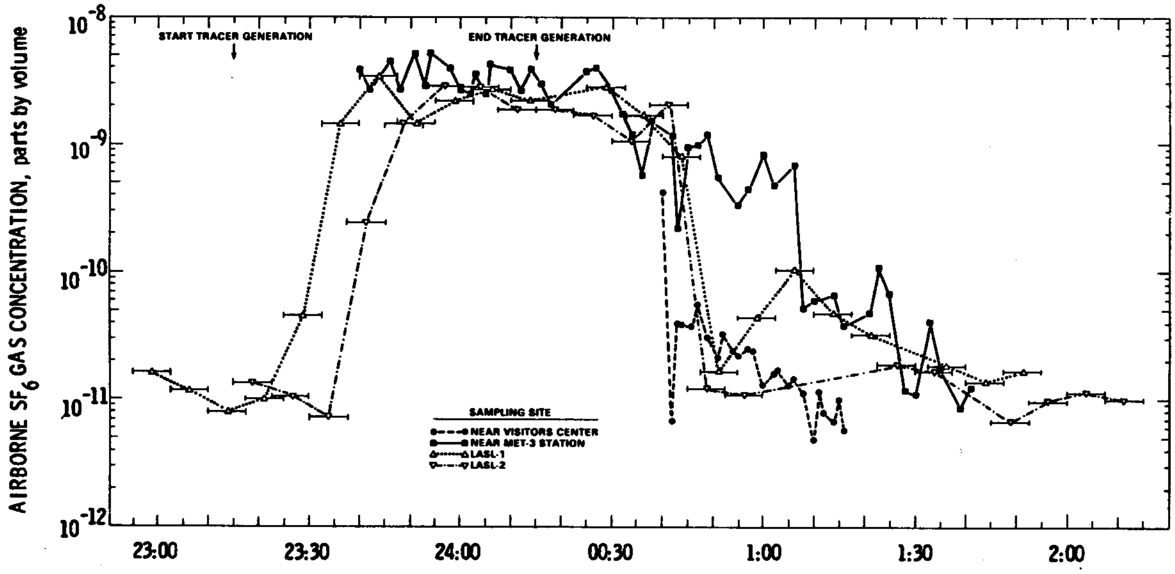

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FIGURE 3. Airborne SF$_6$ Tracer Gas Concentrations the Night of August 9 to 10, 1980

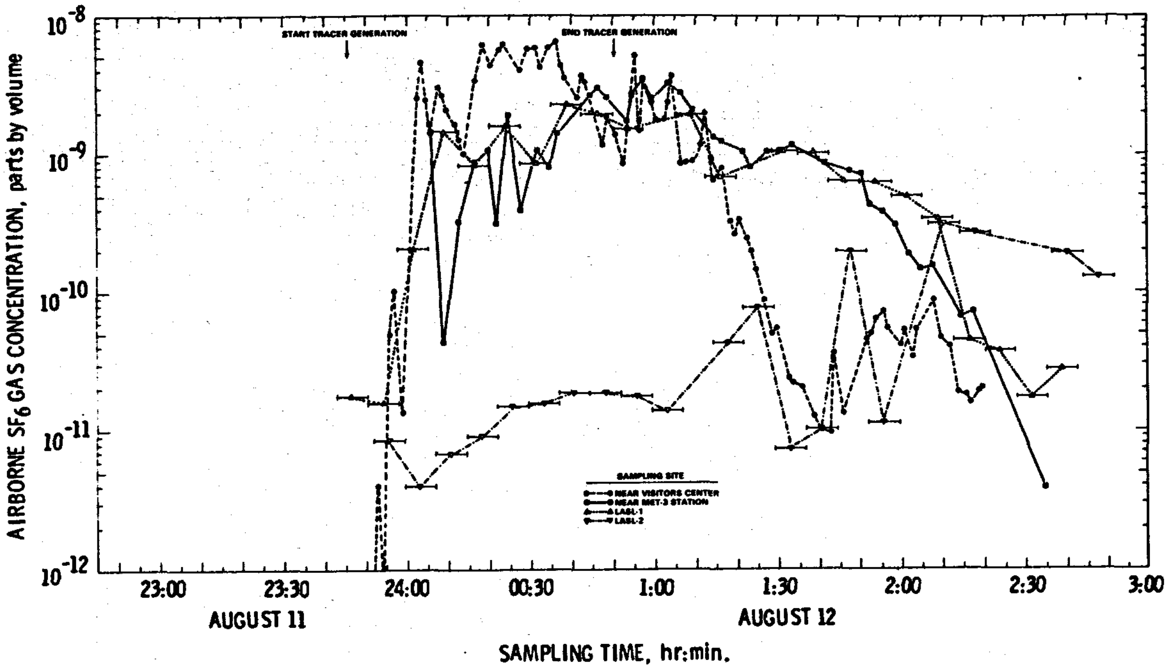

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FIGURE 4. Airborne SF_6 Tracer Gas Concentrations the Night of August 11 to 12, 1980

- August 5 to 6: LANL-1 after 00:34
- August 7 to 8: PNL-1 at 00:19
- August 11 to 12: LANL-2 from 00:00 to 01:00

Concentration changes as a function of distance are also complex, not a simple concentration decrease with increasing distance. The complex changes as a function of distance include the following on August 5 to 6. During times near 00:20, concentrations at PNL-2 (1.6 km) are less than at LANL-1 (3.3 km). PNL-2 is near the confluence with a second main gulch. On this night, concentrations at PNL-2 were low because of partially sampling untraced air coming from the second gulch, Box Elder Gulch.

Conclusions

Significant transient concentration changes occur within a 4.4-km long section of a gulch (6.0 - 1.6 km) even for nocturnal drainage flows. Major transients are caused by cross flows penetrating into and mixing with the drainage flow. Predictive models for average airborne concentrations in the gulch could be based partially upon the frequency of crosswinds. Although these cross-flows can decrease concentrations in the gulch, it is questionable whether cross-gulch effects need to be incorporated into predictive atmospheric transport and diffusion models. In addition, the frequency of these cross-wind penetrations is unknown. If penetration frequency were pursued for model development, the frequency would need to be evaluated by a spacial integration measurement technique, a tracer technique rather than only an anemometer measurement system.

Possibly, models should be based upon conservative concentration estimates, i.e., the maximum expected concentration in a gulch. These maximum concentrations can be estimated using the tracer concentrations normalized by the release rate data.

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• DOE/RL Special Studies

Objectives of these studies are:

Collecting data from an airborne platform on scattering extinction coefficients, relative humidity, sulfate and trace metals over mountainous terrain in the Northwest.

Estimating trace metal budgets over an area of complex terrain.

POLLUTION BUDGETS WITHIN A REGIONAL WATERSHED

R. N. Lee, M. M. Orgill, D. R. Drewes and J. M. Thorp

In the early summer of 1980, an aircraft and surface sampling program was conducted in Washington State to assess the transport, metamorphosis and removal of atmospheric contaminants in a region characterized by complex terrain. The initial examination of aerosol data revealed enrichment of trace elements such as Pb, Cl, S, Zn and Br in the collected samples. Transport of anthropogenic materials from an upwind source area, vehicular emissions and emissions from Mt. St. Helens were suggested to account for the observed enrichment of specific trace elements (Lee and Orgill 1981). Analysis of field data was completed during FY81.

Data interpretation was based on the simple box model shown in Figure 1. The inflow region was defined by the airborne sampling program conducted east of the Seattle-Tacoma metropolitan area and the outflow region by airborne sampling east of the Cascade Crest. Concentration profiles and available wind data were employed to calculate the fluxes of trace elements and estimate their budget over the Cascade Mountains. Non-ideal wind conditions and the scarcity of wind data complicated data interpretation. The generally greater elemental flux at the outflow region, shown in Table 1 for the June 18, 1980 test period, is partially attributed to the transport of the visible Mt. St. Helens plume from the southwest. Those elements displaying the greatest difference in inflow and outflow fluxes are among the most abundant in the volcanic plume.

Ammonium, sulfate and nitrate data have also been examined to provide an indication of aerosol acidity and its spatial variation.

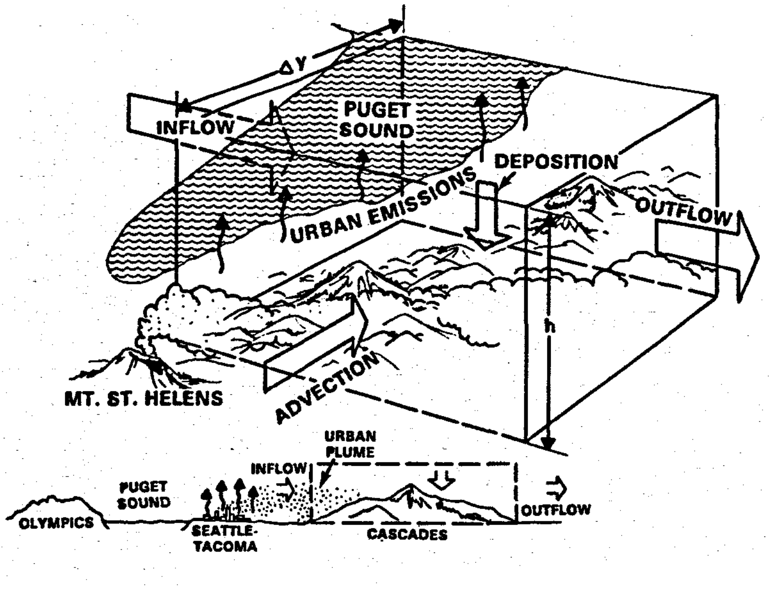

FIGURE 1. Box Model for Estimating Inflow, Outflow and Deposition Over the Cascades

TABLE 1. Elemental Fluxes to the East Along Aircraft Flight Paths

Trace Element	Inflow	Outflow	Difference
Sulfur	658	1650	-992
Titanium	112	153	-41
Lead	20	22	-2
Zinc	18	19	-1
Chromium	16	12	+4
Manganese	13	50	-37
Nickel	5	9	-4
Copper	5	8	-3
Bromine	3	11	-8
Potassium	278	891	-613

While NH_4:(NO_3 + SO_4) ratios generally suggest only partial neutralization of the strong acids HNO_3 and H_2SO_4, an increase in the equivalence ratios from west to east and from higher to lower elevations is consistent with neutralization during transport and the surface origin of ammonia gas responsible for neutralization.

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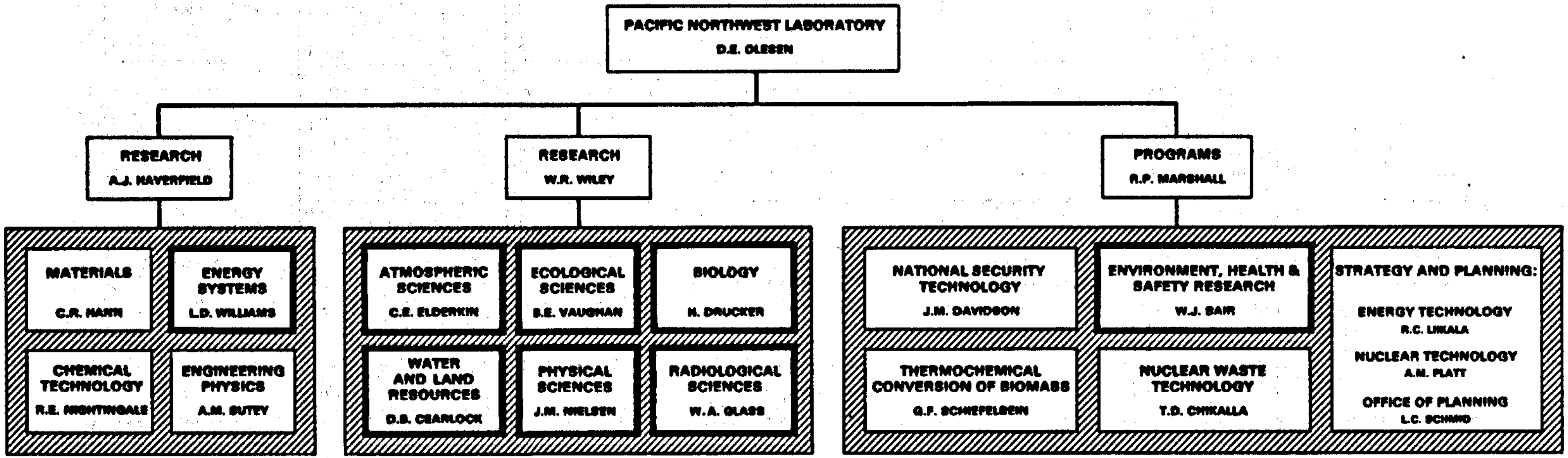

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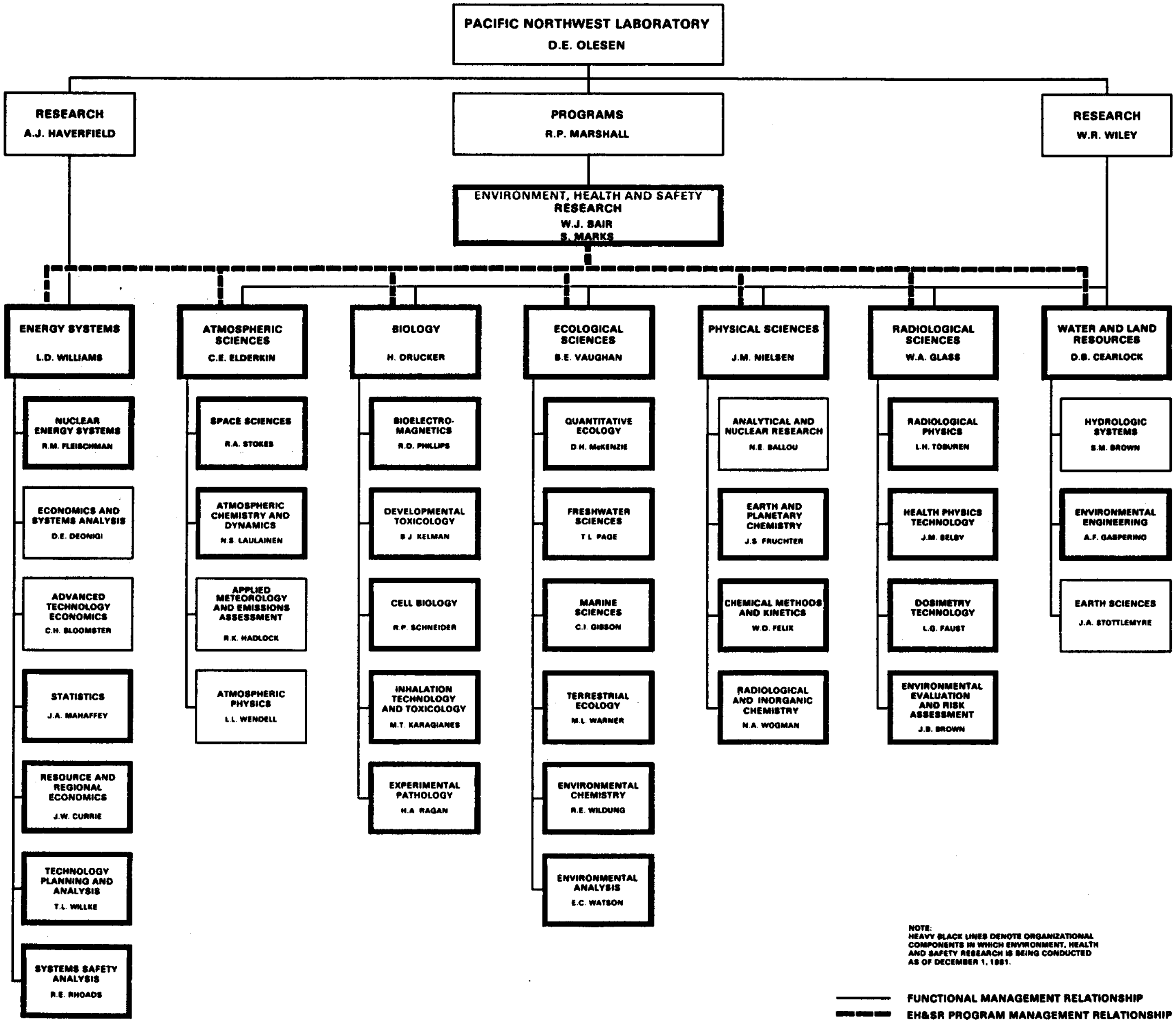

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