Pacific Northwest Laboratory Annual Report for 1982 to the DOE Office of Energy Research

Part 3 Atmospheric Sciences February 1983

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

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

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Pacific Northwest Laboratory
Richland, Washington 99352

PREFACE

This 1982 annual report from Pacific Northwest Laboratory (PNL) to the Department of Energy (DOE) describes research in environment, health, and safety conducted during fiscal year 1982. 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.

The parts of the 1982 Annual Report are:

Part 1: Biomedical Sciences		
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Part 2: Environmental Sciences		
Program Manager - B. E. Vaughan	B. E. Vaughan, Report Coordinator	C. M. Novich, Editor
Part 3: Atmospheric Sciences		
Program Manager - C. E. Elderkin	N. S. Laulainen, Report Coordinator	E. L. Owczarski, Editor
Part 4: Physical Sciences		
Program Manager - J. M. Nielsen	J. M. Nielsen, Report Coordinator	J. E. Danko, E. M. Toomey, Editors
Part 5: Environmental and Occupational Protection, Assessment, and Engineering		
Program Managers - S. Marks W. A. Glass	W. J. Bair, 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.

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 editor in chief, who directed the total effort.

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
1981	PNL-4100, 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 conventional energy resources (coal, gas, oil and nuclear power) as well as alternative energy resources.

The description of atmospheric research at PNL is organized in terms of generic studies:

- Theoretical Studies of Atmospheric Processes
- Pollutant Characterizations and Transformation
- Boundary Layer Meteorology
- Dispersion, Deposition and Resuspension of Atmospheric Pollutants

Theoretical Studies of Atmospheric Processes

Mathematical models are major investigative tools describing and predicting the nature and fate of atmospheric pollutants from the point of emission until their eventual deposition to the earth's surface. These models attempt to account for the transport and diffusion in air, the physical and chemical transformations during transport, and removal by wet and dry deposition of pollutants. Theoretical studies of various atmospheric processes provide the necessary physical basis for the development of models.

Pollutant Characterization and Transformation

An assessment of the nature and fate of atmospheric pollutants must begin with a description of the chemical and physical nature of the materials being emitted from the source. Because many atmospheric pollutants are changed to other physical states and/or chemical forms, understanding of these transformations is essential in any assessment. Of particular concern are those atmospheric transformations that produce potentially environmentally harmful or hazardous substances such as strong acids and mutagenic and carcinogenic compounds.

Boundary Layer Meteorology in Complex Terrain

Air pollution in mountainous regions is a particularly difficult problem because of the complexity of meteorological conditions over spatial scales ranging from individual valleys to systems of many valleys and ridges of a region and the diurnal coupling/decoupling phenomena between individual valleys and the regional convective boundary layer. In order to assure acceptable siting of energy development facilities, such as oil shale retorts, models and field measurements of the complex airflow and dispersion conditions in this complex setting must be developed and undertaken.

Dispersion, Deposition and Resuspension of Atmospheric Pollutants

Dispersion, deposition, and resuspension processes are linked intimately with the dynamics of the boundary layer. The former two processes act to reduce air concentrations of gaseous and particulate material while the latter process acts to increase air concentrations of particulate material, particularly in the large particle size classes. In many energy development areas of the West, all three processes will play an important role in determining the effective residence times of potentially hazardous particles in the atmosphere. The current program is aimed at gathering necessary field data and developing models for the prediction of the dispersion, deposition and resuspension of atmospheric pollutants.

This report describes the progress in FY 1982 for each of these generic areas. A divider page summarizes the goals of each area and lists, as bulleted items, project titles that support research in each area.

CONTENTS

PREFACE	iii
FOREWORD	v
POLLUTANT CHARACTERIZATION	
Transformation of Energy-Related Pollutants	1
Reaction Rates of Nitric Oxide Gas with Solid Surfaces of Polycyclic Aromatic Compounds, <i>D. R. Kalkwarf and V. W. Cope</i>	1
Reactions Between Solid Surfaces of Polycyclic Aromatic Compounds and Ozone Gas, <i>V. W. Cope and D. R. Kalkwarf</i>	2
BOUNDARY LAYER METEOROLOGY IN COMPLEX TERRAIN	
Atmospheric Diffusion in Complex Terrain	9
Temperature Inversion Breakup in Mountain Valleys and the Ventilation of Pollutants—The Joint Grama-Ascot Field Experiment 1982, <i>M. M. Orgill, C. D. Whiteman, R. N. Lee and T. R. Heimbigner</i>	9
Synoptic and Local Scale Interactions in the Geysers Geothermal Resource Area—An Overview, <i>M. M. Orgill</i>	11
Atmospheric Boundary Layer Studies	15
Improvements in Modeling Nocturnal Slope Winds, <i>J. C. Doran and T. W. Horst</i>	15
Slope Flow Data from the Rattlesnake Mountain Site, <i>T. W. Horst and J. C. Doran</i>	16
DISPERSION, DEPOSITION AND RESUSPENSION OF ATMOSPHERIC POLLUTANTS	
Atmospheric Diffusion in Complex Terrain	23
The Transport of H ₂ S and Trace Materials from the Geysers Geothermal Research Area, <i>M. M. Orgill, R. N. Lee and R. I. Schreck</i>	23
Plume Rise Calculations from the Ascot 1981 Geysers Experiments, <i>J. M. Thorp and M. M. Orgill</i>	26
Atmospheric Boundary Layer Studies	31
Evaluation of a Simple Plume Depletion Model, <i>T. W. Horst</i>	31
Air Pollution Dry Deposition: Radioisotopes as Particles and Volatiles	33
Particle Density Effects on Dry Deposition Velocities Measured in Wind Tunnel Experiments, <i>G. A. Sehmel and W. H. Hodgson</i>	33
Development of a Near-Monodispersed Particle Generator for Field Dry Deposition Experiments, <i>G. A. Sehmel and W. H. Hodgson</i>	34
Oil Shale Fugitive Air Emissions	37
Fugitive Emission Flow Paths Investigated with Tracers in Complex Terrain of the Oil Shale Region of Colorado, <i>G. A. Sehmel</i>	37
Simulation of Plume Depletion of Fugitive Emissions with Dual Tracer Particle Dry Deposition Experiments in Brush Creek, Colorado, <i>G. A. Sehmel</i>	38

Particle Resuspension and Translocation	41
Wind Resuspension Investigated with Real-Time Tracer Concentration Measurements, G. A. Sehmel	41
Tracer Resuspension Source-Strength Spread on the Surface by Wind Resuspension, G. A. Sehmel	43
THEORETICAL STUDIES OF ATMOSPHERIC PROCESSES	
Theoretical Studies and Applications	49
Theoretical Studies and Applications, W. G. N. Slinn	51
PUBLICATIONS	57
PRESENTATIONS	59
AUTHOR INDEX	63
ORGANIZATION CHARTS	67
DISTRIBUTION	69

Pollutant Characterization

POLLUTANT CHARACTERIZATION AND TRANSFORMATIONS

- **Transformation of Energy-Related Pollutants**

Because of their complex chemical forms, the combustion and conversion of fossil fuels gives rise to a host of polycyclic aromatic compounds (PACs) that can be emitted into the atmosphere. A number of these PACs have been shown or are suspected to be mutagenic or carcinogenic. Once they are in the ambient air, these PACs can react with other pollutant species, such as ozone and nitrogen oxides, to form new compounds. Some of these new compounds are less mutagenic or carcinogenic than the unreacted species, while others become more so. It is thus vital that the amount and composition of these substances be characterized with respect to their source and that their subsequent transformations in the atmosphere be understood.

The recent emphasis at PNL on identifying and studying potentially harmful pollutants has been the study of the reaction kinetics of ozone and nitric oxides with solid surfaces of polycyclic aromatic compounds. Since many reaction products are possible, even under controlled laboratory conditions, those reactions which produce mutagenic or carcinogenic products are given highest priority for study.

• Transformation of Energy-Related Pollutants

Objectives of this study are:

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

Correlating transformation rates of these compounds with distance from the source, ambient temperature, light intensity and concentrations of other reactant species, such as nitrogen oxides and ozone.

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

REACTION RATES OF NITRIC OXIDE GAS WITH SOLID SURFACES OF POLYCYCLIC AROMATIC COMPOUNDS

D. R. Kalkwarf and V. W. Cope

The purpose of this study was to measure the reaction rates of nitric oxide gas with solid surfaces of polycyclic aromatic compounds (PAC) which are representative of those found on coal fly ash. Nitric oxide is generally the preponderant oxide of nitrogen in the stack gas from a coal-fired power plant, and so its reactions must be considered in assessing the possibilities for chemical transformation of PAC in stack emission plumes. Although no reactions between nitric oxide and either pyrene or benzo[a]pyrene were detected by earlier investigators (Hughes et al. 1980), re-examination of these systems was considered important in view of the high mutagenicity shown by nitro derivatives of these compounds.

Reactions were conducted between monomolecular layers of PAC adsorbed on Pyrex® glass and a standard mixture of 193 ppm (7.0×10^{-3} mol/m³) nitric oxide in nitrogen gas. The monomolecular layers were prepared on the inside surfaces of 500-ml round-bottom flasks by the method described by Cope and Kalkwarf (1983), and the gas mixture was introduced after purging oxygen from the flasks with the system shown in Figure 1. After various periods of time, the nitric oxide was purged from a flask with air, and the organic material on the inside wall was dissolved in methylene chloride and analyzed by gas chromatography. When a large stoichiometric excess of nitric oxide was used to maintain a constant concentration of this gas in the flask, the amount of PAC decreased exponentially with exposure time.

(®)Trademark of Corning Glass Works, Corning, New York.

FIGURE 1. Apparatus for Exposing Monomolecular Layers of Polycyclic Aromatic Compounds (PAC) to Ozone Gas.

This relationship suggested that the transformation rates could be expressed by an equation analogous to those devised earlier for reactions of PAC with nitrogen dioxide (Kalkwarf 1982) and with ozone (Cope and Kalkwarf 1983), namely:

$$\frac{d(\text{PAC})}{dt} = -kA(\text{PAC}) [\text{NO}]$$

where: (PAC) = moles of original PAC remaining at any time t
 k = rate constant for PAC transformation in units of m/s
 A = molar surface area of monomolecular-layer PAC in units of m²/mol
 $[\text{NO}]$ = concentration of nitric oxide in contact with the PAC in units of mol/m³.

Rate constants derived from data for reactions of nitric oxide with pyrene, benzo[a]pyrene and perylene are listed in Table 1.

TABLE 1. Rate Constants for Reactions of Nitric Oxide with Monomolecular Layers of PAC Supported on Glass at 25°C

PAC	k, m/s
Benzo[a]pyrene	9×10^{-8}
Pyrene	7×10^{-8}
Perylene	3×10^{-8}

These reactions are considerably slower than analogous reactions of PAC with nitrogen dioxide (Kalkwarf 1982), which may account for their concealment in previous studies. Although the products of these reactions have not been studied in detail as yet, it was clear that the principal product of the reaction between nitric oxide and pyrene was 1-nitropyrene, a potent mutagen.

The concentrations of nitric oxide in the stack emission plume from a coal-fired power plant decrease rapidly with travel time from the stack, and values reported for a 600-MW plant range from 710 ppbv at 5 minutes to 50 ppbv at 34 minutes (Hegg and Hobbs 1979). Assuming that the PAC on fly ash in such a plume are distributed as monomolecular layers and are exposed to an average concentration of 50 ppbv (2×10^{-7} mol/m³) during the first hour, the half-time, T , for reactions of PAC with nitric oxide was estimated with the equation:

$$T = 0.693/kA [\text{NO}]$$

Values ranging from 70 to 300 days were obtained, and it was concluded that reactions with nitric oxide made negligible contributions to the transformation of PAC in stack emission plumes.

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Kalkwarf, D. R. 1982. "Transformation of Energy-Related Pollutants." In *Pacific Northwest Laboratory Annual Report for 1981 to the DOE Office of Energy Research, Part 3, Atmospheric Sciences*. PNL-4100 PT-3, pp. 15-17, Pacific Northwest Laboratory, Richland, Washington.

REACTIONS BETWEEN SOLID SURFACES OF POLYCYCLIC AROMATIC COMPOUNDS AND OZONE GASS

V. W. Cope and D. R. Kalkwarf

The purpose of this study was to characterize the reactions between ozone gas and solid surfaces of polycyclic aromatic compounds (PAC) which are representative of those found on coal fly ash. Characterization included measurement of the reaction rates, identification of as many products as possible and investigation of their mutagenicities. Ozone is present in the atmosphere at concentrations of about 50 ppbv (2×10^{-6} mol/m³) and reacts with most organic compounds. Ozone could be a key reagent for the atmospheric transformation of PAC in stack effluents from coal-fired power plants. Ozone gas has been reported to react with solid deposits of benzo[a]pyrene, benzo[b]fluoranthene and benzo[k]fluoranthene, (Lane and Katz 1977, Pitts et al. 1978) but more information on the rates and products of ozone with PAC is needed to adequately assess how such reactions would alter the health hazard of stack emission plumes.

Reactions were conducted between monomolecular layers of PAC adsorbed on Pyrex glass and ozone gas from a continuous generator as shown in Figure 1. The ozone was formed by photolysis of oxygen in air; and concentrations ranging from 40 to 200 ppbv were used. The monomolecular layers were prepared by evaporating solutions of PAC in methanol on the inside walls of 500-ml, round-bottom flasks. After addition of the solution, the flask was rotated until the inside surface was uniformly coated with liquid. This coating was then dried rapidly by passing clean air through the rotating flask. Under ultraviolet light, the dry surface emitted a weak and uniform fluorescence indicating the absence of macroscopic PAC crystals.

FIGURE 1. Apparatus for Exposing Monomolecular Layers of Polycyclic Aromatic Compounds (PAC) to Nitric Oxide Gas.

The initial reaction rates of ozone with pyrene, benzo[a]pyrene, perylene and chrysene were each found to increase linearly with ozone concentration up to at least 200 ppbv. The initial rates were also found to increase linearly with the amount of PAC in the flask up to a value corresponding to a monomolecular layer of PAC over the entire flask wall. With larger amounts of PAC, the rates remained essentially constant. These relationships indicate that the rate of PAC transformation can be expressed by the following equation:

$$\frac{d(\text{PAC})}{dt} = -kA(\text{PAC})[\text{O}_3]$$

where: (PAC) = moles of original PAC remaining at any time t
 k = rate constant for PAC transformation in units of m/s
 A = molar surface area of monomolecular-layer PAC in units of m^2/mol
 $[\text{O}_3]$ = concentration of ozone in contact with the PAC in units of mol/m^3 .

The values for the rate constants resulting from these measurements at 25°C are listed in Table 1.

Assuming that the PAC on fly ash from coal-fired power plants are distributed as monomolecular layers and are exposed to a constant average concentration of 25 ppbv ($1 \times 10^{-6} \text{ mol}/\text{m}^3$) ozone in the stack emission plume, we estimate the half-times, T , for reactions of PAC with ozone with the equation:

$$T = 0.693/kA[\text{O}_3]$$

TABLE 1. Rate Constants for Reactions of Ozone with Monomolecular Layers of PAC Supported on Glass at 25°C

PAC	k , m/s
Benzo[a]pyrene	6×10^{-4}
Pyrene	1×10^{-4}
Perylene	4×10^{-5}
Chrysene	5×10^{-6}

The resulting values are listed in Table 2.

Data previously reported on samples of the Colstrip stack plume indicated half-distances of 8 and 12 km for pyrene and benzo[a]pyrene, respectively (Kalkwarf and Garcia 1979). Since these samples were collected when the wind velocity was 2 km/hr, the corresponding transformation half-times were 4 and 6 hr. Comparison of these values with those in Table 2 shows that the transformation of pyrene and benzo[a]pyrene in the Colstrip plume could be accounted for entirely by reactions with ozone.

To date, only the reaction products of pyrene with ozone have been investigated in detail. Analysis of these product mixtures by high-performance liquid chromatography indicated 5 major components. Two of these were identified to be pyrene-1,6-quinone and pyrene-1,8-quinone by comparison of their chromatographic retention times with those of authentic samples and from their color reactions with sulfuric acid and with sodium hydrosulfite. Neither of these quinones was found to be mutagenic as measured by the modified Ames Salmonella assay on thin-layer chromatographic plates (Bjorseth et al. 1982). One of the unidentified components, however, was found to be mutagenic in this test and its identity is being sought.

TABLE 2. Estimated Half-Times for Reactions of Ozone with Monomolecular Layers of PAC in a Stack Emission Plume at 25°C

PAC	A (m^2/mol)	$[\text{O}_3]$ (mol/m^3)	T (hr)
Benzo[a]pyrene	5.8×10^5	1×10^{-6}	0.6
Pyrene	4.4×10^5	1×10^{-6}	4
Perylene	4.8×10^5	1×10^{-6}	10
Chrysene	4.9×10^5	1×10^{-6}	80

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Boundary Layer Meteorology in Complex Terrain

BOUNDARY LAYER METEOROLOGY IN COMPLEX TERRAIN

- **Atmospheric Diffusion in Complex Terrain**
- **Atmospheric Boundary Layer Studies**

Major reserves of fossil fuels are located in mountainous or other complex terrain. As the use of these fuels as an energy source increases, the emission of air pollutants, such as sulfur and nitrogen compounds, trace metals, and fugitive dust produced by the combustion, conversion and extraction of these fuels, will increase also. The analysis of the fate of these pollutants is particularly difficult in mountainous terrain settings, but is urgently needed to assure that the nation's energy plan can proceed efficiently and yet be environmentally sound.

The research activities at PNL are related to the interlaboratory ASCOT program. During FY 1982 the area of study focus shifted from the Geysers Geothermal Site in California to the oil shale region in western Colorado. A wealth of data from the Geysers area is being analyzed and tested against a variety of models accounting for the transport and dispersion of air pollutants under special but pertinent meteorological conditions. An effort also is being made to determine how much of the experience gained at the Geysers can be extended to the oil shale regions of Colorado.

The field activities in the ASCOT program include the micrometeorological description of drainage winds from data obtained at Hanford (Rattlesnake Mountain), the Geysers (Anderson Creek, Big Sulfur Creek) and Colorado (Brush Creek). Other measurement programs include radiation (energy budget) measurements and tracer experiments.

• 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.

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.

Evaluating and developing various transport and dispersion models for complex terrain.

TEMPERATURE INVERSION BREAKUP IN MOUNTAIN VALLEYS AND THE VENTILATION OF POLLUTANTS-THE JOINT GRAMA-ASCOT FIELD EXPERIMENT 1982

M. M. Orgill, C. D. Whiteman, R. N. Lee and T. R. Heimbigner

One of the major objectives of the EPA GRAMA Program being conducted at the Pacific Northwest Laboratory (PNL) is the development of a validated valley air pollution model. An important aspect of the model is its ability to simulate the breakup of temperature inversions and pollutant plumes by the growth of convective boundary layers over the valley floor and sidewalls, and the development of upslope flows over the sidewalls. During this inversion breakup period, fumigation and ventilation of air pollutants occur in the growing convective boundary layer.

To test the model with actual air quality and meteorological data, the PNL Atmospheric Studies in Complex Terrain (ASCOT) and GRAMA programs supported a sulfur hexafluoride (SF_6) aircraft sampling program during the joint DOE-EPA field experiment, conducted between July 26 and August 7, 1982. The experiment was conducted at Brush Creek, Colorado, approximately 55 km north-northeast of Grand Junction.

The purpose of the aircraft sampling was to provide data on the locations, heights and concentrations of SF_6 within the Brush and Roan Creek valleys. The aircraft sampling supplemented a surface sampling network which consisted of sampling sites dispersed along the valley axis and across the valley at a location about 3 km from the entrance of Brush Creek. Figure 1 shows the topography of the Brush Creek area and the experimental setup.

Sulfur hexafluoride tracer gas was released from a site approximately 11 km up Brush Creek from its confluence with Roan Creek.

The material was released at a height of about 100 m by a balloon-borne hose with an outer diameter of 1.3 cm. The SF_6 tracer was released at a rate that varied from 3 to 9 kg/hr for a duration of 3 to 6 hr. Release rates and durations varied from experiment to experiment.

During these experiments the airborne sampling was conducted with the PNL Cessna 411 aircraft, which carried a real-time SF_6 analyzer and a DAS-32 data acquisition system. The real-time SF_6 sampling was supplemented with periodic grab samples taken in double-walled polyethylene bags. Data from the SF_6 tracer analyzer were monitored by strip chart and recorded on magnetic tape. Location and supplementary temperature, humidity, wind and turbulence data were also recorded on tape.

Four experiments were conducted in the early morning hours (0100 to 1000 MST) of July 29, July 31, August 4 and August 6, 1982. The airborne sampling was conducted during the period 0530 to 0930 MST for four experimental periods. Concentrations in the Brush Creek valley were of principal interest, with aircraft observations taken over the valley center, and over the west and east sidewalls. Sampling also extended throughout the Roan Creek valley but this was of secondary importance.

In this report, data are presented for August 6, and preliminary analyses are presented. At the time of writing real-time aircraft data are not yet available for analysis. Figure 2 depicts a time series of the SF_6 concentration data taken at the GC-1 site on the west sidewall of Brush Creek (1952 m, see Figure 1). Also plotted on Figure 2 are the aircraft grab sample concentrations taken at various locations within the valley. Further details on the locations and times of the aircraft grab samples are given in Table 1.

FIGURE 1. Brush Creek Topography Showing the Location of Surface Sampling Sites (dots), Gas Chromatographs (triangles) and Aircraft Sampling Flight Paths.

FIGURE 2. Sulfur Hexafluoride Concentration at Site GC-1 as a Function of Time on August 6, 1982. Aircraft SF_6 grab sample concentrations are indicated by dots. See Table 1 for location of samples. GC-1 data were collected by Mr. Bill Bernardo of Region VIII, EPA.

TABLE 1. Locations, Altitudes and Times of SF₆ Aircraft Grab Samples Taken During the August 6 Experiment. Concentrations in ppt.

Location/Time	Altitude (m M.S.L.)				
	1981 (6500 ft)	2134 (7000 ft)	2286 (7500 ft)	2408 (7900 ft)	2530 (8300 ft)
Brush Crk. Entrance					
A 0556-0600	219				
0608-0611	184				
0623-0626		4.6			
0634-0637			ND		
0701-0704		12.2			
0725-0729			Trace		
Brush Crk. Valley					
B 0652-0656			ND		
0718-0721			10.1		
0758-0801			213		
0813-0816			222		
0857-0901				37.5	
West Sidewall (Brush Mtn.)					
C 0712-0714					Trace
0751-0754					116
0806-0809					113
East Sidewall (Skinner Rdg.)					
D 0820-0825					ND
0836-0840					Trace

On August 6, the SF₆ tracer gas was released from 0410 to 0946 MST. The increase in concentration at the GC-1 site, shown in Figure 2, is due to the valley filling with tracer gas, which is then transported downvalley to the site. A rapid increase in concentration after 0630 MST at the site may be due to the initiation of upslope winds on the sunny west sidewall after sunrise (0610 MST) and the fumigation of pollutants into the growing boundary layer. The decrease of concentration after 0645 MST may be related to deepening of the convective boundary layer in the valley (0640 MST) and upslope winds resulting in enhanced fumigation and ventilation of the SF₆. The slight increase in concentration around 0735 MST appears to be related to the change from downvalley winds to upvalley winds. Further decrease in the concentration is brought about by the establishment of upvalley winds and the transport of the SF₆ tracer upvalley.

A point of special interest is the detection of SF₆ over Brush Mountain by the aircraft. At 0712 MST, only a trace of SF₆ was detected over Brush Mountain. By 0751 MST, concentrations had increased to over 100 ppt and concentrations in the higher elevations of the valley had increased to over 200 ppt. These increases in concentrations coincided with

the period when winds were shifting from downvalley to upvalley (~0730 MST). These results and the variation of concentration at the GC-1 site suggest that the SF₆ tracer was carried up the sunny west sidewall by upslope air currents. The east sidewall, which was still in the shadows, received only small amounts of SF₆ tracer.

These preliminary results indicate that the air quality data taken by aircraft and surface sampling during the joint ASCOT-GRAMA field experiment will have important validation value for the GRAMA valley model and will also increase our understanding of physical transport mechanisms operating during local wind cycles. Additional analysis of the ASCOT-GRAMA data base will provide further details concerning this particular problem.

SYNOPTIC AND LOCAL SCALE INTERACTIONS IN THE GEYSERS GEOTHERMAL RESOURCE AREA--AN OVERVIEW

M. M. Orgill

During July 1979, September 1980 and August 1981, the Department of Energy sponsored multilaboratory field programs (called Atmospheric Studies in Complex Terrain, or ASCOT)

in the Geysers-Calistoga Geothermal Resource Area (GGRA) to study the development and characteristics of nocturnal katabatic (drainage) winds and their effects on atmospheric transport and diffusion. During the past 3 years the ASCOT group at PNL has utilized the ASCOT data base to examine the effect of larger-scale (synoptic and meso-scale) wind systems on the development and persistence of local nocturnal katabatic flow. This work is a continuation of past work reported at the Second Conference on Mountain Meteorology in Steamboat Springs, Colorado (Orgill, Schreck and Whiteman 1981).

The present work has examined a portion of the September 1980 and August 1981 ASCOT data base in order to extend the analysis past the July 1979 time period. The results from all three ASCOT experimental periods are being summarized in an article to be submitted to the Bulletin of the American Meteorological Society under the title "Synoptic and Local Scale Interactions during the DOE-Sponsored ASCOT Field Experiments - An Overview." Topics that are discussed in this overview are the local wind cycle, marine air intrusion, effects of easterly winds, temperature inversion variations, nocturnal warming events, and transport and dispersion of fluorescent particle (FP) tracer plumes.

The purpose of this report is to summarize briefly the principal results described in the overview article to be published in the near future.

Local Wind Cycle

Topography, the radiational climate and the seasonal synoptic pressure gradient combine to give the east side of the GGRA a three-phase diurnal wind cycle. This wind cycle consists of a morning upslope (easterly) flow followed by an afternoon southwesterly flow. In the evening hours, radiational cooling initiates and supports a northwesterly drainage flow on the Anderson Creek slopes unless ambient winds are too strong. Generally, winds on the Mayacmas ridge of 7 m/s or more are unfavorable for persistent nocturnal drainage winds. On the west side of the GGRA, in Big Sulphur Creek valley, preliminary analyses show a diurnal wind cycle consisting of two components--upvalley and downvalley flow.

Marine Air Intrusion

The mean sea-level pressure differential between the coast and inland areas and the observed winds at the GGRA undergo long-term

(several-day) and diurnal oscillations during the summer season. Surges of modified marine air (westerly flow) invade the GGRA about every afternoon. These surges normally decrease in the late evening but may persist if supported by enhanced pressure gradients or migratory weather disturbances. The nocturnal drainage wind occurrence on the west side of the GGRA depends on the strength of the marine air intrusion. Surges of marine air will overpower the local nocturnal drainage wind.

Effects of Easterly Winds

A mild "Santa Ana" wind effect was observed at the GGRA during the September 1980 ASCOT experiments. The formation of high pressure to the north or low pressure to the west of the area resulted in the presence of easterly winds over the GGRA, primarily during the evening hours when the westerly marine air intrusion was retreating. These easterly winds were strongest on the ridges and, on some evenings, descended into the Anderson Creek drainage area. This coupling between the east winds and the lower levels eventually destroyed any drainage wind present on the slopes.

Temperature Inversion Variations

Nocturnal or radiational cooling always produced surface and elevated temperature inversions over the GGRA. On "good" drainage nights, the drainage wind layer on the slopes of Anderson Creek appeared to be about 2 to 3 tenths of the total stable layer (top of the highest temperature inversion). Generally, inversion and drainage layer depth would increase with distance downslope. The total stable layer or top of the inversion layer was eroded downward by westerly or easterly upper-level synoptic winds of 6 m/s or more.

Nocturnal Warming Events

Temperature data from several tower and tethered sonde locations in the Anderson Creek area revealed the occurrence of warming periods (0.5 to 3.5°C) during the normal nocturnal cooling period. These warming periods decreased the normal nighttime cooling rates on the slopes and probably interfered with the development of deep persistent drainage winds. The causes of the warming periods are not yet known in a quantitative manner, but a number of possible mechanisms were examined, such as advection, differential sinking and warming of air,

oscillations in the drainage wind and turbulent mixing.

Transport and Dispersion

During the "good" drainage wind nights on the slopes, oil fog smoke tracer releases showed the movement of the drainage flow down the slope into the Middletown area. As expected, synoptic-mesoscale coupling with the local drainage wind affected the transport and dispersion of tracer plumes. One example, FP tracer releases on the nights of July 18 and 22, 1979, is discussed in relation to coupling effects on plume trajectories and changes in plume widths.

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Orgill, M. M., R. I. Schreck and C. D. Whiteman. 1981. "Synoptically Driven Downslope Winds and Their Effects on Local Nocturnal Drainage Airflow in the Geysers Geothermal Resource Area." Paper presented at Second Conference on Mountain Meteorology, November 9-12, 1981, Steamboat Springs, Colorado.

• 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.

IMPROVEMENTS IN MODELING NOCTURNAL SLOPE WINDS

J. C. Doran and T. W. Horst

A one-dimensional model to describe the structure of nocturnal drainage winds over simple slopes has been developed. The inclusion of advection terms enables the model to determine changes in the structure with downslope distance, in effect giving a two-dimensional model, although changes in slope can be handled only in an approximate manner.

A number of features have been added to an earlier version of the model (Doran and Horst 1982). These include an improved algorithm for calculating diffusion terms, which is based on the approach described by Mahrer and Pielke (1978), and the imposition of a lower bound of 0.2 m for the mixing length relating the eddy diffusivity K to the turbulent kinetic energy E . Substantial improvements in the comparisons between modeled results and observations obtained on Rattlesnake Mountain in July 1980 have been obtained as a result of these modifications. These comparisons are shown in Figures 1 and 2, which present modeled and observed temperature profiles at towers A and B and wind speed profiles at tower B, respectively. A description of the Rattlesnake Mountain site and the location of the towers are given in Horst and Doran (1981).

Comparisons were also made between model simulations and measurements made in July 1981. In this case, the ambient winds were upslope, in contrast to the downslope winds found in July 1980. Simulations of the temperature profiles were again very good, but the observed wind profile at tower B showed a deeper katabatic layer than predicted by the model. A possible contributing cause is the failure of the model to handle advection effects properly for ambient upslope

FIGURE 1. Comparison of Measured and Modeled Temperature Profiles at Towers A (upper) and B (lower), July 1, 1980. Model (—); measured (□--□)

FIGURE 2. Comparison of Measured and Modeled Downslope Wind Component at Tower B, July 1, 1980. Model (—); measured (□--□)

winds. Further work in this area as well as the inclusion of a surface heat budget are being considered.

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SLOPE FLOW DATA FROM THE RATTLESNAKE MOUNTAIN SITE

T. W. Horst and J. C. Doran

Measurements of the wind and temperature structure of nocturnal slope flow were made on Rattlesnake Mountain near Richland, Washington during the summers of 1980 and

1981 (Horst et al. 1982). These measurements were made as part of Project ASCOT (Atmospheric Studies in Complex Terrain), a multilaboratory research effort sponsored by the U.S. Department of Energy.

ASCOT's initial focus has been on nocturnal drainage winds. Drainage winds begin when air adjacent to a sloped surface flows down the slope because it is cooled more, and is denser 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; however, the Rattlesnake Mountain site was selected for measurements of slope flow in the simplest possible geometry.

Wind and temperature profiles were measured on a portion of Rattlesnake Mountain that is uniform in the cross-slope direction and vegetated with short grasses and scattered sagebrush. The upper part of the slope closely approximates a simple, tilted plane. Measurements were made from three towers at different distances from the ridge crest to determine the development of slope flow with downslope distance. Two towers were located on the upper slope, which has a uniform slope angle of 21 degrees, and the third tower was located below a change in the slope from 21 to 8 degrees. In each case the towers extended through the local depth of the katabatic flow.

The data have been published in a report by T. W. Horst and J. C. Doran entitled Simple Nocturnal Slope Flow Data from the Rattlesnake Mountain Site, PNL-4406/ASCOT-82-5. This report lists the tower wind and temperature data from the four best cases of slope flow observed in 1980 and 1981: June 19-20 and July 1-2, 1980 and July 2-3 and October 16, 1981. Also listed are the ambient wind and temperature profiles to a height of 300 m from a Tethersonde® flown at the experimental site and surface winds from a network of telemetry stations surrounding the site. For each case a brief summary is given of the synoptic situation, the surface winds over the Hanford area from the telemetry network, the Tethersonde observations at the experimental site, and the tower wind and temperature profiles.

These data have already been used by PNL and other DOE laboratories to describe the characteristics of slope flow and to evaluate models of slope flow. References to reports of these investigations are included in the bibliography.

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Dispersion, Deposition and Resuspension of Atmospheric Pollutants

DISPERSION, DEPOSITION AND RESUSPENSION OF ATMOSPHERIC POLLUTANTS

- **Atmospheric Diffusion in Complex Terrain**
- **Atmospheric Boundary Layer Studies**
- **Air Pollution Dry Deposition**
- **Oil Shale Fugitive Air Emissions and Plume Depletion**
- **Particle Resuspension and Translocation**

The concentration in air of pollutant species is governed by the rate of input from sources, the rate of dilution or dispersion as a result of air turbulence, and the rate of removal to the surface by wet and dry deposition processes. Once on the surface, pollutants also may be resuspended, depending on meteorological and surface conditions. An understanding of these processes is necessary for accurate prediction of exposures of hazardous or harmful pollutants to humans, animals, and crops.

At PNL several research programs focus attention on these processes through a combination of field, laboratory and modeling studies. In the field, plume dispersion and plume depletion by dry deposition are studied by the use of tracers. A unique application of tracer technology at PNL is the simultaneous release of depositing and non-depositing tracers. Studies have been conducted to evaluate a simple plume depletion model. Resuspension is also studied using tracers on contaminated surfaces. The objective of the resuspension studies is the development and verification of models for predicting the airborne concentrations of pollutants over areas with surface contamination. These models will be of particular relevance to the evaluation of deposition and resuspension of radionuclides.

• 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.

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.

Evaluating and developing various transport and dispersion models for complex terrain.

THE TRANSPORT OF H₂S AND TRACE MATERIALS FROM THE GEYSERS GEOTHERMAL RESEARCH AREA

M. M. Orgill, R. N. Lee and R. I. Schreck

The transport of pollutants from energy production areas in complex terrain is an important aspect in the assessment of whether the pollutants may cause an impact on the general public and environment through dry deposition, acid precipitation and impaction.

The Atmospheric Studies in Complex Terrain (ASCOT) group at the Pacific Northwest Laboratory (PNL) is utilizing airborne and surface sampling results to assess the transport of pollutant materials from the Geysers-Calistoga Geothermal Resource Area (GGRA). These sampling results were accomplished during the ASCOT Cooling Tower experiments in August 1981 (Orgill, Lee and Hannigan 1981). The purpose of this sampling was threefold: 1) to define the types and quantities of airborne materials such as H₂S, R, As, Cd, Pb, Cr, and Hg that may be emitted from the geothermal operations, 2) to collect data on the location, height and concentration of the airborne materials and 3) if possible, to estimate the flux of pollutant material being transported from the GGRA. The purpose of this paper is to discuss the initial data analysis and results in respect to the first two goals. A final analysis and conclusions will be reported in the future.

The airborne sampling was conducted during the daytime hours of August 20, 21, 23 and 24 as part of the ASCOT limited mixing experiment. Sampling procedures and instrumentation were described previously (Orgill, Lee and Hannigan 1981). To supplement the airborne sampling, six surface-based high-volume filter samplers (for aerosols only) were located at sites within, east, and west of the GGRA operations (Figure 1). These samplers were operated during the seven hours of airborne sampling for each day.

The principal sampling transects were 5 to 6 km and 12 to 15 km from Units 9 and 10. These transects were flown in an approximate racetrack manner at three different altitudes for an hour or more for each altitude. Each sampling transect was flown from three to five times, except for a transect at 18 to 20 km which was flown once. Background sampling transects were flown at two altitudes to the west of the GGRA either before or after the principal sampling (see Figure 1). The combination of sampling aerosols and the real-time sampling of H₂S (and SF₆) limited the range of altitudes and distances for sampling H₂S (and SF₆). Aerosol measurements required flying on a fixed path and altitude for a minimum of an hour in order to collect sufficient material for analysis.

During the four days of sampling, 41 filter samples were taken. An initial selection of eight filters was made for x-ray fluorescence (XRF) and neutron activation analysis (NAA). Three were examined by XRF and all eight were examined by NAA. Thirty-three to thirty-six trace elements were examined by XRF and NAA, respectively, to obtain air concentrations. Only fourteen trace elements had air concentrations of sufficient magnitude to warrant further assessment. In order to determine whether these fourteen elements were enriched in comparison to crustal material, the crustal enrichment factor

$$EF_c = (C_x/C_{FE})_{\text{aerosol}} / (C_x/C_{FE})_{\text{crust}}$$

was calculated. An EF_c value close to unity indicates that little or no enrichment occurred for a given element in the aerosol relative to Fe. A value <1 indicates depletion of an element, whereas a value >1 indicates enrichment of a given element relative to Fe. Generally, an enrichment factor of 10 or more is considered significant with respect to anthropogenic input to the atmosphere. Table 1 shows the calculated enrichments for the selected surface and airborne samples.

FIGURE 1. Geographic Setting of the Geysers—Calistoga Geothermal Resource Area, The Geysers, and Location of Aircraft Sampling Flights and Surface High-Volume Filter Samplers.

Trace elements of particular interest in geothermal operations are As, Cd, Hg, S, Pb and Cr. The XRF analysis shows a wide range in enrichments for these elements. The NAA analysis shows an enrichment for As, Br and Cr and these results are consistent with the XRF analysis. The cadmium and mercury NAA results were subject to large analysis errors and are not shown. The enrichments for the aircraft samples are especially small indicating small concentrations of materials and/or insufficient sampling time. One of the primary pollutants from the GGRA is sulfur in the form of H₂S. Both surface and airborne samples indicated sulfur enrichment in the air. As expected, H₂S (in the form of gaseous sulfur) was detected in the eastern quadrants from the GGRA since the winds are generally from southwest to northwest at speeds varying from 4 to 8 m/s. H₂S was detected up to 20 km from the GGRA. Peak concentrations varied from 15 to 61 ppb but

most concentrations were below 20 ppb. On August 20 and 21, H₂S was detected up to 1800 m M.S.L. at concentrations between 7 and 25 ppb at distances 6 to 12 km from Units 9 and 10. Figure 2 shows one example of the real-time airborne ambient and background H₂S data for the afternoon of August 24. H₂S was detected at 1524 m M.S.L. northeast of the GGRA (shaded area). The airborne sampling measurements were consistent with airborne lidar measurements of cooling tower plume rise. Uthe (1982) reported an average maximum plume rise from Units 9 and 10 of around 1100 m (2068 m M.S.L.) on the afternoon of August 24. Apparently, a deep convective layer over the GGRA allowed the cooling tower plumes to rise to heights above Cobb Mountain.

The results presented above are preliminary since a large amount of data still needs to be processed and analyzed during the next

TABLE 1. Crustal Enrichment Factors for Trace Elements Detected in Selected Surface and Airborne Filter Samples in the GGRA.

Element	Enrichment Factor							
	Surface Samples (m ³)					Airborne ^(a) Samples (m ³)		
	R.S. 8/23 (215)	Sawmill 8/23 (198)	R. S. 8/24 (165)	Brangus 8/24 (209)	Delfino 8/24 (165)	ZF-45 8/23 (13.3)	ZF-41 8/24 (11.2)	ZF-49 8/24 (14.5)
XRF								
As	<201		<35				ND	
Br	145		32				ND	
Cd	4798		1467				ND	
Cr	3	3				32		
Hg	<2670	NO	284	NO	NO	NO	ND	NO
Mn	2	XRF	2	XRF	XRF	XRF	3	XRF
Ni	3		3				ND	
Pb	19		13				ND	
S	214		26				85	
Sn	<1014		136				ND	
V	2		2				ND	
Zn	8		3				ND	
NAA								
As	28	24	4	18	6	<2	ND	<5
Br	81	139	14	104	78	<5	ND	<7
Co	1	1	1	1	1	2	ND	<2
Cr	3	3	2	4	4	<4	ND	<8
Na	0.2	0.3	0.1	0.3	0.2	0.2	ND	0.1
Zn	8	8	3	4	13	<11	ND	<20

(a) ZF-45 August 23 914 m M.S.L.
ZF-41 August 24 1128 m M.S.L.
ZF-49 August 24 1524 m M.S.L.

fiscal year. Further results of this work are scheduled to be presented at the Sixth Symposium on Turbulence and Diffusion, March 22-25, 1983, in Boston, Massachusetts.

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FIGURE 2. Transport of Gaseous Sulfur (H_2S) Plume from The Geysers as Depicted by Aircraft Sampling. Locations: M—Middleton; C—Cobb Mountain; B—Boggs Mountain; H—Mt. Hannah.

PLUME RISE CALCULATIONS FROM THE ASCOT 1981 GEYSERS EXPERIMENTS

J. M. Thorp and M. M. Orgill

The August 1981 ASCOT field experiment at The Geysers Geothermal Resource Area resulted in five tests of tracer release into the steam plume from a power plant cooling tower. Three of the tests were conducted at night during light valley drainage flow conditions, and two of the tests were conducted during afternoons with limited mixing conditions.

A continuous photographic record of the visible cooling tower plume was an integral part of the experiments. This record consisted of sequential 5-minute exposures of the plume on color slides and black-and-white film (Thorp and Glover 1982).

This report is a brief discussion of the plume rise calculations made from the photographic record. Details of the analysis will be published later in a journal article.

Analysis

Using geometric techniques (Thorp 1976) on the projected slide image or photographic print, the visible plume height has been calculated at approximate 15-min intervals for each of the five 7- to 8-hour experiments (Thorp et al. 1982). One additional test, that of August 12-13, has also been analyzed for plume rise. On that night conditions were never favorable for a tracer release, but a 4-hour photographic record was made, and tethered profiles of wind velocity, temperature and relative humidity were taken for 8 hours. The steam plumes photographed

on that night were by far the highest plumes observed during the nighttime experiments.

Brief descriptions of the plume rise measurements and the related meteorology follow in the next subsections. A general comparison between experiments can be made from Table 1.

Nighttime Drainage Flow Experiments

August 12-13. Though the drainage flow was never strong enough to warrant a tracer release this night, meteorological information was obtained from 1900 to 0300 PST, and photographs were made from 2300 to 0300 PST. Before midnight, the average wind direction through the 400-m boundary layer was somewhat variable, but was more persistently southeast after midnight. Speeds were very light all night, generally 1 m/s or less. Temperature and relative humidity averages within the 400-m layer decreased and increased respectively during the night. Visible plume heights were somewhat variable between 250 and 350 m.

August 15-16. Average wind direction in the 200-m boundary layer was northwest prior to 2000 PST. Southeast drainage flow averaging 2 to 3 m/s set in at about 2000 PST and continued for the duration of the experiment. The average temperature fell until 2100 PST, then remained nearly constant to 20°C. The average relative humidity, however, rose to a peak of 46% at 2100 then decreased. The visible cooling tower plume retained a tapered flame-like appearance throughout the night, and rose to a maximum height of ~100 m at

2200 PST then slowly decreased to 50 m by 0100, and remained near that height for the rest of the experiment. Figure 1 shows a plotted example of the analysis of this experiment.

August 16-17. Average wind direction within the 200-m boundary layer was northwest until 2100 PST; after then a light drainage flow from the southeast set in with speeds of 2 m/s. The average temperature within the layer fell until ~2100 PST then was relatively constant at 23°C. Relative humidity peaked at 37% near 2000 PST then fell to 25% for most of the night. A rise to 32% was noted at 0400 when the experiment ended. The visible plume rise during this experiment was the most constant of any, and was also the lowest. It increased slowly during the night from 25 to 40 m. The plume photographs did not show the tapered plume outline observed the previous night.

August 21-22. A slightly stronger southeast drainage wind of ~3 m/s was established this night between 2100 and 2200 PST. The average temperature in the 200-m boundary layer fell from 25°C to ~20°C by 2000 PST and remained near 20°C the remainder of the experiment. Average relative humidity within the layer had minor fluctuations around 30%. Illumination of the plume was poor for this experiment as the moon was waning and rose late. In general the visible plume was similar to that of August 16-17, rising slowly during the night from about 25 m at 2000 PST to a 60-m maximum near 0230 PST, then decreasing. Detached puffs from the main plume were observed to reach heights of 90 to 120 m.

TABLE 1. Plume Rise and Meteorological Summary

Date Aug 1981	Type	Layer Averages						
		Temp., °C	Relative Humidity, %	Wind Speed, m/s	Depth of Layer, m	Plume Height, m	Inversion Height, m	Lidar Plume Height, m
12-13	Night-Wk Drainage	23-17	49-68	<1.0	400	350-250	>500	UNK
15-16	Night-Drainage	26-20	46-16	2.2	200	100-50	>500	150-400
16-17	Night-Drainage	29-22	36-25	1.7	200	25-40	50-285	275-500
21-22	Night-Drainage	25-20	32-27	2.0	200	25-60	50-260	200-500
23	Day	16-14	69-37	3.1	350	450-50	300->475	150-600
24	Day	19-14	42-60	4.2	350	35-250	None	100-1300

FIGURE 1. Results of Analysis for August 15-16 Experiment

Daytime Limited Mixing Experiments

August 23. The average wind velocity through a 350-m boundary layer was southwesterly 1 to 3 m/s from 1100-1400 PST, shifting gradually to northwest 3 to 5 m/s during the late afternoon and early evening. Average temperature through the layer hovered near 15°C except for a brief dip to 13.5°C at 1900 PST. Relative humidity was between 50% and 70% all afternoon with peaks at 1300 and 1900 PST. After 1900 the humidity dropped to 40% or less for the duration of the experiment. The visible plume was quite tall, ~500 m, at 1100 PST when photography commenced, but ranged between 70 and 150 m from 1300 to 1830 PST. An increase in plume height to ~300 m occurred between 1830 and 1930 PST. Two lobes of the plume were apparent on the photographs after 1830; the highest lobe was >400 m.

August 24. There were several differences in the meteorology and plume behavior between this and the previous day. The wind velocity averaged through a 350-m layer was more westerly and stronger (3 to 5 m/s) during the

afternoon. The average wind backed to southwest at 5 m/s at 1600 to 1700 PST then was northwest 4 to 5 m/s until the end of the photographic record at 2100 PST. Average temperature for the layer rose from 15°C at 1100 PST to nearly 19°C in the afternoon then fell slowly to 13.5°C by 2100. Relative humidity in the layer generally averaged 40% to 50% until 1600 then rose to 60 to 70% in the early evening. The visible plume was low during the daylight hours; much of the afternoon it was 50 m or less. At about 1800 PST however, it began to rise and was ~250 m when photography ceased at 1915 PST.

Conclusion

Table 1 summarizes the analyses of the photographic records. The depths of the layer for averaging temperature and humidity were individually chosen to encompass the plume rise for each experiment. The invisible plume heights measured by the airborne lidar system (Uthe 1982) indicate that the time-integrated plume rise visible photographically was generally about 1/10 that "seen" by the lidar.

This is primarily because the lidar is sensitive to the detection of subvisible particulates at the near-infrared wave lengths. Secondly, in order to provide an average visible plume envelope, a long photographic exposure was chosen to eliminate short term excursions and detached puffs from the plume photo. Conventional photographic exposures would have produced evidence of greater visible plume rise.

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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.

EVALUATION OF A SIMPLE PLUME DEPLETION MODEL

T. W. Horst

The Gaussian plume model is commonly used to estimate airborne pollutant concentrations downwind of a point source release. The dimensions of the Gaussian plume are empirical functions of downwind distance and atmospheric stability, which have been determined from experimental programs using nondepositing tracers. Since the Gaussian plume model is based on empirical data and does not explicitly model the vertical diffusion process, it is not easy to modify to account for depletion of the plume caused by the loss of depositing pollutants.

Plume depletion is commonly modeled with the Chamberlain (1953) source depletion technique. This technique distributes the loss due to dry deposition throughout the vertical extent of the plume and thus retains the Gaussian vertical profile. Since deposition actually depletes the portion of the plume adjacent to the surface, source depletion overestimates the ground-level concentration, overestimates the deposition, and underestimates the remaining airborne material.

Horst (1977) developed an exact, surface depletion technique to account for the depletion of an airborne plume by deposition and applied it to the Gaussian plume model to quantify the errors of the source depletion technique. Unfortunately the surface depletion model is computationally expensive and hence is inappropriate for most applications of the Gaussian plume diffusion model. Further, it does not account for gravitational settling of large particles. An alternative plume depletion model has recently been added to the Environmental Protection Agency (EPA) PAL Gaussian diffusion model (Rao 1981). This model is attractive because it includes both settling and deposition, and because it is computationally inexpensive. The PAL

plume depletion model has been evaluated by comparing its predictions for nonsettling pollutants to the Horst surface depletion model.

The PAL plume depletion model is an exact solution of the advection-diffusion equation with a deposition boundary condition and a constant eddy diffusivity, or equivalently for σ_z proportional to the square root of downwind distance. The PAL and Horst models give identical results for the case of nonsettling particles and σ_z proportional to $\sqrt{x}$, where they are both an exact solution. However, this is not a physically realistic solution, since in the atmosphere σ_z near the source is linear in x . More realistic forms of σ_z are used in the model by applying the approximation $\sigma_z^2 = 2Kx/u$, where K is the eddy diffusivity and u is the wind speed.

For an unstable atmosphere the PAL model is particularly poor because σ_z is linear in x for all x (until mixing is limited by the top of the convective boundary layer). In this case the PAL model overpredicts the ground-level contamination $C(z=0)$ and the Chamberlain model is better. However, except for large particles this is not serious because deposition is usually small. For neutral conditions PAL still overpredicts $C(z=0)$, but its predictions fall between those of the Chamberlain and Horst models and thus it is an improvement on the Chamberlain model. For stable atmospheres PAL is also intermediate between the two models, until at large x it underpredicts $C(z=0)$. Except for unstable conditions the PAL model is generally a good approximation to the exact solution.

Two types of improvements have been made to the PAL model. Its predictions have been improved by forcing it to conserve mass and the range of applicability of PAL has been increased by improving its algorithms.

Since PAL assumes that σ_z is proportional to $\sqrt{x}$, it does not conserve mass when this is not the case. By keeping track of the losses due to deposition and forcing PAL to conserve mass, its predictions are noticeably improved, particularly for large deposition velocities and for unstable atmospheres. In the latter case the PAL model is then at least as good as the Chamberlain model.

PAL gives nominal, default returns for large values of quantities such as

$$(h - z - wx/u)/\sqrt{2} \sigma_z,$$

where h is the release height. This is no problem for small settling velocities w, since then $C(z=0)$ is very small and no deposition occurs. PAL has been improved,

however, to return the correct $C(z=0)$ for large settling velocities.

References

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• 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.

PARTICLE DENSITY EFFECTS ON DRY DEPOSITION VELOCITIES MEASURED IN WIND TUNNEL EXPERIMENTS

G. A. Sehmel and W. H. Hodgson

Particle dry deposition velocities have been measured in both field and wind tunnel experiments. Although many experimenters have investigated dry deposition for field conditions (Sehmel 1980), results have not been generalized because of limited control of experimental parameters. In contrast, parameters affecting particle dry deposition can be more readily investigated in wind tunnel experiments. As a result of this control, it was possible to develop a predictive dry deposition velocity correlation (Sehmel and Hodgson 1980). Predictions are based upon mass transfer resistances calculated from experimentally measured dry deposition velocities, which were evaluated from reference airborne concentrations measured 1 cm adjacent to deposition surfaces. However, dry deposition velocities are usually applied as a boundary condition at a height of about 1 m in meteorological transport, diffusion, and deposition models. Thus for ease of modeler usage, deposition velocities were subsequently calculated for a concentration reference height of 1 m by analytically combining mass transfer resistances below 1 cm and above 1 cm, from 1 cm to 1 m.

Particle dry deposition velocities predicted from the correlation are a function of the three parameters investigated: friction velocity, aerodynamic surface roughness, and monodispersed particle diameter for a 1.5 g/cm^3 particle density. The correlation was used to extrapolate deposition velocity predictions as a function of particle density at the 1-m concentration reference height. However, in the prediction calculations particle density effects were assumed negligible in the lower 1 cm adjacent to the deposition surface.

The objective of the experiments reported here was to experimentally investigate particle density effects on deposition velocities that are calculated using a concentration reference height near the deposition surface.

Experimental Procedures

Effects of particle density on dry deposition velocities were investigated using monodispersed uranine-traced particles in wind tunnel experiments. Uranine is a fluorescent water-soluble organic dye. The deposition surface was a crushed railroad rock, +3.8 cm to -5.1 cm (+1.5 in. to -2.0 in.) sieve size. Experiments were similar to prior experiments except for the particle densities investigated. Results to date were calculated using airborne concentrations from isokinetic samples collected 5 cm above the deposition surface. Both airborne concentrations and deposition fluxes were evaluated fluorimetrically from water solutions. Water washes were used to remove isokinetically collected and surface-deposited uranine in the uranine-traced particles.

The particle density was altered by adjusting the relative proportions of uranine (at a theoretical density of 1.5 g/cm^3) and of salts of greater density in the particles. Particle composition by mass was 70% salt and 30% uranine. Particles were generated from three salts: KI, $\text{Ba}(\text{NO}_3)_2$, and BaI with theoretical densities of 3.13, 3.24, and 5.15 g/cm^3 , respectively. In order to calculate particle densities, the assumption was that uranine and salts formed ideal solids. Particle densities were 2.4 g/cm^3 for the KI-uranine and $\text{Ba}(\text{NO}_3)_2$ -uranine particles and 3.0 g/cm^3 for the BaI-uranine particles.

Dry deposition velocities were investigated with particles generated by either a spinning disc aerosol generator (particle diameters

equal or greater than about $1.0 \mu\text{m}$) or a collision generator (particle diameters equal or less than about $0.9 \mu\text{m}$).

Results

Particle dry deposition velocities are shown in Figure 1 for particle densities of 1.5, 2.4, and 3.0 g/cm^3 and for average friction velocities of 15 and 107 cm/s. Deposition velocities can be significantly affected by both particle density and resuspension.

For the 15 cm/s friction velocity, deposition velocities are comparable for both particle densities of 1.5 and 2.4 g/cm^3 . However, significant deposition velocity increases are shown for particle diameters from about 1 to $2 \mu\text{m}$ for the largest particle density investigated (3.0 g/cm^3).

For the 107 cm/s friction velocity, deposition velocities are independent of particle density only in the diameter ranges from about 2 to $5 \mu\text{m}$. For diameters smaller than about $2 \mu\text{m}$, calculated dry deposition velocities increased rapidly as particle density was increased above 1.5 g/cm^3 . For diameters greater than about $6 \mu\text{m}$, concurrent resuspension occurred. Indeed for particle diameters

greater than about $9 \mu\text{m}$, the observed deposition velocities may be less than the gravity settling velocity for a particle density of 1.5 g/cm^3 .

Conclusions

Particle density effects on calculated dry deposition velocities can be significantly greater than predicted from the deposition velocity correlation, which was based on experimental deposition velocities for a constant particle density of 1.5 g/cm^3 . The greatest effects were observed in the "well" in which dry deposition velocities are often considered minimum as a function of particle diameter. These particle density effects need more evaluation.

References

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DEVELOPMENT OF A NEAR-MONODISPersed PARTICLE GENERATOR FOR FIELD DRY DEPOSITION EXPERIMENTS

G. A. Sehmel and W. H. Hodgson

A method for producing large quantities of monodispersed tracer particles is required for conducting improved field experiments in which particle removal by dry deposition is investigated. Large quantities of particles are required since airborne concentrations are rapidly decreased by diffusion and transport.

Particle dry deposition velocities are a function of particle diameter and other variables. For discussion purposes, values for these other variables are assumed to remain constant. Particle dry deposition velocities are nearly constant, and minimum, only for the particle diameter range from about 0.1 to $1 \mu\text{m}$. For particle diameters above about $1 \mu\text{m}$, dry deposition velocities increase rapidly with increases in particle diameter. The deposition velocity increase

FIGURE 1. Deposition Velocities to a Railroad Rock, +3.8 to -5.1 cm (+1.5 to -2.0 inch)

is continuous with increasing particle diameter until simultaneous reentrainment occurs. Reentrainment often occurs for particle diameters greater than about 10 μm . Between particle diameters of about 1 to 10 μm , the relative deposition velocity increase is proportional to a power function of particle diameter, d : d^n where the exponent n is 2 or larger.

If deposition velocities are investigated with polydispersed particle diameter tracers, experimental results are usually biased toward dry deposition velocities for the larger particle diameters. This bias reflects that experimental results (when tracer mass or concentration, not number, is measured) must be interpreted by considering particle mass distributions, not the particle number distributions. In order to relate particle dry deposition velocities to particle mass distributions, mass-weighted dry deposition velocities as a function of particle diameter are required. Summations are required: particle number distributions must be cubed for each particle diameter, and the cube multiplied by the deposition velocity for that particle diameter.

Although particle dry deposition velocities have been reported for many field experiments (Sehmel 1980), results have not been generalized, in part, because of insufficient control of tracer particle size characteristics. An experimental problem was that size distributions for tracers included wide ranges of particle diameters. This prior use of polydispersed particle size distributions is a severe restriction for developing generalized models from reported results.

The objective here was to circumvent the mass size-distribution effects by developing a particle generator for investigating particle dry deposition during field experiments. The development constraints are that all particles are in the 0.1 to 1 μm diameter range, the range in which dry deposition velocities are minimum and nearly independent of particle diameter.

Experimental Procedures

The tracer particle generator was developed by modifying a commercial paint sprayer. This paint sprayer has the general features of a laboratory-scale spinning disc aerosol generator used to generate monodispersed diameter aerosol particles. Essential features of this commercial device include a rotating bell (rather than the flat disc used in the

laboratory generator), an air-driven turbine for rotating the bell, and a governed speed control. Speed control of the bell is essential for generating monodispersed particle diameters. Rotational speeds are adjustable under liquid-feed conditions to 40,000 RPM by controlling air pressure to the air-turbine inlet.

The bell geometry is similar to a truncated cone with the bell axis concentric with the turbine drive shaft. In order to generate liquid droplets, liquid is injected into the bell into a hollow cone base adjacent to the turbine. Centrifugal force distributes this liquid to the outer edge of the hollow base. Subsequently, liquid flows through holes (drilled at the internal edge of the hollow cone base) uniformly onto the internal surface of the bell. Next, liquid flows along the internal surface of the expanding cone until droplets are formed at the base of the truncated cone. Liquid droplets are formed along the rounded edge of the base, which has a diameter of 6.8 cm.

Since airflow dissimilarities exist between this field generator and laboratory generators, both primary and satellite droplet sizes are generated with this field generator. Satellite droplets are smaller droplets formed during breakup of liquid bridges between primary droplets and the rounded edge. Satellite droplets are removed in the laboratory-scale spinning disc aerosol generator, while they are not in the paint sprayer. However, provisions for satellite particle removal could be incorporated into the paint sprayer system through airflow modifications.

Although satellite particles were not removed, the paint sprayer geometry was modified to remove some of the largest particles generated. Large particle removal was required since particle sizes were not monodispersed because the bell edge was rounded, instead of sharp as in a laboratory-scale particle generator. Removal of large particles was accomplished by liquid droplet impaction onto a concentric cylinder, 15 cm in diameter, which was added around the paint sprayer. The axis of the impaction cylinder extended approximately 30 cm in front of the paint sprayer, and approximately 15 cm in back.

The particle size distribution characteristics of generated solid particles were measured within a closed space, a wind tunnel for which the air-circulating blower was not turned on. Particle size distributions were measured with a 10-stage quartz crystal

microbalance (QCM)[®] particle cascade impactor. The 50% collection efficiencies for each QCM stage are 0.05, 0.1, 0.2, 0.4, 0.8, 1.6, 3.2, 6.4, 12.5, and 25 μm . This QCM permitted investigation of both particle size distributions and times required in the wind tunnel to obtain dry particles by complete evaporation of liquid.

Particles were generated from a 1 g/liter solution of uranine dye in a solution of 20% water and 80% ethyl alcohol. Different solution feed rates were investigated as a function of particle size distributions. These included liquid feed rates used subsequently in two field experiments. Satisfactory solution feed rates included 219 and 113 $\text{cm}^3/\text{minute}$, with 31 and 15%, respectively, of the solution lost by impaction on the concentric cylinder.

After impaction, all particles were collected on the 0.05 to 1.6 μm stages of the QCM. Although statistics need further evaluation, approximately 70% by weight of the particles were collected on the 0.8 and 1.6 μm stages.

Conclusions

Operating characteristics of a commercial rotary paint sprayer were investigated for the purpose of developing a monodispersed particle generator for releasing airborne tracer particles in field conditions. Results were satisfactory, but further improvements are planned by adding provisions for satellite particle removal and using a sharper edged rotating bell.

Generated tracer particle diameters were nearly all within the diameter range for which particle dry deposition velocities are minimum and nearly independent of particle diameter. Consequently, the particle generator was used in field dry deposition experiments (uranine particle release heights of 1.4 and 2.5 m on two nights) concurrently with tracer releases on 19 October and 4 November 1982 sponsored by the Environmental Protection Agency (EPA) at the Hanford meteorological diffusion grid. The purpose of the EPA experiments was to investigate particle dry deposition with depositing ZnS tracer particles (density of 4.1 g/cm^3) and nondepositing SF_6 tracer gas. After collected filter samples are analyzed for ZnS for the EPA experiment, filter samples will be subsequently analyzed for uranine tracer-particle concentrations.

Expected results from airborne uranine tracer particle (density of 1.5 g/cm^3) concentration measurements and comparisons with the other two tracers will include particle size and density effects on dry deposition, more refined evaluation of the minimum dry deposition removal rates as a function of particle diameter, and an indication of diffusion-transport effects for multiple tracer releases.

Reference

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[®]Model PC-2 QCM Cascade Impactor, California Measurements Incorporated, Sierra Madre, California.

• 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.

FUGITIVE EMISSION FLOW PATHS INVESTIGATED WITH TRACERS IN COMPLEX TERRAIN OF THE OIL SHALE REGION OF COLORADO

G. A. Sehmel

Increased airborne particulate concentrations could result from the production of shale oil, and thus limit development of this resource. In addition, spent-shale disposal areas could become prime fugitive emissions sources. However, the prediction accuracy of downwind airborne concentrations is uncertain in the complex terrain of the oil shale region in western Colorado. Consequently, atmospheric tracer investigations were conducted from a proposed spent shale disposal site (in July 1981) to decrease this uncertainty at Federal oil shale lease Tract C-a. Results have been reported (Sehmel 1982) and are summarized here. The objective of the July 1981 experiments was to simulate pollutant transport and dispersion from a proposed spent-shale disposal site during both day and nighttime conditions. The terrain is a sloping, undulating area with many tributaries between hilltops along Corral Gulch and Dead Horse Ridge.

Experimental Procedures

Airborne tracer concentrations were investigated using one tracer release site and multiple airborne concentration sampling sites. The tracer simulant was SF₆ gas, a nondepositing and nonreactive gas. Tracer release times ranged from 3 to 9 hours. There were four experimental time periods: two in the evening and two in the morning. Tracer release times included airflow transition times around sunset and sunrise. Airborne tracer concentrations were determined from air samples collected in gas bags and syringes. The time-integrated gas-bag samples were collected at 32 radio-controlled sampling sites, which were from 0.6 to 6 km

from the tracer release site. The near-instantaneous syringe samples were collected as a function of time by sampling along roads at distances of 3 to 13 km from the tracer release site. Meteorological data that were recorded include wind speed, wind direction, and vertical temperature difference ($T_{60\text{ m}} - T_{10\text{ m}}$). There were three meteorological measurement sites in addition to one at the tracer release site.

Results

Meteorological data and airborne SF₆ concentrations were summarized (Sehmel 1982) for each experiment. Concentrations were shown as a function of identical time periods at the integrated gas-bag-sampling sites (sampling times were identical since sites were activated and deactivated by radio control), and as a function of time and distance along roads with the syringe-collected samples. Although 32 radio-controlled sampling sites were used, the additional syringe sampling sites were desirable to increase understanding of local airflow in complex terrain. The results from the syringe samples, though near-instantaneous samples, complemented and were used to further explain concentrations determined from the integrated gas-bag samples. Concentrations from the more spacially frequent syringe samples confirm, in more detail, plume-location and plume-width results from gas-bag samples.

Conclusions

Fugitive emissions transport and dispersion were simulated in tracer-gas experiments during conditions of relatively low wind speeds; i.e., wind speeds less than the threshold wind speed causing soil suspension. Although spent shale will suspend due to wind stresses only for greater wind speeds, fugitive emission rates will be nearly independent of wind

speed if suspension were mainly caused by mechanical stresses. For instance, constant vehicular traffic on spent shale can produce relatively constant fugitive emission-flux source terms that are nearly independent of wind speed. If this proposed emission scenario occurs, maximum downwind airborne concentrations will similarly occur during low wind speed conditions. Airborne concentrations will remain relatively large since airborne pollutant concentrations will not be diluted by high wind speeds, u ; i.e., downwind airborne concentration decreases are proportional to $1/u$.

Multiple plume flows and plume interactions were identified for fugitive emissions transport from the proposed spent shale disposal site. The tracer plume segmented. The greatest concentrations occurred during drainage flows. Plume interactions were between traced plumes and ambient air plumes. Since multiple plume flows and plume interactions were identified, model predictions of downwind transport of fugitive emission plumes on a local scale must include predictions of plume segmentation and interaction.

Predictions on a local scale are essential if maximum "fence-line" concentrations are considered. Two parameters in model predictions are fugitive emission rates, Q , and airborne concentrations, χ . The only controllable parameters in these models are pollutant release heights, release times, and release rates. Predictions of χ/Q ratios at the fence-line, and downwind, are used both by industry and in regulations. Although further analysis is planned for evaluations of predictive models, the χ/Q ratios can nevertheless be estimated from the reported tracer concentrations and the constant tracer release rate.

Reference

Sehmel, G. A. 1982. "An Atmospheric Tracer Investigation of Fugitive Emissions Transport in the Colorado Oil Shale Region". Presented at the Fifth Symposium on Fugitive Emissions: Measurement and Control, May 3-5, 1982, Charleston, South Carolina.

SIMULATION OF PLUME DEPLETION OF FUGITIVE EMISSIONS WITH DUAL TRACER PARTICLE DRY DEPOSITION MEASUREMENTS IN BRUSH CREEK, COLORADO

G.A. Sehmel

Particle dry deposition velocities are a function of particle diameter when other con-

trolling parameters are constant. Deposition velocities are minimum, and nearly constant, for a particle diameter range from about 0.5 to 1 μm . This diameter range is estimated from both experimental field investigations (Sehmel 1980) in which polydispersed particle sizes were used and from empirical predictions (Sehmel 1980, Sehmel and Hodgson 1980) based upon wind tunnel particle dry deposition experiments with monodispersed particles.

The objective here (Sehmel 1982) was to investigate dry deposition removal of fugitive emissions (for the particle diameter range of minimum dry deposition) by using dual tracers during nocturnal drainage flows in a canyon. Nocturnal flows were investigated because of the time and direction persistences.

Theoretical Approach

The tracers were a nondepositing gas, (SF_6), and depositing lithium-traced particles (Li_2CO_3). The experimental technique was to release both tracers at constant release rates at the same release location and measure concentrations, χ , downwind simultaneously for both tracers. Airborne tracer concentrations were measured with sampling instruments located at one sampling location (at multiple sampling heights). Subsequently, plume depletion by particle dry deposition was calculated from tracer concentration ratios, R , i.e., the tracer particle concentration (χ_p) divided by the SF_6 tracer gas concentration (χ_g).

The fractional plume depletion was calculated by integrating tracer fluxes as a function of sampling height to obtain the average tracer particle and gas concentrations. These average concentrations were compared to the maximum tracer concentrations which were measured. Maximum concentrations were assumed to correspond to concentrations for which the plume was undepleted by dry deposition. The fractional plume depletion up to the highest concentration measurement height, 15 m, is

$$\text{Fractional Depletion} = \frac{(\chi_{\text{SF}_6})_{\text{maximum}}}{(\chi_{\text{particles}})_{\text{maximum}}} \frac{\int \chi_{\text{particles}} u \, dz}{\int \chi_{\text{SF}_6} u \, dz} \quad (1)$$

where u is the windspeed at height z .

Experiments

Tracer release procedures minimized differential plume cooling of SF₆ gas and subsidence at the release site. The SF₆ tracer gas was released through rotometers from gas cylinders in which the SF₆ was in the gas phase. The lithium-traced particles were generated from a water solution of Li₂CO₃ sprayed through nozzles. Experiments were run on four nights. Both tracers were released at elevated heights: 8.5 m on the first night and 12 m on the other three nights.

The tracer concentration sampling tower was centrally located in the canyon 2.8 km downwind from the tracer release site and was immediately adjacent to the downwind edge of a forested area. The average tree height in this area ranged from about 10 to 20 m. Depending on wind direction, the forested fetch length immediately upwind from the sampling tower ranged from about 0.04 to 0.3 km.

Simultaneous tracer sampling was a function of either 4 or 5 heights, from 7 cm to 15 m. In addition, cup and vane anemometers were located at 5 heights from 0.3 to 11 m. Tracer concentrations were determined using real-time detectors for sensing lithium-traced particles and gas chromatographs for measuring SF₆ tracer gas concentrations. The

SF₆ tracer gas concentrations were determined using gas bag-collected samples in which concentrations were integrated with time. Concentrations were usually determined for time periods of about 50 minutes.

Results

Results for the four nights' experiments are summarized in Table 1. With only one exception, for the experiment on July 29 to July 30 between 0130 to 0240 MDT, calculated plume depletion (by dry deposition below 15 m height) ranged from 0 to 16%, with an average of 8% plume depletion for the 9 experimental time periods.

Conclusions

Since the tracer arrival speed was always greater than measured wind speeds at either the tracer release or sampling tower sites, the tracer was vertically mixed into the air (above the wind speed measurement heights) and was transported downwind by winds at elevated heights. Thus, these measured plume depletions have general applicability and may typify dry deposition removal from an elevated source, as well as from a near-surface source release.

TABLE 1. Particulate Plume Depletion Below 15 m Height in Brush Creek.

Date July-August 1982	Experimental Time Periods, MDT		Speeds, m/s				Integrated Plume Depletion Below 15 m, %	
			Wind at Release	Tracer Arrival	Sampling Tower Winds			
	Release	Sampling	Tower, 13 m	Time	4.5 m	11 m	Exp.	Average
27-28	0047			1.9		2.1		11
	to	0210-0310			0.7	1.3	9	
	0618	0502-0552			0.7	1.1	12	
29-30	2200		0.9	1.9		1.1		
	to	0130-0240			1.0	1.1	4	
	0713	0340-0430			1.0	1.3	16	9
		0530-0620			1.0	1.4	7	
31-1	0003		0.7	1.9		1.6		1
	0700	0230-0330			0.9	1.2	0	
		0450-0550			0.8	1.0	1	
4-5	2117		0.7	1.9		1.5		10
	to	2310-2400			0.7	1.4	5	
	0220	0110-0200			1.0	1.3	15	

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of 1977 and Energy Considerations for Air Pollution Control. Symposium Series No. 196, Vol. 76, pp. 218-230, American Institute of Chemical Engineers, New York, New York.

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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.

WIND RESUSPENSION INVESTIGATED WITH REAL-TIME TRACER CONCENTRATION MEASUREMENTS

G. A. Sehmel

Airborne concentrations of wind-resuspended surface contaminants and tracer simulants have been investigated (Sehmel 1982) as a function of several parameters, including time since pollutants were originally deposited on the ground surfaces. Changes in airborne concentrations with time have been described (Sehmel 1980) by a weathering half-life, the time required for airborne concentrations to decrease to one-half the original value. This time is expected to depend upon wind speed and vary according to the availability of surface contamination to resuspend.

The objective of this investigation was to develop an experimental technique for investigating weathering half-lives and the wind speed dependencies of airborne concentrations by measuring airborne concentrations of a resuspended lithium-traced particulate tracer in realtime. Only the measurement technique was evaluated.

Weathering half-lives need further experimental evaluation since the historical data for weathering half-lives are inconsistent and possibly misleading. Airborne concentrations of a radionuclide after the Plumbob nuclear test decreased with a weathering half-life of 35 to 40 days (Wilson et al. 1960). More recent results (Olafson and Larsen 1961) near the same study location indicated a great uncertainty about these half-lives. In the original study starting in May 1957 (Wilson et al. 1960), the 35-day half-life was determined for an air sampling time period of less than 5 months. Subsequently in the summer of 1958, airborne concentrations were measured intermittently for times up to 15 months. In comparing results from these two studies, Healy (1974) reported no appreciable change in airborne concentra-

tions within the data scatter at a site 230 m from the original test location.

There is further evidence from the literature on airborne radionuclides that weathering half-lives are on the order of years. This evidence is deduced from the fact that airborne plutonium concentrations currently obtained during monitoring activities at nuclear locations are usually near fallout concentrations. In contrast if the 35- to 40-day half-life were valid, back calculation from current concentrations to predict concentrations in earlier years would predict relatively high airborne plutonium concentrations. At least to the author's knowledge, such high concentrations were not measured in earlier years.

Weathering half-lives on the order of years are also supported (Sehmel 1982) by results from wind resuspension experiments using inert tracer particle sources (calcium molybdate). Results from these experiments show that concentrations of airborne resuspended tracer are a function of wind speed and show seasonal variations, resuspension rates being greatest in the fall.

Experiments

Real-time lithium-traced particle resuspension was investigated between November 1981 to April 1982 using an aged resuspension source of lithium carbonate particles. The site was near the Hanford Meteorological Station (HMS) in a sandy area (Sehmel and Lloyd 1976). The source was originally deposited from a water solution sprayed uniformly (using a portable garden sprayer) on the ground on October 3, 1980. The average source strength was 2.4 g/m².

Airborne tracer particle concentrations were measured with two lithium detectors in each experiment, one 7 m upwind of the source and

the other at the downwind edge of the tracer source area. The length of the tracer source area between detectors was 30 m. Lithium detectors were placed to investigate concentrations of resuspended tracer particles during high wind speeds, from a direction of about 225°.

Since the lithium detectors are sensitive to some particles other than lithium, the purpose of the upwind detector was to measure background concentrations of particles to which the lithium detectors might respond. In order to evaluate concentrations of resuspended lithium-traced particles, these background concentrations were to be subtracted from concentrations measured downwind.

Airborne concentrations of lithium-traced particles were investigated as a function of time near respiration height, 1.5 m. Airborne concentrations were investigated with flame photometric detectors. Sampled air was drawn into a detector at a flow rate of 50 cm³/minute and burned in a hydrogen flame. The flame spectra for lithium is at 6708 Å. For recording tracer particle concentrations, the flame spectra are transmitted through a narrow band-pass filter to a photomultiplier tube. The wave length for this filter is 6717 Å and has a full-width half-maximum (FWHM) of 1040 Å. The filter transmission is about 45% over a band width of 50 Å.

Results

Airborne concentrations were measured during several time periods of expected high wind speeds. Concentrations for one day are shown in Figure 1. Although concentrations increased downwind, the data are qualified by detector responses for other time periods during which concentrations also increased at the upwind site.

There are three possible explanations for simultaneous concentration increases at the upwind site: the extent of the tracer source had spread by prior resuspension in the upwind direction, the detectors responded to the continuous light spectra of soil particles heated in the hydrogen flame, and the detectors responded to the continuous light spectra of combusted particles. It is not known which explanation dominated detector response.

FIGURE 1. Airborne Particle Concentrations on Feb. 17, 1982 (Calculated from Lithium Detector Counts).

Conclusions

Real-time airborne concentration measurements of resuspended lithium-traced particles are confounded by detector response to other than tracer particles. Nevertheless, the real-time measurement technique shows promise for investigating real-time airborne concentrations from resuspension if spurious light spectra can be eliminated. Further experimental evaluations are planned using a narrower band pass filter, a 20 Å band pass around 6708 Å, in order to reduce spurious signals.

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TRACER RESUSPENSION SOURCE-STRENGTH SPREAD ON THE SURFACE BY WIND RESUSPENSION.

G. A. Sehmel

Wind-caused particle resuspension rates have been investigated with molybdenum tracer (calcium molybdate) at a 29.9 m circular resuspension site near Horn Rapids Dam in the Hanford area. The tracer was deposited 5-1/4 years ago, on May 29, 1977. Resuspension rates were investigated previously (Sehmel 1982) by sampling resuspended tracer with air sampling equipment mounted as a function of height on a centrally located sampling tower at each site. These sampling systems automatically collected resuspended tracer particles as a function of wind speed increments, sampling height, and aerodynamic particle diameter.

The objective of this investigation was to experimentally confirm resuspension rates evaluated using the central air sampling tower by investigating resuspension-source area changes and spread.

Ground source strength data are desired since resuspension rates were calculated using several assumptions to derive airborne tracer fluxes from measured airborne tracer concentrations. Isokinetic correction factors as a function of particle size were assumed negligible, wind speeds were assumed independent of height, crosswind diffusion was assumed negligible, and the cross-products of fluctuating concentrations and wind speed were assumed negligible compared to the products of average concentration multiplied by average wind speed.

Resuspension rates were calculated from total airborne tracer exposures (A) per unit width (m) as a function of height, tracer ground sources (G) from the edge of the tracer area to the sampling tower, and sampling times. Total airborne exposures (g/s) were calculated by integrating airborne fluxes as a function of height. Resuspension rates ranged from about 10^{-12} to 10^{-7} fraction resuspended per second. In general, resuspension rates increased as power functions of wind speed, U^n , where the exponent n was greater than 3.

Experiments

Surface soils were sampled in September 1982 to investigate tracer-source changes and strengths in the original area and beyond. In order to investigate these changes and source strengths, surface soils at each sampling site were collected from a 84 cm² surface area. The average sampling depth was 1 cm, a soil depth which permitted soil sampling below the cheat grass and detritus. Surface-soil samples were collected along concentric circles from the sampling tower.

After soil collection, soil samples were dried, weighed, and leached with 2 M citric acid for dissolving and chelating the molybdenum. Leach solutions were analyzed with an induction-coupled argon plasma atomic emission spectrometer (ICP) for molybdenum.

Results

As shown in Figure 1, tracer-source strengths ranged from about 10^{-4} to 10 g Mo/m² in September 1982, 5-1/4 years after the tracer was deposited. Initially, the tracer source strength was 3.1 g Mo/m² within a radial distance of 29.9 m from the sampling tower. By September 1982, the average source strength within this original tracer-source area had decreased to 1.9 g Mo/m², which was only a 39% decrease.

Tracer source-strength decreases with distance were not uniform as a function of direction from the tower. These nonuniformities might reflect preferential resuspension by wind gusts in the nonuniform vegetative cover, or by inadequate soil sampling. Also, the tracer source had spread on the ground during the 5-1/4 years after deposition. If high wind speeds were the principal cause of source spread on the ground as a function of direction, the near-uniform angular spread is unexpected as a function of direction. A

FIGURE 1. Tracer Source Strength per m² of Ground Surface as a Function of Direction and Distance

preferential source-strength decrease from the original tracer area was expected since the frequency of high wind speeds (i.e., highest expected resuspension rates) is highest from near 225°. This decrease was not observed. Furthermore, a preferential increase in source strength beyond 29.9 m at a direction of 45° was expected since tracer transport by surface creep and saltation also should have been greatest in the direction of high wind speeds. This increase also was not observed.

Conclusions

Wind-caused resuspension rates for respirable-size particles range from about 10^{-12} to 10^{-7} fraction resuspended per second when the resuspension source strength is assumed independent of tracer particle size (Sehmel 1982). Resuspension rates on the order of 10^{-7} fraction resuspended per second correspond only to high wind speeds; yearly average wind speeds are much lower. These resuspension rates infer that the tracer source on

the ground does not appear to rapidly change since there are 3.15×10^7 sec/yr.

A confirmation of the experimental air sampling technique and adequacy for measuring resuspension rates lies within the following interpretation of tracer source-strength data and an average resuspension rate. If an average resuspension rate for 5-1/4 years is assumed to cause the 39% average source-strength depletion shown in Figure 1, the 5-1/4 year average resuspension rate is 3.7×10^{-9} fraction resuspended per second. This average resuspension rate is comparable to resuspension rates determined by air sampling.

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Theoretical Studies of Atmospheric Processes

THEORETICAL STUDIES OF ATMOSPHERIC PROCESSES

- **Theoretical Studies and Applications**

Mathematical models, needed to predict consequences of pollution releases and controls, rely on understanding of the governing atmospheric processes. In turn, this understanding requires the synthesis of theory and data. For example, modeling the wet and dry deposition and resuspension of air pollutants requires the synthesis of results from laboratory and field experiments with statistical and other descriptions of the turbulent transfer of momentum, heat, and matter. Chemical and physical transformations of released pollutants are other processes where this synthesis is needed. Knowledge gained from these syntheses can then be used to improve models that simulate the fate of atmospheric pollutants.

During FY 1982, these studies have emphasized modeling of the global-scale fate of atmospheric trace constituents. New results have been obtained for the particle-size dependence of the tropospheric residence time of aerosol particles, and for the wet and dry fluxes of particles and gases to the oceans. New results have also been obtained that are essential in experimental studies of dry deposition of acidic species. Two other highlights in the project have been the hosting of two international conferences: the NATO Advanced Study Institute on the Air-Sea Exchange of Gases and Particles, and the Fourth International Conference on Precipitation Scavenging, Dry Deposition, and Resuspension.

• Theoretical Studies and Application

Objectives of this study are:

Developing theoretical models to characterize and describe the fate of energy-related air pollutants.

Translating theory and data into forms most useful for practical application in evaluating the consequences of air pollutant releases.

THEORETICAL STUDIES AND APPLICATIONS

W. G. N. Slinn

This brief report summarizes progress in this project during FY-1982, in turn reflecting progress on topics explored continuously by the Principal Investigator (P.I.) during the past fifteen years. For a more coherent description of this year's progress, the reader is referred to the 107-page chapter entitled "Air-to-Sea Transfer of Particles" (Slinn 1983a), to be published in a book co-edited by the P.I. (Liss and Slinn 1983).

Aerosol Size Distributions

In 1974, the P.I.'s survey of available data resulted in a plot of typical aerosol-particle "size distributions" in surface-level air (Slinn 1975). This plot has subsequently appeared in a number of undergraduate- and graduate-level textbooks. More recent data has led to the revised plot shown as Figure 1 (Slinn 1983b).

Tropospheric Residence Times

If the continuity equation for a trace substance in the atmosphere is integrated over the volume in the troposphere into which the substance has diffused, and if Greens' theorem is used, then the overall removal rate, τ^{-1} (or tropospheric residence time, τ) is found to be

$$\frac{1}{\tau} = \frac{1}{\tau_w} + \frac{1}{\tau_d} + \frac{1}{\tau_{\text{coag}}} + \dots \quad (1)$$

in which separate (first-order) removal rates have been identified, arising from precipitation scavenging, dry deposition, coagulation, etc. With use of estimates for particle-size dependent wet- and dry-deposition velocities (Slinn 1974; 77; 82a,b; Slinn and Slinn 1980; 81), there results the particle-size-dependent residence times shown in Figure 2 (Slinn 1983c).

Precipitation Scavenging

Wet deposition can be described in terms of scavenging rates, ratios, or efficiencies. During this reporting period, inter-relations among these different descriptions have been derived (Slinn 1983a). Also, the description of scavenging as a stochastic process has been extended beyond earlier descriptions (Slinn et al. 1978, Slinn 1982b). Thus, for the case when the number of storms that the trace constituent experiences is a random variable, $\tilde{n}$, then the fraction of the initial pollution remaining airborne (see Figure 3) is

$$q(t)/q_0 = \prod_{j=1}^{\tilde{n}} (1 - \varepsilon_j), \quad (2)$$

where ε_j is the fraction of the trace constituent scavenged by storm j (with the time, during which the trace constituent is exposed to each storm, short compared to τ_w). Then, for independent storms, the expected value is easily found to be

$$\bar{q}/q_0 = \exp \{-\bar{\tau}/\tau_w^*\}, \quad (3)$$

where $\bar{\tau}^* = [\bar{v} |\ln(1-\bar{\varepsilon})|]^{-1}$, in which $\bar{n} = \bar{v}t$, $\bar{v}$ is the (Lagrangian) average frequency of storms, and $\bar{\varepsilon}$ is the mean of ε_j . This is essentially the same result as for the case when n is deterministic (Slinn 1982b). However, the coefficient of deviation is different. In particular, if the storms form a Poisson process with a common mean and variance for ε_j , then

$$[c.d.]_{\ln q_n/q_0} = \quad (4)$$

$$\frac{\bar{n}^{1/2} [\ln^2(1 - \bar{\varepsilon}) + \sigma_{\ln(1 - \bar{\varepsilon})}^2]^{1/2}}{\bar{n} \ln(1 - \bar{\varepsilon})} \sim \frac{1}{\sqrt{\bar{v}t}},$$

FIGURE 1. Indications of Aerosol Particle "Size Distributions" (here, surface area per unit volume) for a Number of Sources at some "Typical" Locations (from Slinn, 1983b, where data sources are referenced).

which differs from an empirical result obtained by Junge (1974) for gases, but agrees with other results obtained by an entirely different theoretical method (Gibbs and Slinn 1973, Slinn 1982b).

Dry Deposition and Resuspension

Several new results in dry deposition and resuspension have been obtained during this reporting period. They include (see Slinn 1983a, 83d):

- Demonstration of lack of convergence of K-theory formulations near an air-surface interface;

- Reasons for potential failures of second-order closure models of turbulence;
- Explanations for failures of classic theories of mass transfer (by von Karman, Prandtl, and others);
- Causes of inadequacies of similarity theories for mass transfer from turbulent flows;
- Suggestions for the Schmidt and Stokes number dependencies of the mass transfer velocity in air, considering the emerging picture of turbulence near a solid interface (see Figure 4);

FIGURE 2. Estimates for Average, Wet and Dry Deposition Velocities (top) and Tropospheric Residence Times (bottom), as Functions of Aerosol Particle Size (from Slinn, 1983c).

- Descriptions of the influence of the Richardson number on the mass transfer speed in air, k_a , and of the Rayleigh instability on mass transfer speed in a liquid, k_ℓ ;
- Predictions for the influence of waves on k_ℓ and on wave breaking and spray scavenging on k_a ;
- Reasons for the failure of the radon-deficiency method to yield meaningful values for k_ℓ at sea;
- A proposed simple method to account for resuspension by introducing a volumetric gain term in the continuity equation, and retaining the perfect sink boundary condition (see Figure 5).

Another new development, and one that is relatively easy to describe, is the probable reason for failure of eddy-flux and concentration-gradient methods to yield meaningful data for the dry-deposition velocities of submicron particles. The cause appears to be the failure of experimentalists to satisfy the constant-flux condition. To see this quantitatively, the continuity equation is averaged over a proposed constant-flux layer of height h , a Reynolds average is performed, and then the equation is nondimensionalized using the characteristic wind speed $\bar{V}$, length scale for horizontal changes L , and deposition velocity $\sigma C_D \bar{U}$, where C_D is the drag coefficient and $\sigma < 1$ therefore includes influences on deposition from properties of the surface and the

FIGURE 3. Example of the Stochastic Nature of Precipitation Scavenging, Sketched for Scavenging Events Short Compared to the Average Tropospheric Residence Time of the Atmospheric Trace Constituent (from Slinn, 1983a).

FIGURE 4. Sketch of the Emerging Picture of Turbulence Near a Smooth Surface. A picture used to develop predictions for the Schmidt and Stokes number dependencies of the dry deposit velocity (from Slinn, 1983a).

FIGURE 5. A Schematic Indication of the Proposed Method to Replace the Interfacial Boundary Condition for Resuspension by a Volumetric Gain Term, $G(Z)$, Near the Interface (from Slinn, 1983a).

the surface and the trace constituent. For the vertical flux to be essentially independent of height, the dimensional analysis yields the requirement:

$$L \gg h/(\sigma C_D) . \quad (5)$$

For example, for a 10-m constant-flux layer ($=h$), for momentum ($\sigma = 1$) over grass ($C_D \approx 10^{-2}$), then it is necessary to be downwind from nonuniformities in the momentum field a distance $L \gg 10 \text{ m}/(10^{-2}) = 1 \text{ km}$. However, for a 10-m constant-flux layer for $0.1 \mu\text{m}$ particles, with $\sigma \approx 10^{-2}$ for grass (Slinn 1982a), then $L \gg 100 \text{ km}$, a condition

that has not been satisfied in reported results. In other words, it is likely that the reported values of large (positive and negative) deposition velocities, measured by concentration-gradient and eddy-flux methods, actually reflect measurements of vertical fluxes derived from horizontal convergences and divergences, not deposition (and resuspension). Further support for this contention (viz., data for v_d obtained by other methods) is described elsewhere (Slinn 1983e).

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Author Index

AUTHOR INDEX

Cope, V. W.; 1, 2, 59

Doran, J. C.; 15, 16, 57, 59

Heimbigner, T. R.; 9

Hodgson, W. H.; 33, 34

Horst, T. W.; 15, 16, 31, 57, 59

Kalkwarf, D. R.; 1, 2, 59

Lee, R. N.; 9, 23

Orgill, M. M.; 9, 11, 23, 26, 59

Schreck, R. I.; 23

Sehmel, G. A.; 33, 34, 37, 38, 41, 43, 57, 59

Slinn, W. G. N.; 49, 58, 59

Thorp, J. M.; 26, 58

Whiteman, C. D.; 9, 58

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