
**Pacific Northwest Laboratory
Annual Report for 1975 to the
USERDA Division of Biomedical
and Environmental Research
Part 3 Atmospheric Sciences**

March 1976

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PACIFIC NORTHWEST LABORATORY ANNUAL
REPORT FOR 1975 TO THE USERDA DIVISION
OF BIOMEDICAL AND ENVIRONMENTAL RESEARCH
PART 3 ATMOSPHERIC SCIENCES

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

March 1976

Battelle
Pacific Northwest Laboratories
Richland, Washington 99352

PREFACE

The Annual Report for 1975 to the U.S. Energy Research and Development Administration's Division of Biomedical and Environmental Research is organized by major categories according to our schedule-189 submissions. Each part is directed toward a particular DBER Research and Development Program: Part 1 to Biomedical Programs, Parts 2 and 3 to Environmental Programs, and Part 4 to Physical and Technological Programs. Each part of the Annual Report comprises project reports authored by scientists from several research departments, reflecting the interdisciplinary nature of the research effort. The Annual Report consists of four parts:

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Annual Report for

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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

W. J. Bair
Manager, Biomedical and Environmental
Research Program

FOREWORD

Given source information for effluents from energy development and utilization, atmospheric transport research provides the necessary tools to describe pollutant behavior in the atmosphere. This behavior includes the action of the atmosphere to modify and distribute pollutants on local, regional, and global scales. It also includes the action of pollutants to alter atmospheric properties and transport mechanisms. Ultimately, the atmosphere delivers pollutants to man and to ecosystems where their effects must be determined. To assure an adequate understanding of pollutant behavior in the atmosphere and its delivery to biological systems, the following atmospheric transport research is being conducted.

- Surveys of effluents released to the atmosphere
- Air flow and dispersion behavior of pollutants
- The disposition and resuspension of contaminants
- The removal of the pollutants by scavenging in clouds and rain
- Physical and chemical transformations of pollutants, as they occur during transport
- Integration of models of pollutant dispersion, transformation, and removal processes
- Effects on weather and climate.

These atmospheric processes are now primarily being investigated within the context of problems linked to the development and utilization of two energy resources: coal and nuclear energy. We plan future investigations of atmospheric problems caused by development of oil shale technology.

Coal Combustion

The behavior of air pollution, involving the physical and chemical transformations that occur during transport and the removal of pollutants by wet and dry scavenging processes, is being defined in connection with expanding coal combustion for power production in the northeast United States. One of the most pressing problems from coal utilization is the release of large quantities of sulfur dioxide to the atmosphere, its transformation to sulfates, and its removal and ultimate delivery to biological systems where serious health and environmental consequences can result. The greatest emphasis in atmospheric transport studies will be placed, in the future, on this area of fossil fuel pollutant transformation and removal. We anticipate additional reorientation of research previously focused on nuclear problems. This expanded non-nuclear research will be conducted in the Multi-State Atmospheric Power Production Pollution Studies (MAP3S).

Nuclear Energy

The concern for long-lived particulates released to the environment from LMFBR operations dictates that research on particle deposition and resuspension processes be conducted in the context of nuclear power production problems. The primary hazard from plutonium is through inhalation; therefore its residence in the atmosphere must be clearly defined. Thus, removal of particulates from the atmosphere by deposition processes (which limits initial exposure) and any future resuspension from the surface presents (a continuing potential for inhalation hazards) are being evaluated. Research is also being done on waste heat inputs to the atmosphere from cooling towers. Proposed

nuclear power parks will concentrate such releases of heat and moisture, representing energy densities comparable to those of severe storms and suggesting potential weather modifications. Thus, a significant fraction of the atmospheric transport program must remain associated with nuclear problems.

Oil Shale

In the future, the significant atmospheric processes connected with another developing energy technology - shale oil production - will be investigated. The general need to understand air flow and dispersion in complex terrain, especially important throughout the western United States, will be approached in connection with oil shale developments in Colorado, Utah and Wyoming. Recent efforts to accelerate the development of synthetic fuels have increased the need for an interdisciplinary evaluation of the health and environmental problems associated with oil shale mining and retorting. PNL plans to undertake this evaluation. The mountainous oil shale region represents a particularly difficult air pollution problem; air is trapped in valleys for extended periods under certain meteorological conditions, and thus especially stringent siting requirements must be fulfilled to meet State and Federal air quality standards. Adequate models are not available for the complex air flow and dispersion conditions of the area; they must be developed to assure acceptable siting.

CONTENTS

(Listing in parenthesis denotes the sponsoring
programs under which the research was done.)

PREFACE.	ii
FOREWORD	iii
<u>ATMOSPHERIC SURVEYS</u>	
Ozone, Radionuclide and Particulate Correlations in Air at Quillayute, Washington - J. D. Ludwick and T. D. Fox (Survey of Ambient Airborne Pollutants)	1
Lead (Pb) Concentrations Associated with 1000 mb Geostrophic Back Trajectories at Quillayute, Washington - T. D. Fox and J. D. Ludwick (Survey of Ambient Airborne Pollutants)	11
Atmospheric Fallout During 1975 at Richland, Washington and Point Barrow, Alaska - C. W. Thomas (Radioactive Fallout Rates and Mechanisms)	16
The Concentrations of ^{241}Pu and ^{99}Tc in Surface Air at Richland, Washington - C. W. Thomas (Radioactive Fallout Rates and Mechanisms)	20
Solvent Refined Coal--Organic Constituents Present in the Process - M. R. Petersen (Atmospheric Chemistry of Coal Conversion Pollutants)	23
Baseline Air Quality Data from Colstrip, Montana--E. A. Crecelius, J. C. Laul, L. A. Rancitelli and R. L. McKeever (Chemical and Physical Properties of Pollutants in the Colstrip Power Plant Effluents)	25
The Use of Elemental Concentration Ratios to Distinguish Between the Plumes of Different Northeastern Cities - J. A. Young, N. S. Laulainen, L. L. Wendell and T. M. Tanner (Tracer Studies in the METROMEX Experiment)	27
<u>AIR FLOW AND DISPERSION</u>	
A Simple Development of Covariant Differentiation - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	35
A Pitfall in the Application of Tensor Analysis to Fluid Mechanics - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	37
Diffusion with Deposition at Short Range - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	39
Some Observations of the Relative Contribution of Real Plume Diffusion and of Meander to Apparent Plume Diffusion - P. W. Nickola (Atmospheric Boundary Layer Studies)	43
Variation of Plume Centerline Concentration with Time - P. W. Nickola (Atmospheric Boundary Layer Studies)	48
Field Measurement of the Effect of Filter Orientation on Sampling of a Particulate Tracer - P. W. Nickola (Atmospheric Boundary Layer Studies)	53

Variation of Sample Wind Component Spectra and Implications for Diffusion Modeling in the Atmospheric Boundary Layer - D. C. Powell (Atmospheric Boundary Layer Studies)	56
Further Studies on a Low Level Model to Compute Montgomery Stream Functions - W. E. Davis (Atmospheric Boundary Layer Studies)	61
<u>DEPOSITION AND RESUSPENSION</u>	
Modeling Preliminaries for Dry Deposition to a Canopy - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences.	65
Rate Limiting Aspects of the Dry Deposition of Gases - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	69
Some Comments on the Surface Boundary Conditions for the Diffusion- Deposition-Resuspension Problem - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	72
Approximations and Solutions for the Deposition-Resuspension Problem - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	76
Resuspension from a Generalized Area Source - T. W. Horst (Particle Resuspension and Translocation)	79
Comparison of Deposition Velocities Predicted from Dimensionless Analysis Correlations - G. A. Sehmel (Radioisotopes as Particles and Volatiles)	83
Particle Deposition and Penetration Through Vegetation Canopies from Wind Tunnel Experiments - G. A. Sehmel and W. H. Hodgson - (Radioisotopes as Particles and Volatiles)	86
Field Deposition Velocity Measurements to a Sagebrush Canopy - G. A. Sehmel and W. H. Hodgson (Radioisotopes as Particles and Volatiles)	89
Resuspension Rates from a Circular Field Source - G. A. Sehmel and F. D. Lloyd (Particle Resuspension and Translocation)	92
An Experiment to Measure Particle Resuspension from a Large Annular Area - G. A. Sehmel and F. D. Lloyd (Particle Resuspension and Translocation)	94
Particle Resuspension from Truck Traffic in a Cheat Grass Area - G. A. Sehmel (Particle Resuspension and Translocation)	96
The Influence of Soil Insertion on Atmospheric Particle Size Distributions - G. A. Sehmel (Particle Resuspension and Translocation)	99
Initial Observations of Airborne Concentrations of Small Particle Size Ranges During Two Dust Storms - G. A. Sehmel (Particle Resuspension and Translocation)	102
Frequency of Airborne Dust in the Continental United States - M. M. Orgill and G. A. Sehmel (Particle Resuspension and Translocation)	105
Wind Profiles in Dust Storms - M. M. Orgill, G. A. Sehmel, and T. J. Bander (Particle Resuspension and Translocation)	109

Frequency of Dust Devils in the Hanford Reservation - M. M. Orgill, J. M. Thorp and L. C. Schwendiman (Atlantic Richfield Hanford Company)	112
Resuspension of Radioactive Isotopes Induced by Burning Forest Logging Slash - G. A. Sehmel and M. M. Orgill (Particle Resuspension and Translocation)	115
Concentrations and Rates of Removal of Contaminants from the Atmosphere in and Downwind of St. Louis - J. A. Young, T. M. Tanner, C. W. Thomas, N. A. Wogman and M. R. Peterson (Tracer Studies in the METROMEX Experiment)	119
Dry Deposition of Sulfur Dioxide and Sulfur Particles from Northwest Fossil Fuel Plants - R. K. Hadlock, J. G. Droppo and R. K. Woodruff (Atmospheric Boundary Layer Studies)	125
<u>POLLUTANT SCAVENGING</u>	
Comments on Diffusive Deposition at Small Fourier Numbers - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	127
Precipitation Scavenging of Monodisperse Aerosol Particles - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	129
A General Formalism for the Precipitation Scavenging Rates for Aerosol Particles - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	131
Approximations for the Precipitation Scavenging Rates for Polydisperse, Active, Aerosolized Pollutants - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	133
Rain Scavenging of Active Aerosol Particles from a Plume - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	136
Washout Ratios for Particulate Pollution with an Application to Convective Storms - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	139
Synoptic Scale Wet and Dry Deposition - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	142
The Battelle-Northwest In-Cloud Scavenging Program--An Overview - J. M. Hales (Precipitation Scavenging)	145
Precipitation Scavenging Field Experiments in March 1975 on the Olympic Peninsula - C. E. Hane and J. A. Young (Precipitation Scavenging)	147
Design for In-Cloud Scavenging Experiments on the Olympic Peninsula - C. E. Hane and J. A. Young (Precipitation Scavenging)	151
A Review of Moist Mesoscale Models with Possible Applicability to Precipitation Scavenging - C. E. Hane (Precipitation Scavenging)	156
An Estimate of In-Cloud Scavenging of a Tracer Injected into Two Frontal Storms - W. E. Davis (Precipitation Scavenging)	160
Development of a DC-3 Aircraft for Atmospheric Pollution Research - J. M. Hales (Precipitation Scavenging)	166
A Computer Algorithm for Reduction and Presentation of Information from the BNW DC-3 Data Acquisition System - D. R. Drewes (Precipitation Scavenging)	169

Scavenging of Urban Pollutants by Thunderstorm Rainfall: Numerical Experimentation - C. E. Hane (Precipitation Scavenging)	172
The Entrainment of Tracers Near the Sides of Convective Clouds - J. A. Young, T. M. Tanner, C. W. Thomas and N. A. Wogman (Tracer Studies in the METROMEX Experiment)	179
A Design for Regional Precipitation Chemistry Measurement and Analysis - M. Terry Dana and J. M. Hales (Precipitation Scavenging in MAP3S)	185
Development of a No-Deposition Precipitation Sampler - D. W. Glover (Precipitation Scavenging in MAP3S)	187
<u>POLLUTANT TRANSFORMATIONS AND INTERACTIONS</u>	
Aerosol Balance Equations; Theoretical Studies - R. L. Drake (Aerosol and Trace Gas Transformations)	189
Aerosol Balance Equations; Numerical Modeling - R. L. Drake (Aerosol and Trace Gas Transformations)	191
The Growth of Aerosol in an Urban Plume - A. J. Alkezweeny (Aerosol and Trace Gas Transformations)	196
Simultaneous Aerosol Size Distributions and Turbidity Measurements over a Metropolitan Area - A. J. Alkezweeny and N. S. Laulainen (Aerosol and Trace Gas Transformations)	202
Turbidity Variations over the Alton, Illinois, Airport During August 1975 - N. S. Laulainen (Aerosol and Trace Gas Transformations)	206
Aerosols and SO ₂ Measurements During the Da Vinci Balloon Flight - A. J. Alkezweeny (Aerosol and Trace Gas Transformations)	208
<u>INTEGRATION OF MODELS FOR ATMOSPHERIC PROCESSES</u>	
An Analysis of Tritium in the Atmosphere Released by a CTR - D. S. Renné, W. F. Sandusky and M. T. Dana (Safety Analysis and Environmental Effects Studies)	211
A Regional Scale Model for Computing Deposition and Ground Level Air Concentration of SO ₂ and SO ₄ from an Elevated Source - L. L. Wendell, D. C. Powell and R. L. Drake (MAP3S Modeling Studies)	218
An Examination of the Effects of Real Time Versus Time-Averaged Precipitation Data on SO ₂ and SO ₄ Removal in the Regional Assessment Model - L. L. Wendell and D. C. Powell (MAP3S Modeling Studies)	224
Preparation of Hourly Maps of Gridded Precipitation Data for Use in Precipitation Scavenging Calculations - L. L. Wendell and C. E. Hane (MAP3S Modeling Studies)	229
Examination of the Wind Shear in Regional Scale Flow Layers Derived from Radiosonde Data - L. L. Wendell and T. D. Fox (MAP3S Modeling Studies)	232
Chemical Deposition and Transformation Parameterizations for Regional Models - R. L. Drake (MAP3S Modeling Studies)	237

Regional Air Quality Assessment for Northwest Energy Development Scenarios - D. S. Renne and D. L. Elliott (Pacific Northwest Energy Related Regional Assessment Program)	243
An Investigation of Regional Flow Patterns over an Area of Complex Terrain - D. L. Elliott, W. F. Sandusky and R. K. Woodruff (Investigation and Modeling of the Meteorology of the Hanford Region)	246
<u>WEATHER AND CLIMATE MODIFICATION</u>	
Climate and Weather: Natural and Anthropogenic Perturbations - R. L. Drake (ERDA/RL Special Studies)	253
A Modified Set of Maxwell's Equations that Possess Galilean Invariance - W. G. N. Slinn (Theoretical Studies in Atmospheric Sciences)	258
Calibration of Sunphotometers - N. S. Laulainen and E. W. Kleckner (Aerosol and Trace Gas Transformations)	261
Atmospheric Turbidity over Hanford During 1975 - N. S. Laulainen (Aerosol and Trace Gas Transformation)	265
Comments on the Determination of Aerosol Optical Properties - N. S. Laulainen (Aerosol and Trace Gas Transformations)	268
Plume Dimensioning by Long Exposure Photography - J. M. Thorp (Cooling Tower and Cooling Pond--Atmospheric Impact)	272
Natural Draft Cooling Tower Plume Characteristics Determined with Airborne Instrumentation - M. A. Wolf (Cooling Tower and Cooling Pond--Atmospheric Impact)	281
PUBLICATIONS AND PRESENTATIONS	289
ORGANIZATION CHARTS	293

ATMOSPHERIC SURVEYS

Surveys are being made of effluents released to the atmosphere, within the source areas, to distances covering several states, and on hemispheric scales. Through these efforts, the Atmospheric Sciences and Radiological Sciences Departments are identifying the most urgent air pollution problems and continually reassessing them to set priorities for more detailed investigations.

Fallout measurements and evaluations are continuing with the essential identification and study of world-wide nuclear test debris, nuclear power effluents appearing in the atmosphere, and the transfer of these world-wide distributions to the surface. New survey efforts are measuring ambient distributions of pollutants from fossil fuel facilities. Measurements are being made describing the wide variety of pollutants introduced into the atmosphere from individual coal facility sources; measurements of pollutant characteristics from urban complexes are defining their physical and chemical characteristics as they are transported large distances. Reference levels of pollutants in air entering a given region are also being defined. The emphasis in ongoing reference measurements on the coast of Washington State will assure future comprehensive coverage of nonnuclear pollutants in addition to the continuing nuclear contaminant evaluations.

- **SURVEY OF AMBIENT AIRBORNE POLLUTANTS**
- **RADIOACTIVE FALLOUT RATES AND MECHANISMS**
- **COAL CONVERSION ATMOSPHERIC CHEMISTRY**
- **TRACER STUDIES IN THE METROMEX EXPERIMENT**

OZONE, RADIONUCLIDE AND PARTICULATE CORRELATIONS
IN AIR AT QUILLAYUTE, WASHINGTON

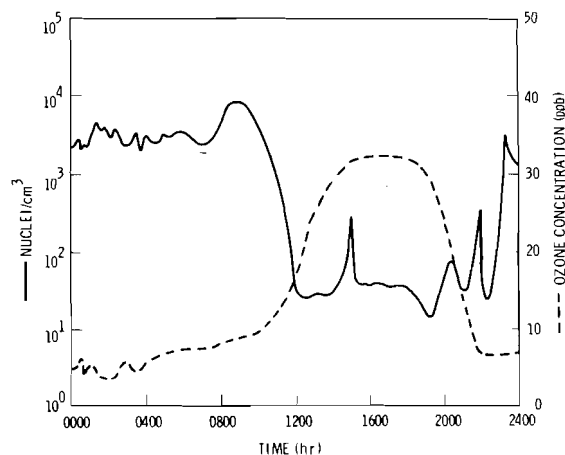
J. D. Ludwick and T. D. Fox

Several results are presented from comparative analysis of atmospheric pollutants measured in 1975 at the Quillayute Air-Reference Station located in northwestern Washington. A description of the site and sampling techniques used were presented¹ in the 1974 Annual Report. Included here are graphic monthly computer readouts of physical and chemical information accumulated since then.

OZONE

Meaningful positive or negative correlations between many atmospheric materials are often masked by physical and chemical processes affecting the species of concern. For example, the influence of pollutant materials on the ozone concentration at low level is illustrated in Figure 1. This is a typical situation at Quillayute; high particulate concentrations occur some 20% of the time due to the presence of moderate atmospheric stability. The resultant stagnation drastically increases the concentration of particulates near ground level. Gaseous pollutants are also trapped under the inversion layer. The data indicate that when this occurs, the ozone is rapidly depleted through reaction with these pollutants. The lack of a significant low level ozone source may also be postulated from these results.

Significant depletion of ozone was observed within 5-20 min of sunrise, indicating a high reaction rate, probably of first order kinetics, not involving a third body. The rapid ozone depletion under stable conditions also suggests that ground inter-

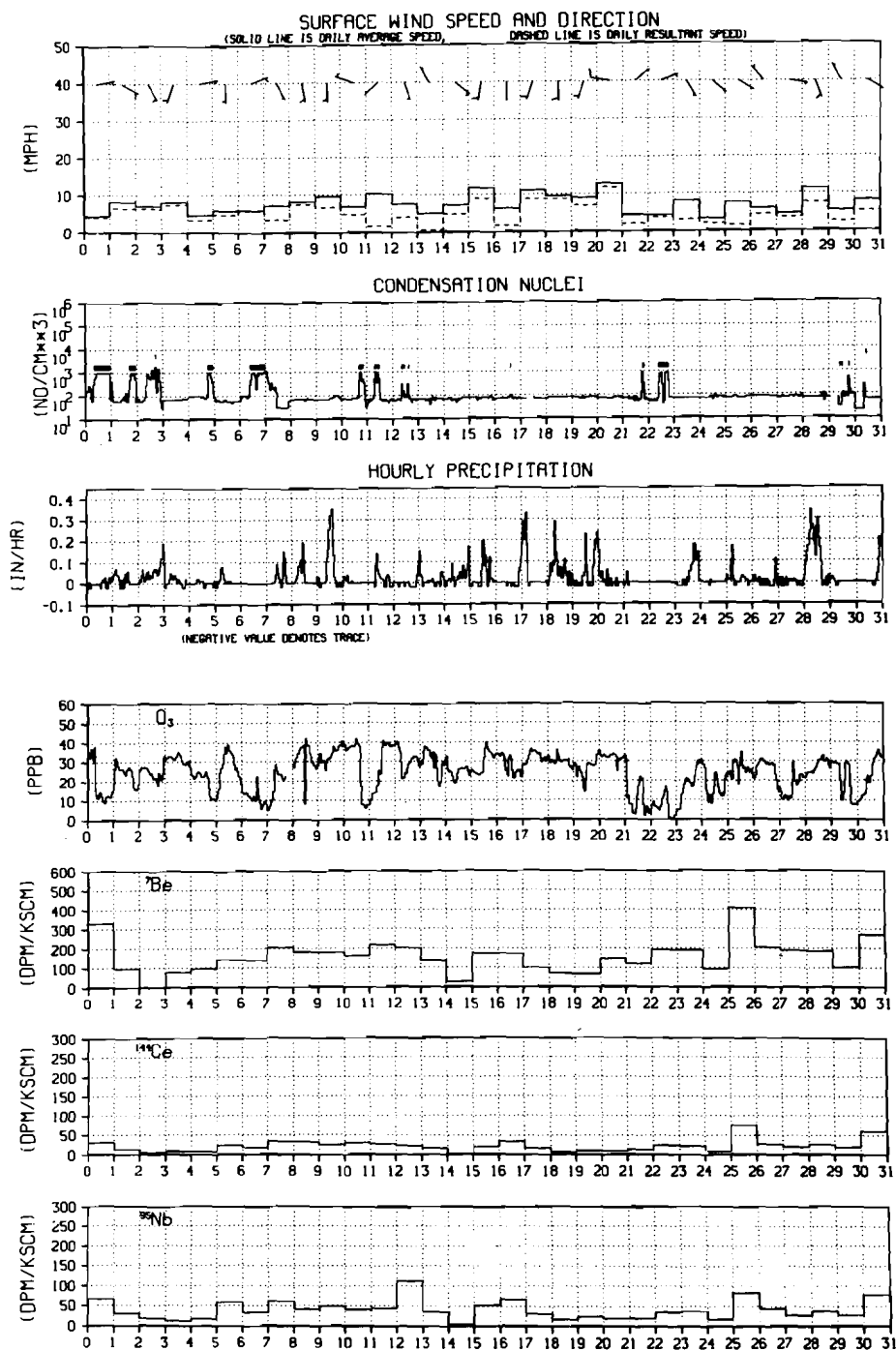


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FIGURE 1. Typical Diurnal Variation of Condensation Nuclei and Ozone Concentrations in Response to Atmospheric Stability Changes Near Ground Level

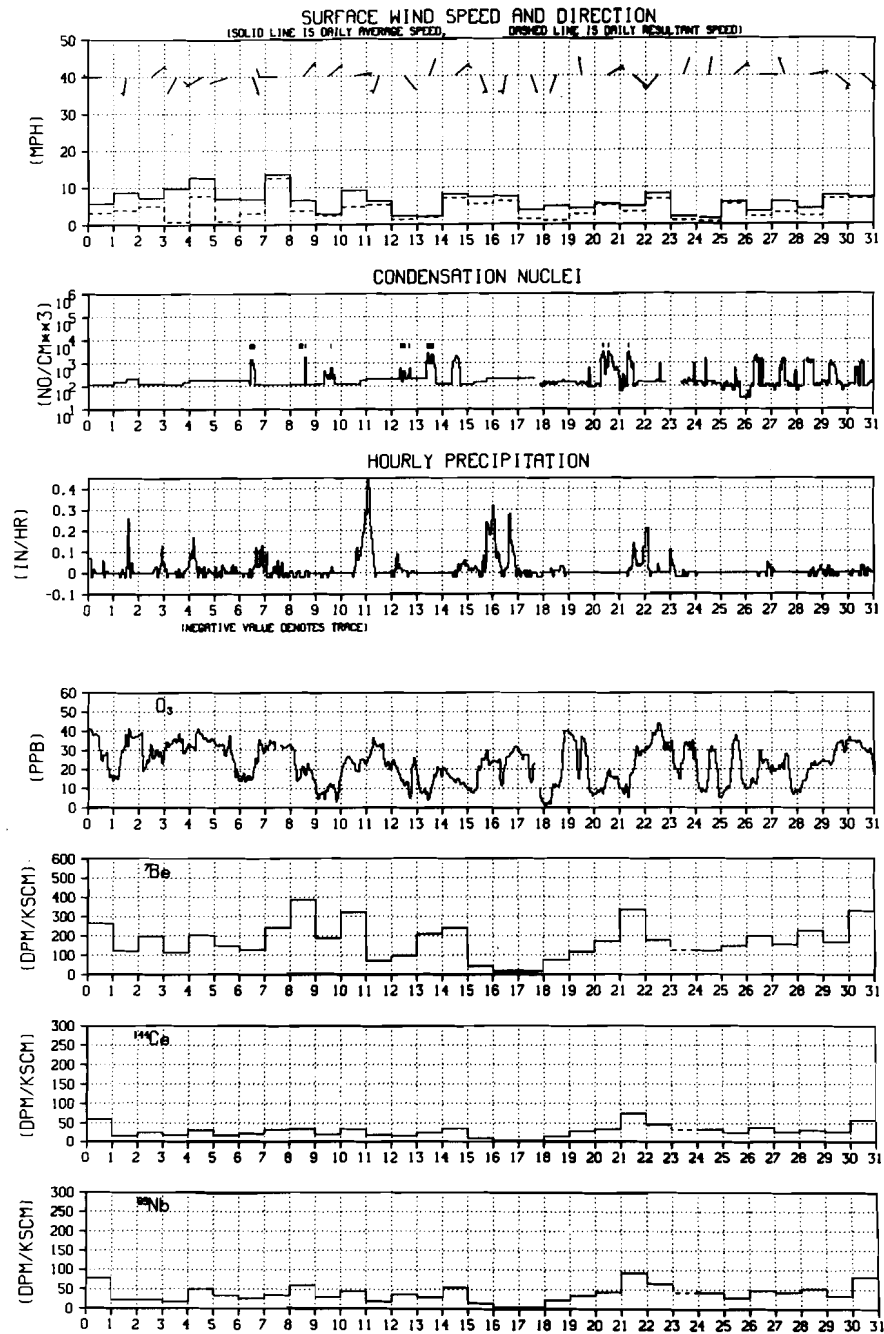
action is not the prime factor, since the sampling port is located 33 m high.

The diurnal effect promoting anti-correlation in the ozone particulate data was not the only one observed



Neg 753676-10

FIGURE 4. Interrelationships of Chemical and Physical Information, Quillayute, Washington, December 1974



Neg 753676-12

FIGURE 5. Interrelationships of Chemical and Physical Information, Quillayute, Washington, January 1975

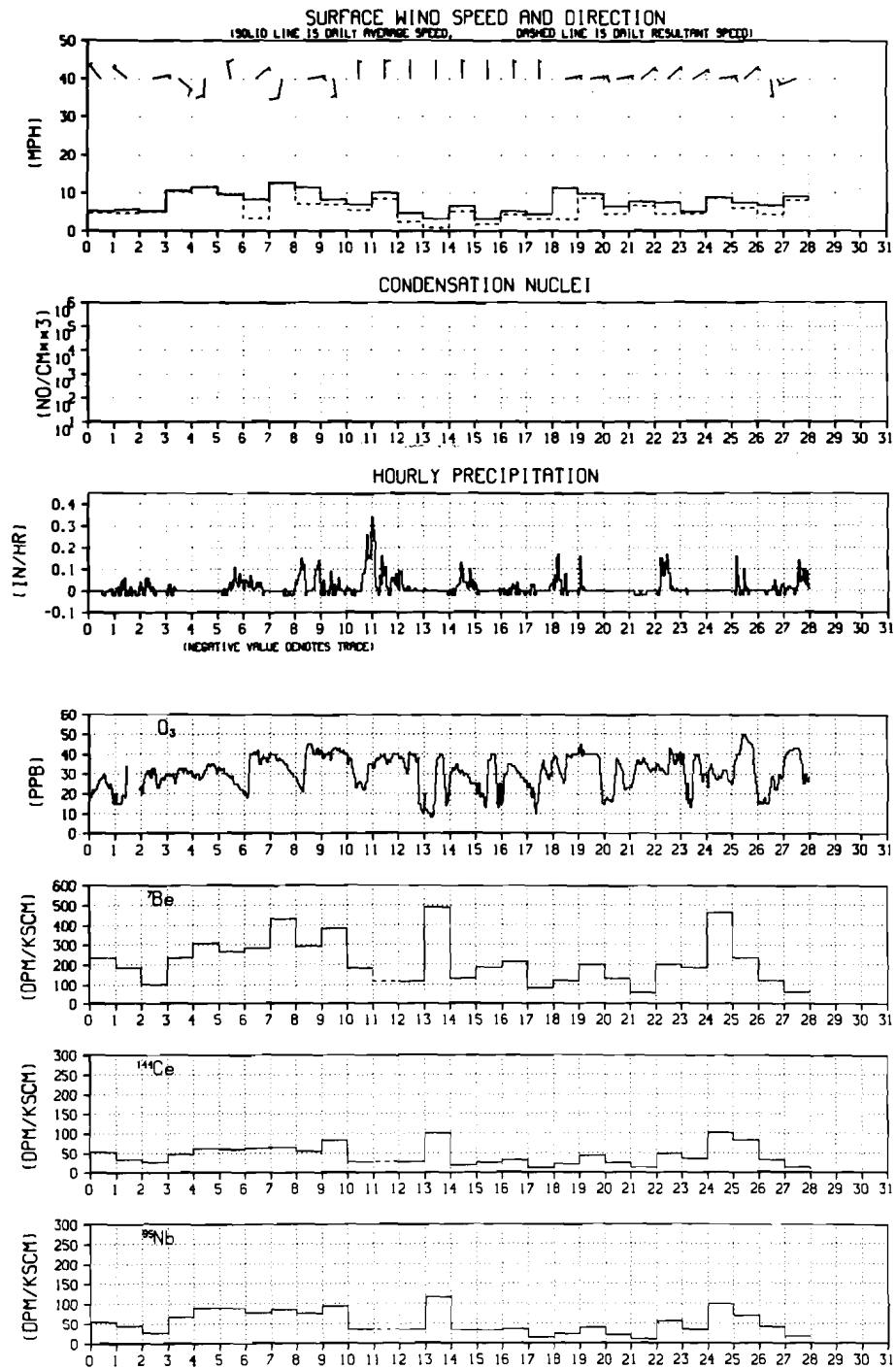


FIGURE 6. Interrelationships of Chemical and Physical Information, Quillayute, Washington, February 1975

Specific cases of such particulate scavenging are difficult to identify with complete confidence; however, the data most certainly suggest that this effect is prevalent. Often, the level of particulates in incoming fresh air is low and the effect of precipitation scavenging is difficult to identify.

In summary, the major chemical and physical factors affecting the ozone and particulates tend to decorrelate their relationships. It is then particularly significant that such cor-

relations exist in the Quillayute data.

RADIONUCLIDE CORRELATIONS

This inference was pointed out in data presented previously.¹ To better illustrate this fact, the ozone data were replotted in an identical form to the radionuclide information. The daily maximum observed ozone concentrations were used as most representative of the tropospheric values in the vicinity of our monitoring station. Figures 7 and 8 illustrate

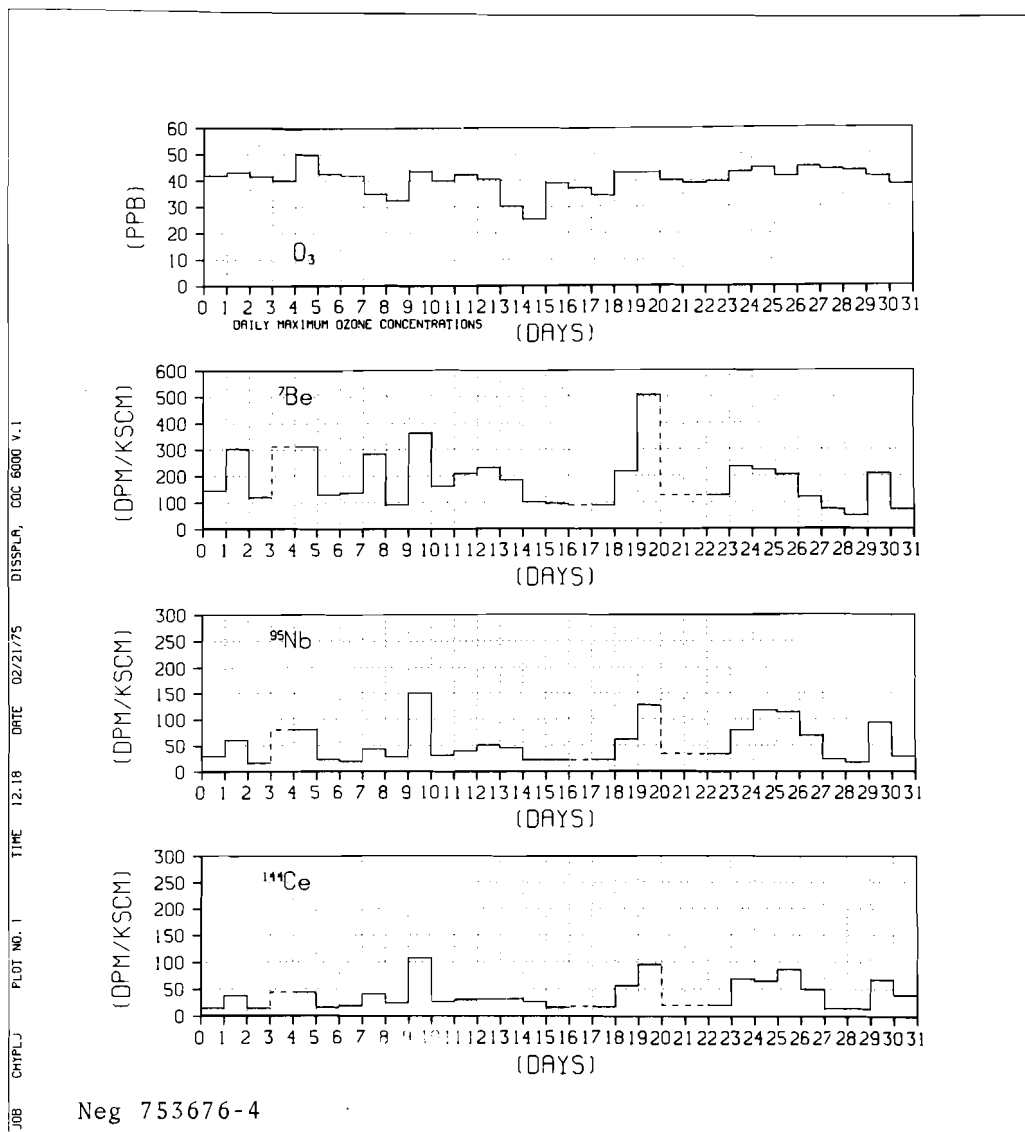


FIGURE 7. Comparison of Daily Maximum Ozone with Daily Average Radionuclide Concentrations at Quillayute, Washington, March 1974

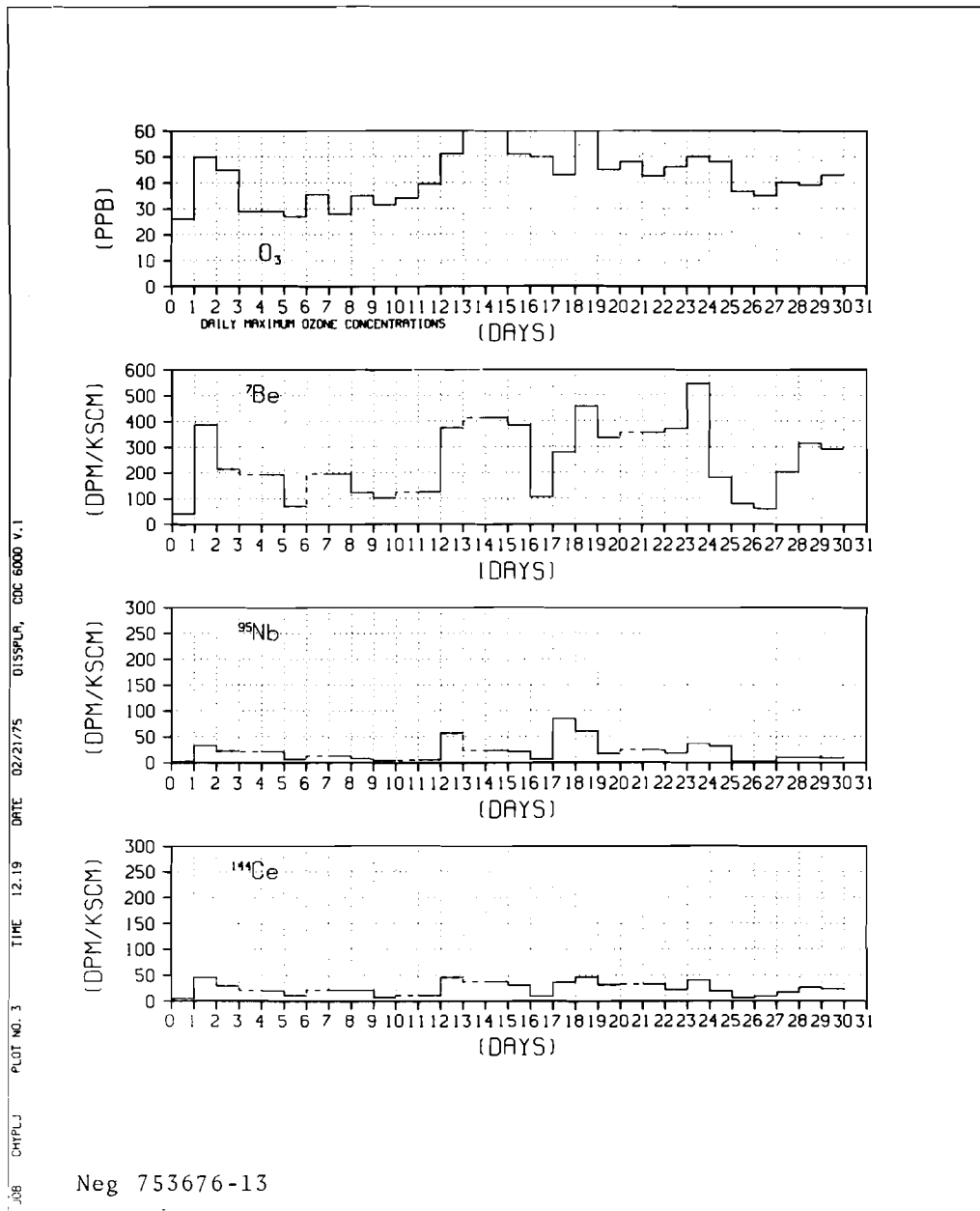


FIGURE 8. Comparison of Daily Maximum Ozone with Daily Average Radionuclide Concentrations at Quillayute, Washington, September 1974

the positive correlation between ozone, beryllium, and fallout radionuclides. During a good deal of September, the meteorological situation was quite well defined. An upper level high prevailed over the region. This was conducive to the transport of high altitude materials

downward. Late night and early morning inversions at Quillayute developed under this high pressure, resulting in depletion of the ozone. However, as daily thermal activity penetrated the stable layer by early afternoon, very large increases in ozone were observed. Accompanying this ozone was a large

shown in Figure 9. Springtime increases of these materials in the troposphere in the northern latitudes, are to be expected, from previous work on fallout patterns.² High values of ozone and Be during the fall are rather unique, perhaps due to the persistent high pressure area previously mentioned.

The annual fluctuations of materials whose major source region is suspected to be stratospheric is

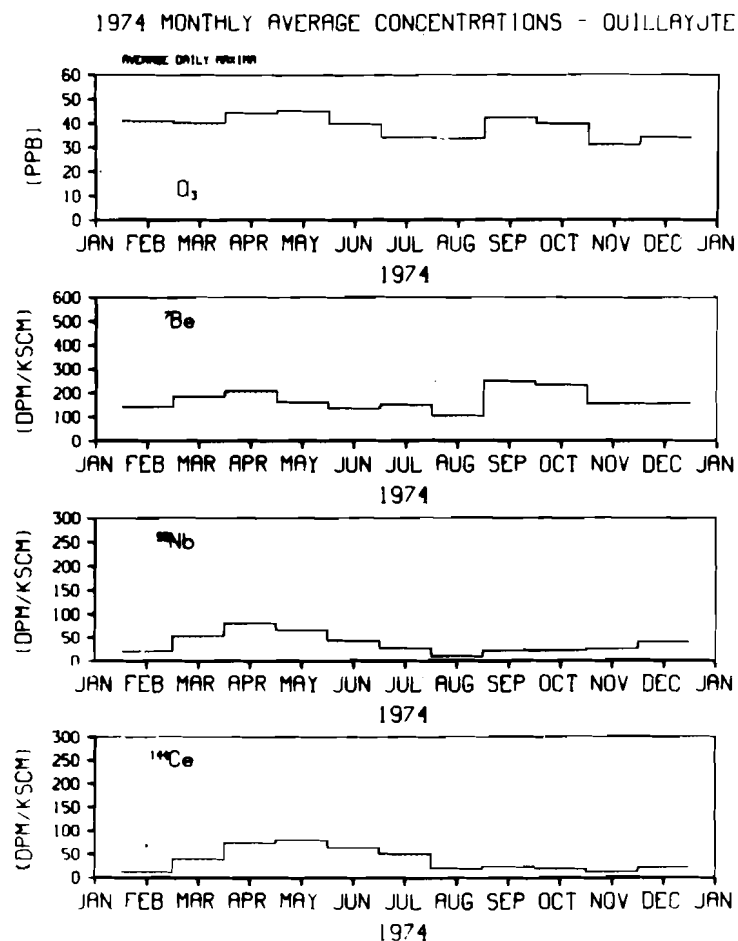


FIGURE 9. 1974 Monthly Averages of Radionuclides and Daily Maximum Ozone Concentrations at Quillayute, Washington

Many recent studies have indicated that rural ozone concentrations are as high or higher than urban concentrations. Frequently they exceed the national air quality standards of 80 ppb.^{3,4,5} Daily maximum ozone statistics from our Quillayute, Washington air monitoring station¹ are compared with similar data from sites in New York and New Jersey in Table 1.⁴

Although the median Quillayute ozone concentration is comparable to the lower median New York concentrations, the upper quartile and upper decile Quillayute values are far below those of the eastern sites. Furthermore, although urban and rural eastern sites occasionally exceeded the air quality standard of 80 ppb, the maximum Quillayute ozone concen-

tration reached between April and November 1974 was 60 ppb.

CONCLUSIONS

The day-to-day changes in daily maximum ozone concentrations at Quillayute have been related to the transport of stratospheric materials.¹ Such fluctuations apparently cannot account for occasionally much higher values reported in more polluted regions by Bruntz and others.⁴ Thus, the Quillayute data indicate that stratospheric-source ozone is probably responsible for only a portion of surface ozone concentrations which exceed the National Ambient Air Quality Standard of 80 ppb. The additional ozone may well be derived from low-level pollution sources as many investigators have long suggested.

TABLE 1. Quantiles of the Daily Maximum O₃ Concentration

Site	Measurement Site	Median (ppb)	Upper Quartile (ppb)	Upper Decile (ppb)	Percentage of days exceeding standard (80)
New Jersey ^{(4)*}	Ancora	74	100	125	44
	Bayonne	64	96	121	38
	Camden	66	88	106	31
	Linden	54	78	104	22
	Newark	46	75	108	21
New York ^{(4)*}	Eisenhower Park	33	60	85	13
	Mamaroneck	45	68	101	20
	Welfare Island	42	78	116	22
	Yonkers	70	106	135	39
Washington ^{(1)**}	Quillayute	39	45	50	0

a. Based on period between May 1, 1973 and September 30, 1975.

b. Based on period between April 1, 1974 and November 30, 1974.

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LEAD (Pb) CONCENTRATIONS ASSOCIATED WITH 1000 mb GEOSTROPHIC BACK TRAJECTORIES AT QUILLAYUTE, WASHINGTON

T. D. Fox and J. D. Ludwick

Daily averaged Pb concentrations in the air at Quillayute, Washington, are related by an objective method to 1000 mb geostrophic backward one-day air trajectories. The results confirm the expectation that land areas are source regions for Pb. The highest Pb values are characteristic of anthropogenic sources within the influence of the air-monitoring station. Low oceanic trajectory Pb concentrations indicate levels to be expected in air over the ocean and away from major Pb sources.

INTRODUCTION

As more attention has been given to the problem of air pollution over populated areas of the United States and the rest of the world, quantitative observation of pollution levels in the air around metropolitan areas has become routine. To evaluate these observations with regard to possible control of suspected sources, background pollutant levels, i.e., the amount of any specific material which is in the air before it is affected by a source or source region,

must be considered. Environmental impact statements relating to the original air quality prior to projected development require such data.

This report is concerned specifically with daily stable lead (Pb) concentrations measured at an air monitoring station on the North American Pacific coast at Quillayute, Washington.¹ They are related with corresponding 1000 mb geostrophic back trajectories computed from Quillayute, to identify reference level Pb in unpolluted conditions.

Directions and/or regions are indicated from which air with generally high or generally low Pb concentrations may be expected.

COMPARISON WITH OTHER REGIONS

Table 1 summarizes the daily Pb concentration data for April through September 1974 at Quillayute. The data consist of samples obtained from high volume ($2.26 \text{ m}^3/\text{min}$), 1-day filters, analyzed by x-ray fluorescence techniques. Values were generally higher in August and especially in September, than in the preceding months when concentrations were typically less than 10 ng/m^3 .

As a basis for comparison, Table 2 presents statistics published by the Health and Safety Laboratory.² These show stable Pb concentrations analyzed from monthly composites of weekly samples from various sites. Monthly values from metropolitan areas range from 500 to 1400 ng/m^3 , while suburban and rural areas are lower. The values from Quillayute fall near the range of remote stations on Greenland and Antarctica, and the oceanic stations at Mauna Loa and Easter Island.

Because the Quillayute averages in Table 1 consider all meteorological situations during the 6-month span, they include and are influenced by situations in which local or regional contamination could be expected. A meteorological analysis including geostrophic back trajectory calculations was undertaken to define average concentrations during periods of local or regional contamination,

while characterizing these periods in terms of regional atmospheric circulation. In addition, meteorological analysis was used to distinguish Pb samples obtained during periods of strictly oceanic source air, whose concentrations might be considered "reference" levels.

DESCRIPTION OF PROCEDURE

To obtain a broad view of the relationship between Quillayute air trajectory histories and the daily average Pb concentrations measured there, a method of objective analysis was implemented. The results of this analysis are shown in Figure 1. This is not an analysis of Pb concentrations in the regions covered; rather, the concentration value assigned any given location on the map is an estimate of the concentration measured 1 day later at Quillayute. The location of this value is determined by back trajectory analyses.

DISCUSSION OF ANALYSIS

Ocean versus Land

Figure 1 indicates that, as might be expected, concentrations are generally high in air coming from continental areas, and low in air which traversed the Pacific Ocean. A region of particularly close packing of isopleths exists near the coast south of Quillayute. The minimum values observed from air originating over the ocean are between 1 and 2 ng/m^3 , while maximum concentrations greater than 32 ng/m^3 relate to air which had previously been southeast of the monitoring station.

TABLE 1. Quillayute, Washington 1974 Pb Concentration
Summary - by month (ng/cm^3)

<u>Month</u>	<u>April</u>	<u>May</u>	<u>June</u>	<u>July</u>	<u>August</u>	<u>September</u>	<u>All Months</u>
Number of observations	29	28	28	31	28	25	169
Mean	9.75	4.37	4.79	7.03	11.08	22.18	9.35
Median	3.25	2.83	3.36	4.06	5.47	21.70	3.82
Maximum	36.8	23.4	20.6	36.2	60.34	62.60	62.60
Minimum	.63	.58	.23	.84	.60	.72	.23

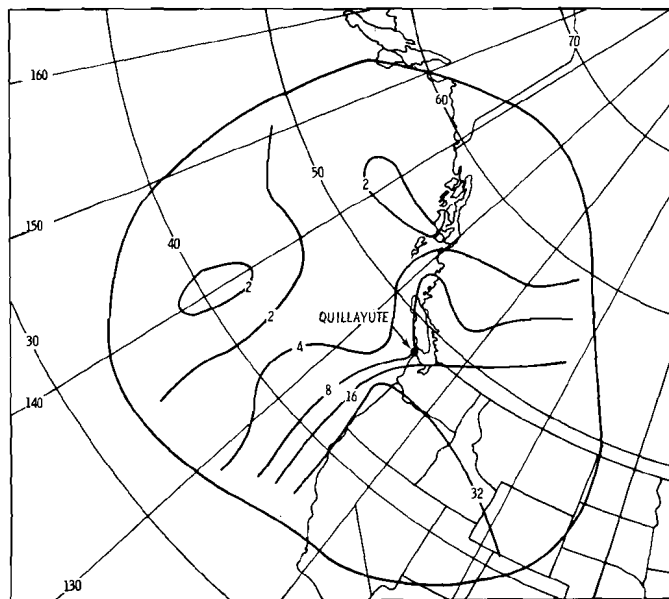
TABLE 2. Stable Lead Concentrations in Surface Air
During 1974 (ng/cm³)

Site	Apr.	May	June	July	Aug.	Sep.
Thule, Greenland	15.10	19.00	11.10	10.80	--	6.81
Kap Tobin, Greenland	3.58	3.18	2.25	1.86	1.85	2.38
Moosonee, Ontario	21.20	21.70	26.40	12.10	13.50	13.70
Helena, Montana	267.00	229.00	381.00	311.00	282.00	613.00
New York, New York	935.00	899.00	1190.00	780.00	1300.00	1370.00
Salt Lake City, Utah	617.00	799.00	785.00	766.00	1120.00	1230.00
Rocky Flats, Colorado #1	135.00	108.00	126.00	138.00	194.00	155.00
Rocky Flats, Colorado #2	633.00	464.00	604.00	603.00	850.00	781.00
Richmond, California	408.00	212.00	522.00	219.00	262.00	900.00
Miami, Florida	822.00	1070.00	1780.00	1450.00	1300.00	1400.00
Mauna Loa, Hawaii	4.14	2.62	3.22	8.35	10.10	4.44
Balboa, Panama	--	219.00	282.00	177.00	215.00	302.00
Guayaquil, Ecuador	298.00	349.00	146.00	346.00	305.00	276.00
Lima, Peru	514.00	491.00	505.00	70.90A	345.00	305.00
Chacaltaya, Bolivia	27.80	30.10	17.90	40.70	30.40	76.20
Antofagasta, Chile	40.90	30.50	22.40	47.40	40.50	24.40
Isle de Pasqua-Easter Is	8.03	4.28	6.64	3.18	4.04	2.79
Santiago, Chile	612.00	664.00	946.00	837.00	1010.00	454.00
Puerto Montt, Chile	11.60	9.43	12.20	12.40	14.80	10.70
Punta Arenas, Chile	51.20	66.40	79.90	87.30	67.30	55.10
Antarctica	3.85	2.95	--	--	--	--
South Pole Station	2.84	7.07	1.44	7.65	3.06	4.59

-- No data

Errors are less than 20% except:

A - error between 20% and 100%



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FIGURE 1. Quillayute Pb Concentrations, Contoured with Respect to the Position of the Air 1 Day Before its Arrival at Quillayute. Air histories based on 1000 mb geostrophic trajectory analyses.

These maximum concentrations are much lower than the typical average concentrations for the general North American urban and suburban areas listed in Table 2. This indicates that considerable dilution and/or Pb removal probably takes place as the air moves over land from the more metropolitan source regions of the Pacific Northwest to and through the relatively source-free area surrounding Quillayute.

Continental Differences

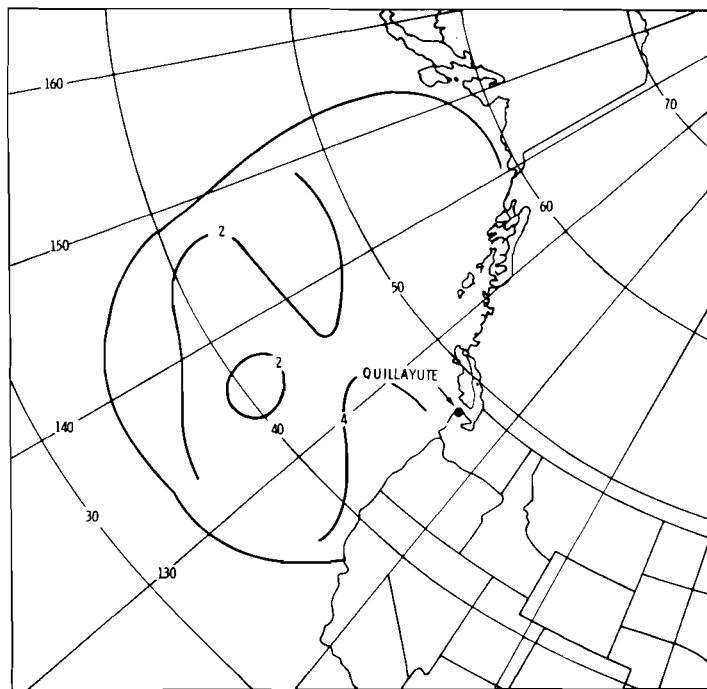
A definite north-south relationship can be seen in Figure 1 with specific respect to continental areas. The highest Pb concentrations arrive in air from southeast of Quillayute, while quite low values are displayed for air derived from western Canada.

More detailed examination of trajectory analyses, along with consideration of topography and typical regional wind flow patterns³ helps to explain this. When 1000 mb geo-

strophic analysis indicates flow from continental areas to the north, the air actually comes from the western, unpolluted side of Vancouver Island. However, during periods of southerly or southeasterly geostrophic flow, Quillayute may be in the path of air from southwest Washington or from the Willamette Valley metropolitan regions, as well as possibly from Puget Sound.

Oceanic Air

In order to determine representative Quillayute Pb concentrations in strictly oceanic air, an attempt was made to eliminate land-intercepting back trajectories. This was done by deleting from the analysis all days between April and September 1974 on which 3-day back trajectory analyses indicated that the air had at some time traversed land. The average Pb concentration for this differentiated data set of 50 trajectories was 2.65 ng/m^3 . A contour analysis of these data is shown in Figure 2.



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FIGURE 2. Quillayute Pb Concentrations in Strictly Oceanic Source Air Contoured with Respect to the Position of the Air 1 Day Before its Arrival at Quillayute. Air histories based on 1000 mb geostrophic trajectory analyses.

Comparison of the values listed in Figures 1 and 2 indicates substantial reduction of the pollutant concentration in air whose recent trajectory history does not intersect land. Low, yet significant pollutant values were derived for some just offshore trajectories to the south of Quillayute, suggesting some slight remaining continental influence. These values may be due to associated stagnant air conditions, or to inherent errors in geostrophic estimation of wind direction.

SUMMARY OF RESULTS AND FUTURE RESEARCH

Baseline Pb concentration values averaging less than 3 ng/m³ were

determined for air entering North America from the Pacific Ocean. Pb concentrations at Quillayute were strongly dependent on the direction from which the air came, with highest concentrations in air from metropolitan regions.

Currently, samples are being analyzed for a variety of other stable elements. These will be studied in the same manner as was the Pb, to obtain corresponding baseline concentration values for these other elements, and to compare directional differences.

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ATMOSPHERIC FALLOUT DURING 1975 AT RICHLAND, WASHINGTON
AND POINT BARROW, ALASKA

C. W. Thomas

The atmospheric radionuclide concentrations in surface air are monitored continuously at Richland, Washington and Point Barrow, Alaska. The radionuclides in the atmosphere at these locations in 1975 resulted primarily from a 2-3 MT Chinese test in June 1973 and a 1 MT Chinese test in June 1974. The concentrations in 1975 of radionuclides with half-lives greater than one year varied from equal to one-half the concentrations measured in 1974.

The atmospheric concentration of radionuclides in surface air is measured continuously at Richland, Washington and Point Barrow, Alaska by filtering large volumes of air through IPC filters, followed by gamma ray spectrometric analysis and chemical analysis for certain selected radionuclides. The spectrum of isotopes consists of natural radionuclides and their daughter products; from fission products, fissile material, activation products associated with nuclear weapons testing, and cosmogenic spallation reactions. The radionuclide concentrations from weapons testing measured in 1975 were attributed essentially to two nuclear tests by the Republic of China. In June 1973 and again in June 1974 the Chinese tested nuclear devices of 2 - 3 MT and 1 MT respectively, inserting relatively large amounts of radioactive debris into the stratosphere. Previous data have shown that radioactive debris inserted into the stratosphere has a residence half-time of about 11 months. During the early part of each year approximately half of the stratospheric inventory is released into the upper troposphere and will be measured in surface air throughout the year. Debris inserted into the stratosphere during one year will not be measured in surface air until the following year.

The fission product concentrations in surface air at Point Barrow and Richland during 1975 of radionuclides with half-lives greater than one year varied from equal to half the value measured in 1974, respectively (Figures 1 and 2). This is about what would be postulated, since the stratospheric inventory in early 1974 consisted of debris from the 3 MT device tested in June 1973 and about 0.8 MT of debris left over from the 1969 and 1970 nuclear tests. The total stratospheric burden was about 3.8 MT. The stratospheric inventory in early 1975 consisted of 1.9 MT residual from previous tests plus the 1.0 MT from the June 1974 test giving an inventory of 2.9 MT.

In the 1973 and 1974 nuclear tests significant amounts of activation products were created and inserted into the stratosphere. The concentration of ^{54}Mn , ^{57}Co , ^{58}Co , ^{60}Co , ^{65}Zn , and ^{88}Y in surface air at Richland and Point Barrow was increased in a manner similar to the fission products. The atmospheric concentrations of the cosmogenic radionuclides ^7Be and ^{22}Na have been measured since 1964. Sodium-22 was produced in significant quantities during the testing in 1961-1962. However, by 1967 the concentration in surface air was due entirely to spallation reactions, as was essentially all the ^7Be since 1964. Monthly averages of atmospheric ^7Be

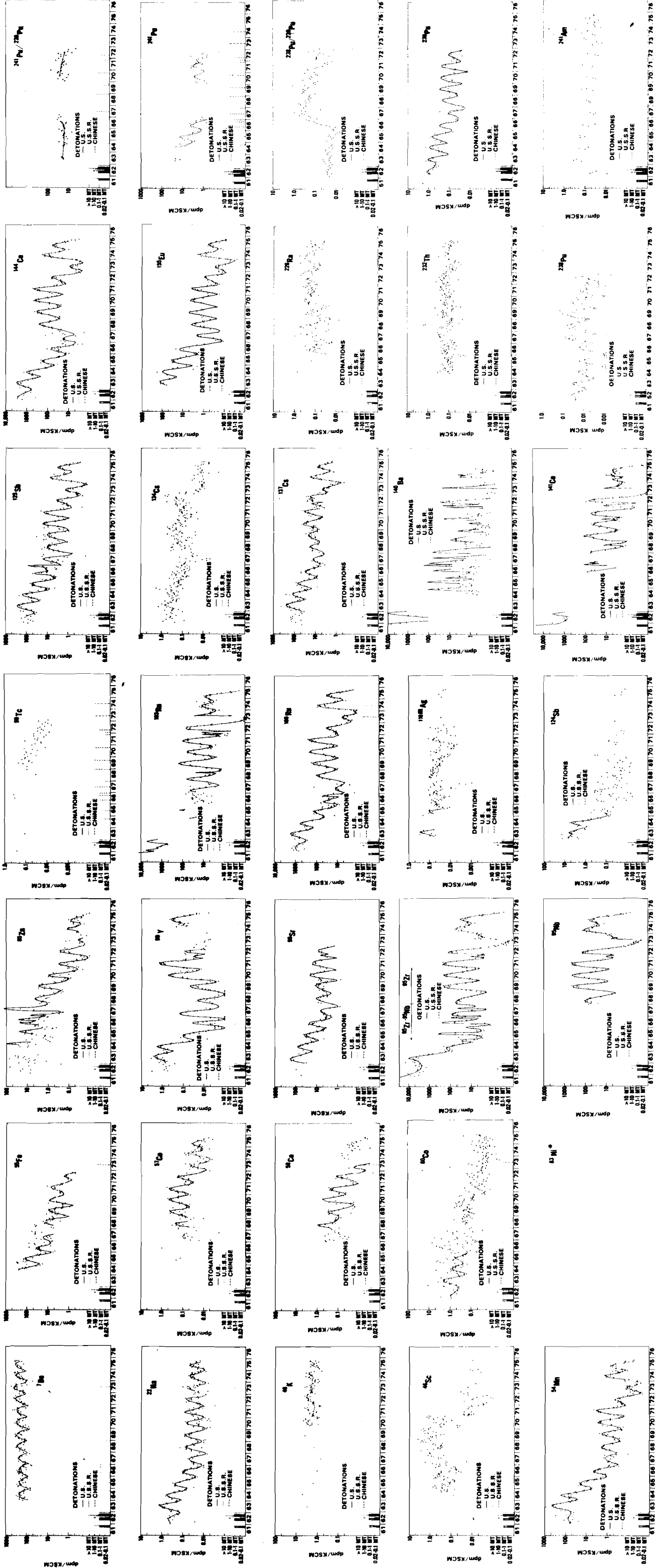
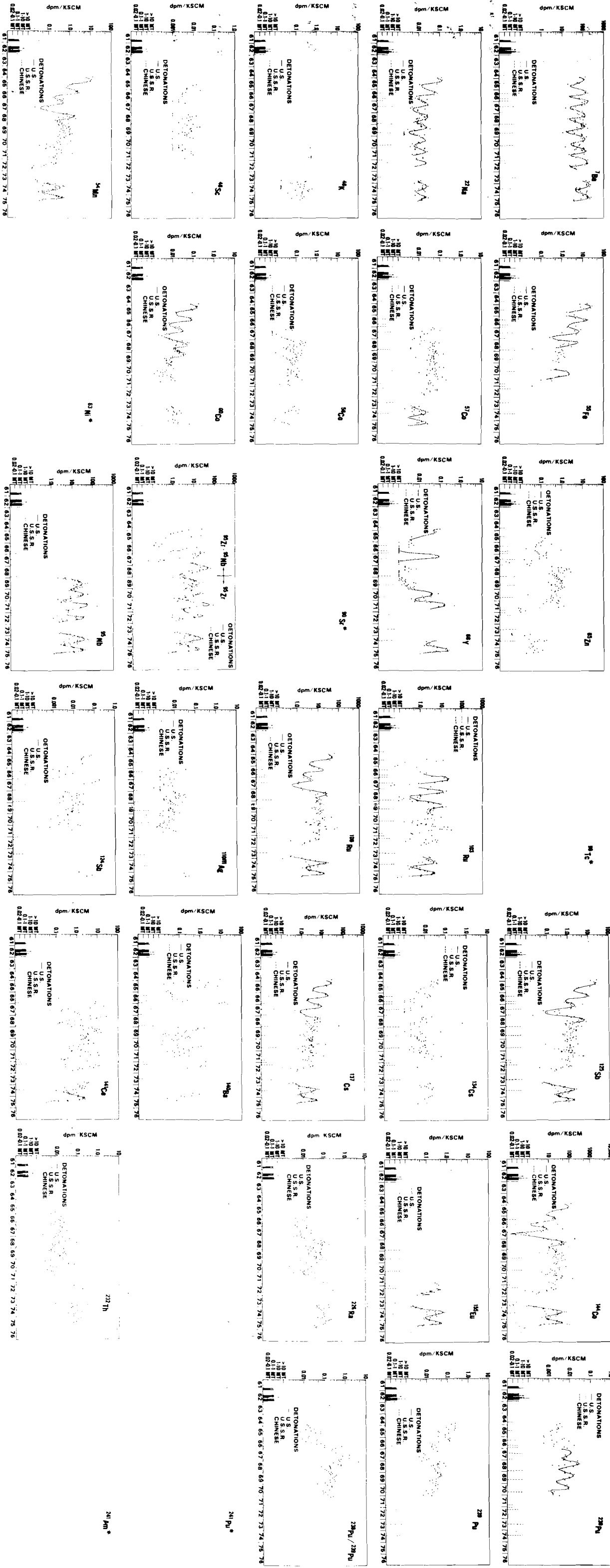


FIGURE 1. Radionuclide Concentration in Surface Air at Richland, Washington

NOTE: CONCENTRATIONS ARE IN DISENTEGRATIONS PER 1000 STANDARD CUBIC METERS (dpm/KSCM)

*ANALYSIS IS IN PROGRESS

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NOTE:
CONCENTRATIONS ARE IN DISINTEGRATIONS PER 1000 STANDARD CUBIC METERS (dpm/KSCM)
*ANALYSIS IS IN PROGRESS

FIGURE 2. Radionuclide Concentration in Surface Air at Point Barrow, Alaska

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from 1964-1975 and ^{22}Na from 1967-1975 are shown in Figure 3. The production rates of ^7Be and ^{22}Na are much higher in the lower stratosphere than near ground level; consequently, these isotopes show seasonal variations typical of radionuclides of stratospheric origin. Peak concentrations occur during May, June, and July

at Richland, while at Point Barrow the peak concentrations are during March and April. Average peak concentration at Point Barrow are always lower than those at Richland. This is possibly due to the transport of radionuclides from the stratosphere near the midlatitude in the region of the tropopause discontinuity.

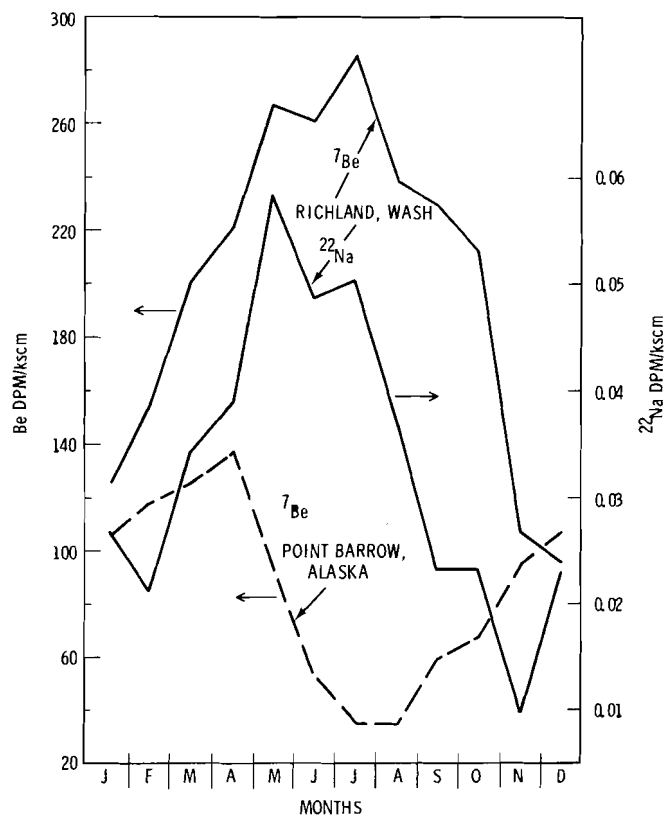


FIGURE 3. Monthly Averages of Atmospheric ^7Be from 1964 to 1975 and ^{22}Na from 1967 to 1975.

THE CONCENTRATIONS OF ^{241}Pu AND ^{99}Tc
IN SURFACE AIR AT RICHLAND, WASHINGTON

C. W. Thomas

Concentrations of ^{241}Pu and ^{99}Tc build up in nuclear wastes from power production reactors. Therefore, the concentrations of ^{241}Pu and ^{99}Tc were measured in air samples collected at Richland, Washington in previous years to provide a baseline against which future concentrations can be compared. Plutonium-241 is of interest because it is a precursor to ^{241}Am , which is as toxic via the human food chain as $^{239-240}\text{Pu}$. The reactions of Tc are similar to those of I in the human body; Tc, like I, is concentrated in the thyroid.

INTRODUCTION

Testing nuclear devices produces many associated radionuclides. Previous studies have measured these short and long-lived radionuclides in surface air, and their impact on the environment has been determined. Periodically, additional isotopes become of interest and they are measured along with these radionuclides.

Recently ^{241}Pu and ^{99}Tc have received considerable attention, mainly because they build up in nuclear wastes from power production reactors. Their concentration in surface air was measured so that a base line concentration in the environment can be determined and compared with future concentration measurements.

PLUTONIUM

During high yield nuclear weapons testing, considerable multiple neutron capture by ^{238}U results in the production of neutron-rich products; these beta decay and produce nuclides along the line of greatest stability. One of the prominent nuclides produced during this process is beta-active ^{241}Pu ($t_{1/2} = 15$ years). The activity associated with ^{241}Pu is many times greater than that of alpha-emitting $^{239-240}\text{Pu}$ but since it is a beta-emitter and has a much shorter half-life the radiotoxicity is much

lower. However, the fact that ^{241}Pu is the precursor of alpha-emitting ^{241}Am ($t_{1/2} = 433$ years), which is as toxic via the human food chain as $^{239-240}\text{Pu}$, the concentration of ^{241}Pu in the environs has become of considerable interest.

It has been estimated that 9450 kCi of ^{241}Pu have been injected into the atmosphere from all nuclear tests, and upon complete radioactive decay will represent 327 kCi of ^{241}Am . The concentration in surface air was determined by measuring the ^{241}Am ingrowth from the radioactive decay of ^{241}Pu on electrodeposited plutonium discs which had been separated several years previously. The ^{241}Am measurements were corrected for the contribution from ^{238}Pu (^{238}Pu has an alpha energy nearly identical to that of ^{241}Am); this had been measured on the discs several years ago. The small correction from the decay products of ^{238}Pu was added previously to determine the radiochemical yield.

The ^{238}Pu interference was small when the only contribution was from weapons-grade plutonium, but after the arrival of debris from SNAP-9A burn-up* the interference was too large to permit accurate measurement

* SNAP-9A was navigational satellite containing 17 kCi of ^{238}Pu .

of the ^{241}Am ingrowth in samples collected from 1967-1969. The concentrations of ^{241}Pu in surface air varied from a high of 20 dpm/KSCM in the 1963 peak maximum, to a low of <0.17 dpm/KSCM in late 1972. The seasonal variations were similar to those of other weapons-produced radionuclides of stratospheric origin. The disintegration rate ratio of ^{241}Pu - ^{239}Pu in surface air is shown in Figure 1. The average ratio was 19.2 with yearly averages varying from 15 to 24, compared with ratios of 27 (measured in debris from the Ivy Mike nuclear test of 1952) and 13 (measured in debris originating during the US-USSR nuclear test period of 1961-1962).

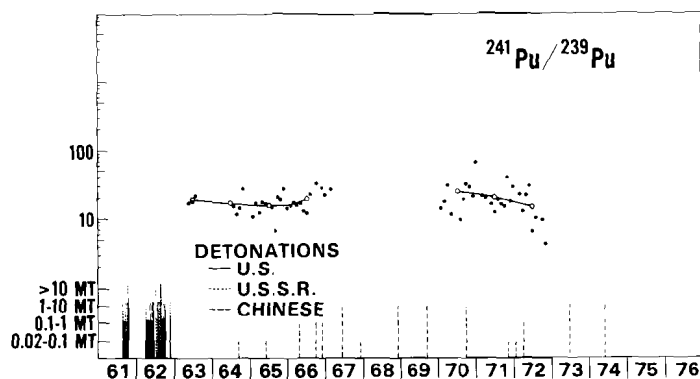
TECHNETIUM

Considerable interest has recently been given to the distribution and source of ^{99}Tc in environmental samples mainly because the reactions of Tc are similar to those of I in the human body (like I, Tc is concentrated in the thyroid). Chemically, Tc is similar to Re. Both form stable XO_4^- anions that give insoluble salts with large cations and both have volatile heptoxides and acid-insoluble heptsulfides. The most common compound is the pertechnetate and at low concentration salts of perhanate, perchlorate and periodate can be used as carriers. The fission yields for ^{99}Tc are slightly greater than 6% from both ^{239}Pu and ^{235}U fission, however, the specific activity is extremely low due to its half-life of about 210,000

years. If we assume ^{99}Tc and ^{137}Cs are present in air at ratios similar to their fission yields we would estimate the ^{137}Cs to ^{99}Tc disintegration rate ratio to be 7210. In nuclear wastes from power producing reactors ^{99}Tc will be the major radionuclide after a few hundred years decay. It has been estimated that by the year 2000 about 3×10^6 Ci of ^{99}Tc will be accumulated in nuclear wastes. Although ^{99}Tc is also formed during activating of molybdenum [$^{98}\text{Mo}(\text{n},\text{r})^{99}\text{Mo} \rightarrow \beta^- ^{99}\text{Tc}$] estimates indicate that in nuclear waste the production ratio by activation as compared to that of fission is very low (0.00028) and will be a minor source for ^{99}Tc production.

The concentration of ^{99}Tc in surface air was measured by chemically separating Tc from air filters and measuring ^{99}Tc disintegration rate on a low-background beta counter. The chemical procedure consisted of using Re as a carrier for Tc, fusing the air sample with a mixture of Na_2CO_3 Na_2O_2 , dissolving the fused residue with nitric acid, scavenging several times with basic ferric hydroxide, and precipitating with tetraphenyl arsonium chloride. The precipitate was washed and dried and the chemical yield determined by weighing the precipitate, after which the ^{99}Tc was measured using a low-background (0.8 cpm) beta proportional counter.

The ^{99}Tc concentration measured in air samples collected at Richland, Washington during the period 1969

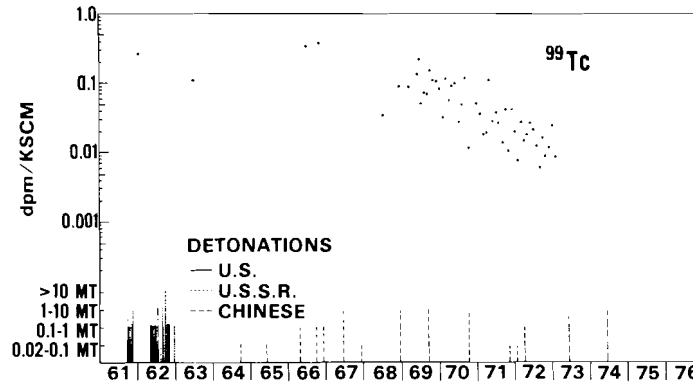


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FIGURE 1. Disintegration Rate Ratio of $^{241}\text{Pu}/^{239}\text{Pu}$ in Air at Richland, Washington

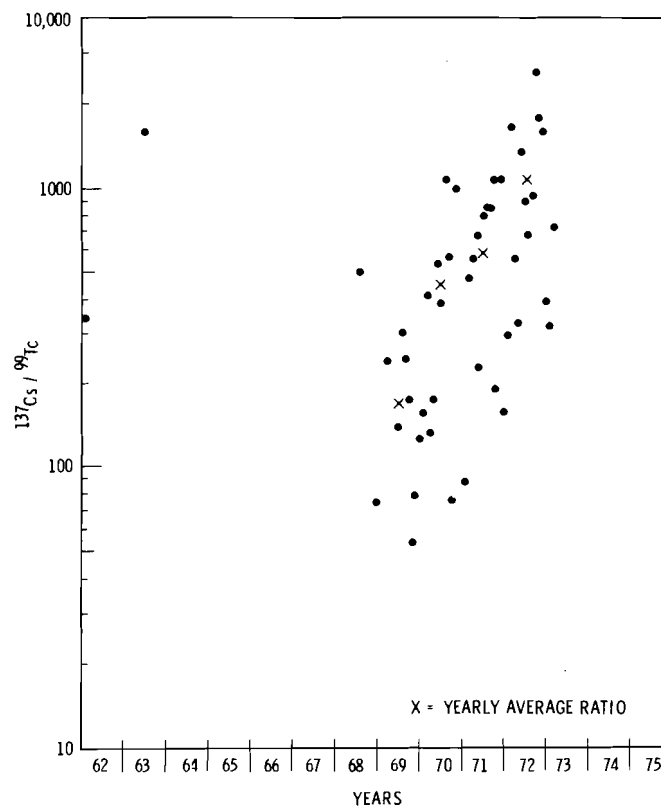
through 1972 varied from a high of 0.4 dpm/KSCM to a low of 0.007 dpm/KSCM, as shown in Figure 2. The disintegration rate ratios of ^{137}Cs to ^{99}Tc shown in Figure 3 varied from 53 to

2600 and can be compared with a ratio of ~ 7210 , an average value calculated by using the fission yields of ^{235}U and ^{239}Pu . The average yearly ratio from Figure 3 indicates the ratio has



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FIGURE 2. Radionuclide Concentration in Surface Air at Richland, Washington



Neg 761052-3

FIGURE 3. Disintegration Rate Ratios of ^{137}Cs to ^{99}Tc

increased significantly since 1969 rising from 167 in 1969 to 1025 in 1972. A ratio change as illustrated in Figure 3 would be possible since fractionation between the 137 and 99 chain occurs during detonation. The most probable Zp for fission fragments of the 137 chain is 24-sec ^{137}I and 252-sec ^{137}Xe giving rise to the high volatility for ^{137}Cs . This can be compared to the most probable Zp for the 99 chain of 33-sec ^{99}Zr and 150-sec ^{99}Nb giving rise to the high refractory characteristics of ^{99}Tc . Fractionation would enhance ^{99}Tc in tropospheric air giving rise to low

^{137}Cs to ^{99}Tc ratios. Unlike nuclear detonations, the fractionation characteristics of ^{137}Cs and ^{99}Tc associated with plants processing nuclear fuels are not based on precursors' characteristics but rather on the chemical characteristics of the isotopes themselves. Consequently in nuclear processing plants ^{137}Cs is not volatile while ^{99}Tc is considered quite volatile; around these plants one could expect possible enhancement of ^{99}Tc . Any contribution of ^{137}Cs and/or ^{99}Tc from nuclear fuels processing plants would enhance the ^{99}Tc in the tropospheric air.

SOLVENT REFINED COAL--ORGANIC CONSTITUENTS PRESENT IN THE PROCESS

M. R. Petersen

Process streams of a solvent-refining coal pilot plant have been sampled and the major organic constituents determined. About 50 organic compounds have been identified in the process water, the process solvent, process naphtha, the fuel product, and the mineral residue. Polycyclic hydrocarbons, phenols and quinolines were found to be main classes of compounds.

What are the potential emissions of any new process developed to obtain energy from fossil fuels? This question is particularly important for processes involving coal or any method to transform coal into a cleaner burning fuel. Since any constituents in the process streams are potential pollutants, knowing their identity and relative concentration before commercializing the process, provides a data base for proper emission control. During this last year we have sampled a pilot plant producing solvent-refined coal. Of the numerous organic constituents in the process streams we have identified about 50 major components.

FACILITY SAMPLING

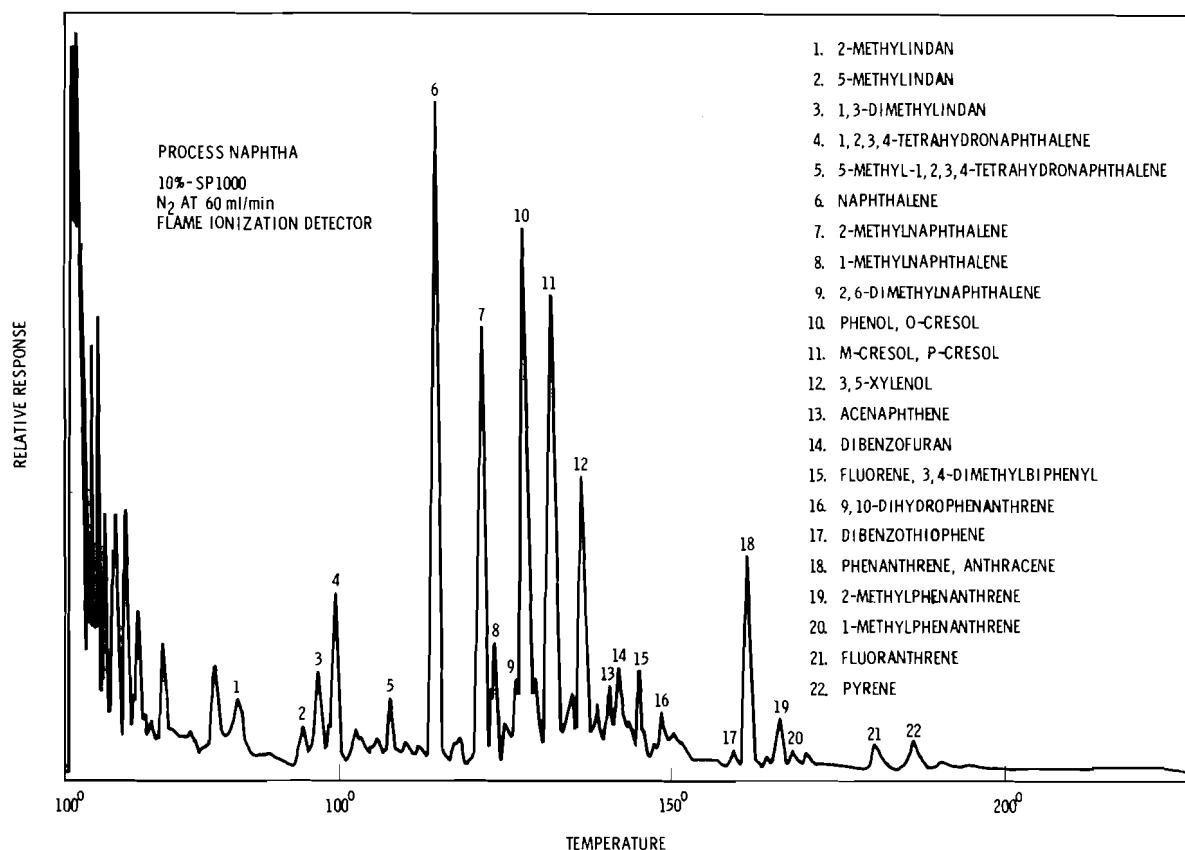
The 50 ton/day pilot plant located near Fort Lewis, Washington, and operated under ERDA contract by the Pittsburgh and Midway Coal Mining Company produces solvent-refined coal which meets design specifications for reduced emissions during firing. In the process, crushed coal is mixed with a recyclable coal-derived solvent and heated to about 825°F in presence of hydrogen at 1000 psi or greater. The coal swells and then dissolves. The solution is filtered to remove the inorganic residue and evaporated. The product which solidifies is a refined coal with less than 1/2% of either sulphur or residual ash.

The objective of our study is to collect samples at various processing points and determine the organic components which may be viewed as potential pollutants. The following samples have been collected: process solvent, mineral residue, process effluent water, process naphtha and the fuel product. The samples or extracts of the samples were characterized using gas chromatography with a flame ionization detector or a combination of gas chromatography-mass spectrometry. A typical chromatogram of the process naphtha is shown in Figure 1; it exhibits the complex mixtures of organic compounds present in the samples. To simplify the complexity of the mixture, the components in each bulk sample were separated into acidic, neutral and basic organic components. It was found that the neutral fraction (consisting mainly of aromatic hydrocarbons) predominates; the acidic fraction (consisting of

phenolic compounds) makes up less than 20% of total compounds; the basic fractions are minor components of less than 5%.

ANALYSIS

About 50 compounds have been identified in the samples collected thus far. Although the positional assignments on some of the methyl groups are tentative, the compounds listed in Figure 1 are the predominant neutral and acidic species common to all the samples. Quinolines are the predominant basic compounds; due to their low concentrations they cannot be seen in the chromatogram shown. Many of the compounds detected are toxic and only one (dibenzothiophene) has been reported as carcinogenic. Although presumably present, benzo(a)-pyrene has not been identified; if present, it appears to be a very minor component.



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FIGURE 1. Gas Chromatogram of Process Naphtha Arising in the Solvent Refining of Coal

Additional sampling is planned at the pilot plant to get offgas samples, particulate samples from product cooling areas and process water samples after biological treatment. Addi-

tional data are being collected on the trace elements including S, Se, Cd, Zn, Ni, Hg, As and Sb, which may be present in the samples.

BASELINE AIR QUALITY DATA FROM COLSTRIP, MONTANA

E. A. Crecelius, J. C. Laul,
L. A. Rancitelli and R. L. McKeever

Colstrip, located in the range land of eastern Montana, is the site of a coal-fired power plant complex. Unit 1 (350 MW) will begin generating power in late 1975. Concern over the possible environmental impact caused by these power plants led to a study of the air quality before the first plant became operational. As expected the air quality was very good. The air quality will continue to be monitored while Unit 1 is operating.

Three air monitoring sites were established near Colstrip in May 1975. The sites were situated to monitor airborne particulate material and gaseous emission from the power plant stacks. During the summer and fall of 1975 both HiVol and Cascade impactor air particulate samples were collected. Vapor arsenic, mercury and SO₂ samples were also collected.

The HiVol samples were analyzed for approximately 30 elements by instrumental neutron activation (INAA) and

x-ray fluorescence (XRF). The elemental concentration ranges for airborne particulate matter are shown in Table 1. These levels are similar to those observed in other rural areas.

During the summer SO₂ levels were measured at the air monitoring sites. The SO₂ levels range from less than the detection limit of 0.003 to 0.5 µg/m³. These are extremely low levels. In comparison, background SO₂ levels upwind of St. Louis are in the range of 1 to 5 µg/m³.

TABLE 1. Concentration of Elements in 13 HiVol Air Particulate Samples from near Colstrip and from Typical Rural Areas (ng/m³)

Element	Colstrip		Typical Rural
	Mean	Range	Range
Al	450	120-960	240-460
As	2.3	0.2-8.8	0.1-5.6
Ba	4.8	0.7-10	-----
Br	4.0	1.1-7.4	<2-20
Cl	120	40-210	150-300
Co	0.12	0.04-0.3	0.07-0.15
Cr	0.75	0.1-2	0.3-0.8
Cs	0.032	0.01-0.1	-----
Cu	0.012	0.0002-0.03	.007-0.014
Fe	270	70-600	170-430
Hf	0.033	0.01-0.1	0.04-0.06
Hg	0.02	0.004-0.05	0.04-.2
K	160	<20-570	-----
La	0.47	0.2-1.0	0.05-0.3
Mn	4.6	1.9-8.2	2.8-8.2
Na	94	30-130	120-440
Rb	0.63	0.1-2	-----
Sb	0.13	0.03-0.3	0.9-2.1
Sc	0.085	0.02-0.2	0.04-0.2
Se	0.22	0.01-0.4	0.05-0.4
Sm	0.05	0.01-0.16	0.01-0.03
Ta	0.016	0.01-0.1	0.01-0.03
Tb	0.0070	0.006-0.02	0.001-0.01
Th	0.10	0.02-0.2	0.02-0.98
Ti	21	<8-42	-----
V	0.6	<0.2-1.4	0.7-1.4
Zn	5.1	1-10	20-90

THE USE OF ELEMENTAL CONCENTRATION RATIOS
TO DISTINGUISH BETWEEN THE PLUMES
OF DIFFERENT NORTHEASTERN CITIES

J. A. Young, N. S. Laulainen,
L. L. Wendell and T. M. Tanner

The atmospheric concentrations of trace elements have been measured downwind of 11 cities in the northeastern quarter of the United States, to determine whether the concentration ratios of the trace elements are sufficiently unique for different cities that the plume of each city can be identified from the concentration ratios. It was found that the ratios of anthropogenic element concentration generally varied the most from city to city. Between any two cities there were generally several concentration ratios which differed by more than an order of magnitude; so, it appears that the concentration ratios can be used to distinguish the plumes of different cities. Concentration ratios measured on different days indicate that the variations with time of the concentration ratios are much smaller than the differences between different cities.

INTRODUCTION

Measurements of the atmospheric concentrations of various trace elements in and downwind of major U.S. cities by several investigators¹⁻⁷ have shown that the relative concentrations of trace elements (especially those of anthropogenic origin) may vary widely from city to city. Air samples for trace element analysis were collected by aircraft, 20-30 miles downwind of 11 major cities from St. Louis eastward, in December 1974. We sought to determine whether the trace element concentration ratios differ sufficiently from city to city so the source of atmospheric contaminants at rural locations can be determined by comparing the concentration ratios measured at the rural locations and the concentration ratios characteristic of various urban-industrial complexes. This information would prove invaluable in the development and verification of computer models to predict the contaminant concentrations in the northeastern United

States. Air samples were collected again downwind of three of these cities in June 1975 to determine whether the concentration ratios characteristic of these cities remain relatively constant with time. Air filter samples were also collected upwind of some of these cities in December 1974 and June 1975 to determine how the concentration ratios in the incoming air affected the concentration ratios downwind of the cities.

EXPERIMENTAL

One air filter sample was collected 20-30 miles downwind of each city over a 60-100-min period while the Battelle-Northwest Cessna-411 twin engine aircraft flew a 20-30 mile track perpendicular to the wind and 1000-2000 ft above ground level. The air intake for the samples was a 10 cm diameter stainless steel pipe, extended at an angle forward through the side of the fuselage and 50 cm out into the air stream. Ram air pressure forced the air through a

27 cm diameter IPC filter paper at a rate of 23 m³/min, giving a linear velocity of 400 m/min across the filter. The air filters were returned to the Richland laboratory; the December samples have been analyzed for trace elements by x-ray fluorescence and instrumental neutron activation. The June samples are being analyzed now. During the collection of the air filters the SO₂ and Aitken nuclei concentrations were monitored in real time using a Sinex SO₂ monitor and a General Electric Aitken nucleus counter, respectively.

In December 1974 samples were collected downwind of St. Louis, Indianapolis, Columbus, Pittsburgh, Buffalo, Rochester, Allentown-Bethlehem, Baltimore, Washington DC, and New York City, in that order. The choice of cities depended partly on the prevailing weather conditions. A few cities, such as Cleveland and Detroit, that we had intended to sample were missed because of bad weather. Samples were also collected in rural areas on flights between cities, and upwind of Washington DC-Baltimore-Philadelphia. In June 1975

samples were collected upwind of Kansas City and upwind and downwind of Indianapolis, Columbus, and Pittsburgh.

RESULTS

The trace element concentrations measured for December 1974 are reported in Tables 1 and 2. The sulfur was in the form of sulfates. The average trace element concentrations measured at ground level, on an arc perpendicular to the wind 20 miles downwind of St. Louis, by Tanner, et al.⁷ on July 17, 1973, and the concentrations measured in New York City by Rancitelli, et al.⁶ during March 1972 are also reported for comparison. The ratios of the concentrations of trace elements to concentrations of iron and lanthanum (two elements generally considered primarily crustal in origin) were compared to the ratios typical of crustal material. Thus we could determine which elements were present in amounts greater than could be accounted for by crustal material. The ratios indicate that most of the S, Cl, Zn, As, Se, Br, Mo, Sb, Hg, and Pb were of noncrustal origin, and therefore were presumably

TABLE 1. Elemental Concentrations Downwind of Cities

LOCATION	ST. LOUIS	ST. LOUIS	INDIANAPOLIS	COLUMBUS	PITTSBURGH	BUFFALO	ROCHESTER	ALLENTOWN BETHLEHEM	PHILADELPHIA	BALTIMORE	WASHINGTON D.C.	NEW YORK	NEW YORK
TIME (LOCAL)	1100-1300	1004-1159	1404-1544	1006-1141	1355-1535	1159-1308	1434-1602	1456-1609	0837-1000	1018-1130	1225-1349	1404-1506	
DATE	7/17/73	12/5/74	12/5/74	12/6/74	12/6/74	12/7/74	12/7/74	12/10/74	12/11/74	12/11/74	12/11/74	12/12/74	MARCH, 72
ALTITUDE (ft)	GROUND LEVEL	2000	2000	2000	2500	2000	2070	2000-2500	1500-2000	1500-2000	1500-2000	2500-3000	GROUND LEVEL
CONCENTRATIONS IN 10 ⁻⁹ M ⁻³													
SODIUM	720	NO	1100	540	300	25*	560	265	1220	310	890	130	2300
MAGNESIUM	840	NO	NO	1900	NO	700	NO	NO	2050	1070	880	ND	150
ALUMINUM	5270	NO	3430	2560	3800	170	340	3450	2700	1690	1250	940	1790
SULFUR	4400	792	3078	1975	2160	2540	574	1342	1160	683	552	1744	-
CALCIUM	4100	339	5190	299	1660	258	414	6130	1280	1340	684	640	-
CHLORINE	520	330	1360	20**	210**	200	390	1070	580	550	600	130**	2300
SCANDIUM	0.68	0.38	0.66	0.50	1.28	0.19	0.11	0.65	0.63	0.62	0.21	0.17	0.44
TITANIUM	440	8**	231	126	264	33*	23**	221	158	86	40	21	-
VANADIUM	9.9	1.5	9.4	6.3	9.0	9.9	10*	14	7.1*	7.9	15	28	174
CHROMIUM	8	28	6.9	13	58	4.1*	4.0	15	8.8	13	2.6	9.2	153
MANGANESE	79	7.3	37	51	89	9.4	7.5	112	56	15	13	22	60
IRON	2380	1980	2440	1190	4540	372	377	3050	2960	2110	471	624	4820
COBALT	1.06	0.61	1.54	0.98	2.79	0.33	0.42	1.08	1.15	1.10	0.42	0.54	2.2
ZINC	190	121*	81	49	144	32	52	551	83	50*	29	101	741
ARSENIC	3.6	2.5	16	12	33	6.0	13	12	50	8.9	14	7.5	-
SELENIUM	3.69	2.32	1.32	4.01	4.64	0.53*	0.50	4.12	1.47	2.09	0.53	0.83	5.67
BROMINE	32	11	89	22*	43	18	38	31	50*	22	18	41	50
STRONTIUM	30	NO	12	5.6	18	NO	NO	24	10	5.4	7.5	5.4	-
MOLYBDENUM	<3	NO	3A*	NO	14	3.7**	NO	NO	NO	NO	NO	3.6	-
ANTIMONY	1.09	0.43	3.22	4.74	2.51	0.65	5.31	1.94	4.88	0.81	0.66	8.6	12
LANTHANUM	4.0	0.15*	1.90	1.27	2.65	0.57*	1.43	2.05	3.53	0.82	0.63	1.21	9.1
SAMARIUM	0.6	0.023	0.34	0.14	0.37	0.08	0.16	0.35	0.48	0.12	0.10	0.11	0.81
EUROPIUM	0.077	0.105	0.021*	0.11	0.21	0.026	0.016*	0.19	0.085	0.11	0.028	0.027	0.021
TERBIUM	0.13	0.10**	0.02**	0.05*	0.10	0.07**	0.02**	0.51*	NO	0.03**	0.05**	0.05	0.14
MERCURY	0.26	NO	0.26*	0.08**	0.18**	NO	0.07**	NO	NO	0.26**	0.09**	NO	1.26
LEAD	170	66	501	53	273	91	139	226	151	63	48	181	997

NO = NOT DETECTABLE

* COUNTING ERROR LIMITS > 25%

** COUNTING ERROR LIMITS > 30%

TABLE 2. Elemental Concentrations Over Rural Areas

LOCATION	ST. LOUIS TO INDIANAPOLIS	COLUMBUS TO PITTSBURGH	SYRACUSE TO ALLENTOWN	UPWIND OF PHILADELPHIA- BALTIMORE-WASHINGTON	WASHINGTON TO NEW YORK (UPWIND)	NEW YORK TO WASHINGTON (UPWIND)
TIME (LOCAL)	1305-1358	1255-1339	1128-1452	1354-1459	0911-1400	1507-1626
DATE	12/5/74	12/6/74	12/10/74	12/11/74	12/12/74	12/12/74
ALTITUDE (ft)	3500-4000	3500-3800	2000-3700	2500-3500	2500-3500	2500-3000
CONCENTRATIONS IN 10^{-9} g m^{-3}						
SODIUM	890	1200	780	180	1530	40
MAGNESIUM	ND	ND	630*	ND	ND	1440
ALUMINUM	70	60	950	1700	1140	1290
SULFUR	51*	220	1090	677	2040	1800
CALCIUM	32	189	985	1530	900	1300
CHLORINE	510	910	810	530	640	165
SCANDIUM	0.007*	0.59	0.20	2.25	0.17	0.56
TITANIUM	47	22	40	67	42	50
VANADIUM	0.1**	7	7.5	54	33	38
CHROMIUM	1.5*	43	7.4	95	6.6	16
MANGANESE	1**	1.5**	19	18	24	25
IRON	80**	3070	840	9600	620	2210
COBALT	ND	1.05	0.61	5.26	0.34*	1.36
ZINC	13*	286	50	202	168	489
ARSENIC	3.5	22	13*	15	30	8.0
SELENIUM	0.14*	3.40	0.35	7.22	0.18	1.39
BROMINE	5.6	3	31	61	67	20
STRONTIUM	6.2	ND	6.3	ND	5.2	4.2
MOLYBDENUM	ND	ND	ND	2.7	3.7	ND
ANTIMONY	0.44*	8.7	2.31	1.76	16	2.54
LANTHANUM	ND	2.27	0.84	0.85	3.38	0.75
SAMARIUM	0.14*	0.26	0.18	0.18	0.36	0.10
EUROPIUM	0.001**	0.17	0.017	0.40	0.031	0.10
TANTALUM	0.10*	0.10**	0.004**	ND	ND	0.20*
MERCURY	0.19*	ND	0.20*	0.48	0.18*	0.56
LEAD	3.3	8.4	121	166	210	116

ND = NOT DETECTABLE

* COUNTING ERROR LIMITS > 25%

** COUNTING ERROR LIMITS > 50%

anthropogenic, although the chlorine could have been of marine origin. Vanadium, Na, and possibly Ca also appeared to be primarily of non-crustal origin for some of the cities. The ratios of the concentrations of Mg, Al, Sc, Ti, Cr, Mn, Sr, Sm, Eu, Ta, and Co to the concentrations of Fe and La were similar to the ratios in crustal materials. Still, these elements could possibly have had significant anthropogenic contributions to their concentration since 1) the crustal concentrations are not very well known and are variable, and 2) there could be a significant anthropogenic contribution to the Fe and La concentrations downwind of cities.

The correlation coefficients between pairs of elements were calculated to determine which elements

were highly correlated, and therefore probably came from the same sources. We also wished to identify those pairs of elements which were completely uncorrelated. The concentration ratios of uncorrelated elements were expected to vary most from city to city, and therefore would be most useful in distinguishing between the plumes of different cities. The elements of crustal origin generally showed correlation with many of the other crustal elements, while the anthropogenic elements generally showed less correlation with other elements. The group of elements Sc, Cr, Fe, Co, Se, and Eu showed very high correlations with each other, but few correlations with other elements. Of this group Se is anthropogenic, but the other elements are generally considered to be of crustal origin. The high correlations between these elements, and the fact

that the Sc, Cr, Fe, and Co concentration ratios were fairly close to average crustal ratios, suggests that these elements are released by some industrial process(es) which emits Sc, Cr, Fe, and Co in roughly the same ratio found in crustal material, but which emits Se in relatively greater amounts. The elements Na, S, Va, and Sb were uncorrelated with the largest number of other elements, thus concentration ratios involving these elements would be the most useful in distinguishing the plumes of different cities.

The concentration ratios of selected pairs of uncorrelated elements of noncrustal origin are shown in Table 3. The ratios varied considerably, sometimes by more than two orders of magnitude between different cities. Generally, however, only some of the ratios were significantly different between any two given cities. Each concentration ratio varied significantly between some pairs of cities, but not between others. For

each pair of cities, the concentration ratios of elements of noncrustal origin which differed by more than an order of magnitude for the two cities are shown in Table 4. For most of the city pairs several elemental ratios differed by more than an order of magnitude. St. Louis-Allentown, Indianapolis-Rochester, and Indianapolis-Washington were the only city pairs which did not have at least one concentration ratio differing by more than an order of magnitude.

The elemental concentration ratios may be used to distinguish between the plumes of different cities only if the ratios in the plumes remain relatively constant with time and distance downwind of the cities, at least until the plumes of different cities intersect. The concentration ratios measured by Tanner, et al.⁷ at ground level from 20-80 miles downwind of St. Louis showed very little change with distance downwind of St. Louis.

TABLE 3. Concentration Ratios of Selected Elements

LOCATION	CONCENTRATION RATIOS																	
	Na V	Na Zn	Na Se	S Na	S Zn	S Se	V Se	V Sb	Zn V	Zn As	Zn Sb	As V	As Se	Br Se	Br Sb	Sb Se	Pb Se	Pb Sb
ST. LOUIS (7/17/73) 20 MILES DOWNWIND	73	3.8	195	6.1	23	1190	2.7	9.1	19	53	174	0.36	0.98	8.7	26	0.30	46	156
ST. LOUIS (7/17/73) 60 MILES DOWNWIND	65	5.2	279	7.3	38	2090	4.3	11.2	13	42	140	0.30	1.3	9.4	25	0.38	54	140
ST. LOUIS (12/5/74)					6.5	341	0.65	3.5	81	48	281	1.7	1.1	4.7	26	0.19	28	153
INDIANAPOLIS	117	13	833	2.8	36	2300	7.1	2.9	8.9	5.3	26	1.7	12	67	28	2.4	382	157
COLUMBUS	86	11	135	3.7	40	493	1.6	1.3	7.8	4.1	10	1.9	3.0	5.5	4.6	1.2	13	11
PITTSBURGH	0.33	2.1	65	7.2	15	466	1.9	3.5	16	4.4	57	3.7	7.1	9.3	17	38	59	109
BUFFALO	2.5	0.78	47	101	79	4780	19	15	3.2	5.3	49	0.61	11	34	28	1.2	172	140
ROCHESTER	54	11	1120	1.0	11	1150	21	2.0	5.0	4.0	9.8	1.3	26	76	7.2	11	278	26
ALLENTOWN-BETHLEHEM	19	0.48	64	5.1	2.4	326	3.4	7.2	39	46	284	0.86	2.9	7.5	16	0.47	55	116
PHILADELPHIA	172	15	830	1.0	14	23	4.8	1.5	12	1.7	17	7.0	34	34	10	3.3	103	31
BALTIMORE	39	6.2	145	2.2	14	327	3.8	9.8	6.3	5.6	62	1.1	4.3	11	27	0.39	30	78
WASHINGTON D.C.	61	31	1679	0.62	19	1040	27	22	2.0	2.1	44	0.97	26	34	27	1.2	91	73
NEW YORK CITY (12/12/74)	4.6	1.3	157	13.4	17	2100	31	3.3	3.6	13	12	0.27	243	49	4.7	10	218	21
NEW YORK CITY (MARCH, 1972)	13	3.1	53	--	--	--	31	15			62			8.8	4.2	2.1	176	83
ST. LOUIS TO INDIANAPOLIS		68	6357	0.06	3.9	364				3.7	30		25	40	13	3.1	24	7.5
COLUMBUS TO PITTSBURGH	63	4.2	353	0.18	0.77	65	5.6	2.2	15	13	33	1.2	6.5	0.88	0.35	2.5	2.5	0.97
SYRACUSE TO ALLENTOWN	104	16	2230	1.4	22	3110	21	3.2	6.7	3.8	22	1.7	37	89	13	6.6	346	52
UPWIND OF PHILADELPHIA - BALTIMORE - WASHINGTON D.C.	3.3	0.89	25	3.8	3.4	94	7.5	31	3.7	13	115	0.28	2.1	8.4	35	0.25	23	94
WASHINGTON TO NEW YORK (UPWIND)	46	9.1	8500	1.3	12	11300	183	2.1	5.1	5.6	11	0.91	167	372	4.2	99	1170	13
NEW YORK TO WASHINGTON (UPWIND)	2.7	0.082	29	45	3.7	1290	11	5.9	33	61	193	0.53	5.8	14	7.9	1.8	83	46

TABLE 4. Concentration Ratios Which Differ by More than an Order of Magnitude Between Pairs of Cities

	INDIANAPOLIS	COLUMBUS	PITTSBURGH	BUFFALO	ROCHESTER	ALLEN TOWN BETHLEHEM	PHILADELPHIA	BALTIMORE	WASHINGTON D.C.	NEW YORK CITY
ST. LOUIS	$\frac{Zn}{Sb}, \frac{As}{Se}, \frac{Br}{Se}, \frac{Sb}{Se}$	$\frac{S}{Cl}, \frac{Cl}{As}, \frac{Cl}{Sb}, \frac{Cl}{Pb}$ $\frac{Zn}{V}, \frac{Zn}{As}, \frac{Zn}{Sb}, \frac{Pb}{Sb}$	$\frac{Cl}{As}, \frac{Zn}{As}, \frac{Sb}{Se}$	$\frac{Na}{V}, \frac{S}{Na}, \frac{S}{Zn}, \frac{S}{Se}$ $\frac{Cl}{V}, \frac{Zn}{V}$	$\frac{V}{Sb}, \frac{Zn}{V}, \frac{Zn}{As}, \frac{Zn}{Sb}$ $\frac{As}{Se}, \frac{Br}{Se}, \frac{Sb}{Se}$		$\frac{Zn}{As}, \frac{Zn}{Sb}, \frac{As}{Se}, \frac{Sb}{Se}$ $\frac{Pb}{Cl}$	$\frac{Zn}{V}$	$\frac{V}{Sb}, \frac{Zn}{V}, \frac{Zn}{As}$	$\frac{Na}{V}, \frac{Cl}{V}, \frac{Cl}{Sb}, \frac{Cl}{Se}, \frac{V}{Zn}$ $\frac{Zn}{Sb}, \frac{As}{Se}, \frac{Br}{Se}, \frac{Sb}{Se}$
INDIANAPOLIS		$\frac{Cl}{S}, \frac{Cl}{V}, \frac{Cl}{As}, \frac{Cl}{Sb}$ $\frac{Sb}{V}, \frac{As}{Sb}, \frac{Sb}{Se}, \frac{Sb}{Sb}$	$\frac{Na}{Sb}, \frac{Cl}{As}, \frac{Cl}{Sb}, \frac{Sb}{Se}$ $\frac{Na}{Mo}, \frac{Cl}{Mo}$	$\frac{Na}{S}, \frac{Na}{V}, \frac{Na}{Zn}$ $\frac{Na}{Sb}$		$\frac{Na}{Zn}, \frac{Na}{Sb}, \frac{S}{Zn}$ $\frac{Zn}{Sb}$	$\frac{S}{Se}$	$\frac{Pb}{Se}$		$\frac{Na}{V}, \frac{Na}{Se}, \frac{Cl}{V}, \frac{Cl}{Sb}$ $\frac{As}{Se}, \frac{Cl}{Se}, \frac{Mo}{Se}$
		COLUMBUS								
			PITTSBURGH							
				BUFFALO						
					ROCHESTER					
						ALLEN TOWN BETHLEHEM				
							PHILADELPHIA			
								BALTIMORE		
									WASHINGTON D.C.	

* RATIOS DIFFERENT BY MORE THAN TWO ORDERS OF MAGNITUDE

The ratios must also be relatively independent or at least vary predictably with time, if they are to be used to distinguish between the plumes of different cities. The various concentration ratios of anthropogenic elements measured downwind of St. Louis in the summers of 1972 through 1975 as part of the Metromex program and in December 1974 have averaged about 40% between different days. The various ratios measured in New York City on July 17, 1973, by Rancitelli, et al.⁶ and those measured downwind of New York on December 12, 1974, also averaged about 40%. However, the various ratios measured by aircraft downwind of Pittsburgh on December 13, 1975, and those measured on June 13, 1975, averaged only 22%. These concentration

ratios measured at different times downwind of a given city differ by far less than the order of magnitude or more difference between ratios of different cities. Thus, the ratios are probably constant enough with time to be used to distinguish between the plumes of different cities.

Probably a large part of the variance between concentration ratios measured at ground level on different days resulted from the large scatter of the elemental concentrations measured in samples collected at ground level over 1-2-hr periods. For example, the lead concentrations between sequential samples collected at a given location downwind of St. Louis in the summer of 1974 varied by 35%. The concentrations measured in samples collected by aircraft have shown

much less variation. The Pb concentrations measured at three different altitudes downwind of Pittsburgh on June 13, 1975, and concentrations measured at three different altitudes downwind of St. Louis in August of 1975 averaged variances of only 14%. The concentration ratios measured in samples collected while flying crosswind downwind of a city are probably much more representative of the city plume as a whole than are ratios measured at ground level.

The elemental concentration ratios in the plume of a city will be unique only if the elements in the plume were introduced into the atmosphere primarily by that city, and were not already present in the air beforehand. The concentrations of most anthropogenic elements were appreciably lower in the samples collected upwind of Columbus, Indianapolis, and Pittsburgh in June of 1975 than in the samples collected downwind of these cities. The concentrations of S, Zn, Se, Pb, As, and Br upwind of Pittsburgh averaged 40% of the concentrations downwind of Pittsburgh. When the upwind concentrations were subtracted from the downwind concentrations to obtain the net concentrations produced by Pittsburgh their ratios changed by an average of only 28%. However, rural samples collected farther east in December 1974 generally contained considerably higher concentrations of the anthropogenic trace elements. In fact, the concentrations measured in the sample collected on a flight from upwind (west) of Washington, DC to upwind (west) of Philadelphia contained

higher concentrations of some of the elements than did the samples collected downwind of Philadelphia, Baltimore, or Washington, DC. Elemental concentrations will have to be measured both upwind and downwind of cities to obtain the net addition of elemental contaminants by the cities in cases where the air entering the cities is heavily contaminated. Possibly the elemental concentration ratios characteristic of the plumes of the eastern seaboard cities may be measured only when the wind is from the east.

SUMMARY AND CONCLUSIONS

The plumes of eleven northeastern cities could be distinguished from one another from differences in the measured elemental concentration ratios. The measurements of elemental concentration ratios downwind of St. Louis during project Metromex indicate that the ratios remain relatively constant with distance downwind, at least until another major plume is intersected. The concentration ratios measured downwind of St. Louis, New York, Indianapolis, Columbus, and Pittsburgh on different dates were somewhat different, but the changes in the ratios with time were unlikely to be great enough to prevent the use of the ratios to distinguish between the plumes of different cities. To obtain accurate values for the ratios characteristic of a given city, the upwind concentrations should be subtracted from the downwind concentrations to obtain the net addition by the city.

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AIR FLOW AND DISPERSION

The dispersion of a pollutant, as it is introduced into the atmosphere and moves downwind from the source position, is the phenomenon which has allowed the atmosphere to be used for waste disposal purposes. In an attempt to curtail abuse of the atmosphere, government agencies have been established to regulate atmospheric pollution. For the most part this regulation is based on dispersion estimates derived from the Gaussian assumption, with diffusion parameters which were empirically derived in a practical manner for rather limited purposes. The almost universal application of these techniques has been quite disconcerting, especially to those who introduced them. The work reported in this section examines some of the fundamentals of atmospheric dispersion with theoretical analyses as well as detailed experimental investigations.

Emphasis has been placed during the past year on analyzing and summarizing previous experimental dispersion data with more realistic and fundamentally sound approaches to plume behavior. The goal is to finalize improved short-range dispersion models from existing data, removing inconsistencies and inadequacies in presently applied assessment models. Dispersion and transport efforts in the future should aim toward evaluating plume behavior on meso and regional scales. The complex features of flow and dispersion through storms, and in the vicinity of significant terrain characteristics influencing local to regional circulations must receive future emphasis.

- **THEORETICAL STUDIES IN ATMOSPHERIC SCIENCES**
- **ATMOSPHERIC BOUNDARY LAYER STUDIES**
- **PRECIPITATION SCAVENGING**

A SIMPLE DEVELOPMENT OF COVARIANT DIFFERENTIATION

W. G. N. Slinn

A simple method is developed to generalize the partial derivatives of components of tensors to curvilinear coordinates.

INTRODUCTION

Quantities obtained from the partial differentiation of components of tensors with respect to the coordinates are themselves the components of tensors, only under linear transformations of the coordinates. Thus, if new coordinates $\bar{y}^i$ are obtained from a linear transformation of the Cartesians, y^i , that is,

$$\bar{y}^i = a_j^i y^j \quad (1)$$

with the a_j^i constants and α_j^i the inverse of a_j^i , then for a tensor whose components are t_j^i in Cartesians,

$$\frac{\partial t_j^i}{\partial y^k} = \frac{\partial}{\partial y^k} \left(a_\alpha^i \alpha_j^\beta t_\beta^\alpha \right) \frac{\partial y^\ell}{\partial y^k} \quad (2)$$

Using (1) and the constancy of a_j^i and α_j^i , this yields

$$\frac{\partial t_j^i}{\partial y^k} = a_\alpha^i \alpha_j^\beta \alpha_k^\gamma \frac{\partial t_\beta^\alpha}{\partial y^\gamma} \quad (3)$$

which demonstrates the tensorial nature of the derivative.

It can be rather complicated to develop a generalization for partial

differentiation (that is, to the concept of covariant differentiation) for a general functional coordinate transformation, say of the form

$$x^i = x^i(y^1, y^2, y^3) \quad (4)$$

where x^i are general curvilinear coordinates and y^i are Cartesian coordinates. Thus, McConnel¹ introduced the concept of a parallel vector field and had to extend the quotient theorem; Block² introduced basis vectors and their derivatives; and Einstein³ used the invariance of the derivative of a scalar with respect to an arc length, a method which appeared simple but only because he had previously developed the equation for a geodesic. This note demonstrates a simple method requiring the introduction of no new concepts.

THE METHOD

The proposed method is best described by applying it to a simple case. Let V^i and v^i be the contravariant components of a vector $\vec{V}$ in Cartesian and the curvilinear coordinates, respectively. Then, with (4),

$$v^i = \frac{\partial y^i}{\partial x^k} V^k \quad (5)$$

What is desired is a derivative that reduces to partial differentiation in Cartesians. Then from (5)

$$\frac{\partial V^i}{\partial y^j} = \frac{\partial}{\partial x^l} \left(\frac{\partial y^i}{\partial x^l} v^k \right) \frac{\partial x^l}{\partial y^j}. \quad (6)$$

Expanding (6) and multiplying by $\partial y^i / \partial x^m$ (and summing on j) gives

$$\frac{\partial y^j}{\partial x^m} \frac{\partial V^i}{\partial y^j} = \frac{\partial y^i}{\partial x^k} \frac{\partial v^k}{\partial x^m} + v^k \frac{\partial^2 y^i}{\partial x^m \partial x^k}. \quad (7)$$

Multiplying (7) by $\partial x^n / \partial y^i$ then gives

$$\frac{\partial x^n}{\partial y^i} \frac{\partial y^j}{\partial x^m} \frac{\partial V^i}{\partial y^j} = \frac{\partial v^n}{\partial x^m} + v^k \frac{\partial^2 y^i}{\partial x^m \partial x^k} \frac{\partial x^n}{\partial y^i}. \quad (8)$$

But, from the definition of the metric tensor

$$g_{\alpha\beta} = \frac{\partial y^i}{\partial x^\alpha} \frac{\partial y^i}{\partial x^\beta} \quad (9)$$

and its inverse $g^{\alpha\beta}$, we have

$$g^{\beta\gamma} g_{\alpha\beta} = \delta_\alpha^\gamma = g^{\beta\gamma} \frac{\partial y^i}{\partial x^\alpha} \frac{\partial y^i}{\partial x^\beta}. \quad (10)$$

Therefore, multiplying (10) by $\partial x^\alpha / \partial y^i$, we see that the last term in (8) can be changed to

$$v^k g^{\beta n} \frac{\partial^2 y^i}{\partial x^m \partial x^k} \frac{\partial y^i}{\partial x^\beta} = v^k \left\{ \begin{matrix} n \\ mk \end{matrix} \right\} \quad (11)$$

which defines the Christoffel symbol of the second kind. Thus (8) becomes

$$v^n_{,m} = \frac{\partial v^n}{\partial x^m} + v^k \left\{ \begin{matrix} n \\ mk \end{matrix} \right\} = \frac{\partial x^n}{\partial y^i} \frac{\partial y^j}{\partial x^m} \frac{\partial V^i}{\partial y^j} \quad (12)$$

which is the desired generalization from partial differentiation to the covariant derivative, $v^n_{,m}$. Obviously $v^n_{,m}$ are the mixed components of a second order tensor since the transformation rule is correct.

Application of this method to higher order tensors follows similarly. It is hoped that the method shown here is of some benefit in the study of tensors.

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A PITFALL IN THE APPLICATION OF TENSOR
ANALYSIS TO FLUID MECHANICS

W. G. N. Slinn

An inconsistency is exposed between the conventional use of the convective acceleration in fluid mechanics, $\vec{v} \cdot \nabla \vec{v}$, and the definition for the gradient of a vector $\nabla \vec{v} = v_{i,j} \hat{e}^i \hat{e}^j$. Two remedies are proposed.

INTRODUCTION

In fluid mechanics, the gradient of the velocity vector appears in the momentum equation in conjunction with the total time derivative. Customarily, this is written as

$$\frac{D\vec{v}}{Dt} = \frac{\partial \vec{v}}{\partial t} + \vec{v} \cdot \nabla \vec{v}. \quad (1)$$

In tensor analysis, the gradient of a vector is defined (e.g., see Block¹) as

$$\nabla \vec{v} = \frac{\partial \vec{v}}{\partial x^i} \hat{e}^i \quad (2)$$

where the summation convention is used and $\hat{e}^i$ is the set of reciprocal basis vectors defined via

$$\hat{e}^i = \nabla x^i. \quad (3)$$

Here x^i is a set of curvilinear coordinates, not necessarily orthogonal, and

$$\nabla x^i = \frac{\partial x^i}{\partial \xi^j} \hat{c}^j \quad (4)$$

where ξ^i is the Cartesian coordinates with orthonormal basis vectors $\hat{c}^i$.

This report notes an inconsistency between Eq. (1) and (2) and presents remedies.

THE INCONSISTENCY

If the derivative in Eq. (2) is expanded it becomes

$$\nabla \vec{v} = v_{i,j} \hat{e}^i \hat{e}^j \quad (5)$$

where $v_{i,j}$ is the covariant derivative of the covariant components of $\vec{v}$

$$v_{i,j} = \frac{\partial v_i}{\partial x^j} - \left\{ \begin{matrix} i \\ k \end{matrix} \right\} v_k \quad (6)$$

in which the usual symbol for the Christoffel symbol of the second kind appears. Then, from (5),

$$\vec{v} \cdot \nabla \vec{v} = g^{ik} v_k v_{i,j} \hat{e}^j = v^i v_{i,j} \hat{e}^j \quad (7)$$

where g^{ik} are the contravariant components of the metric tensor. In Cartesian coordinates (7) reduces to

$$v_i \frac{\partial v_i}{\partial \xi^j} \hat{c}^j. \quad (8)$$

In contrast, the conventional meaning for the convective acceleration in

fluid mechanics reduces, in Cartesian coordinates, to

$$\vec{v} \cdot \nabla \vec{v} = v_i \frac{\partial v_j}{\partial x^i} \hat{c}_j. \quad (9)$$

Clearly (8) and (9) are inconsistent.

REMEDIES

There are two obvious ways to remedy the inconsistency exposed above. In the first, which will probably not gain wide acceptance, the convective acceleration is written as $(\nabla \vec{v}) \cdot \vec{v}$. Then, with (2) or (5), we obtain

$$(\nabla \vec{v}) \cdot \vec{v} = v_{i,j} \hat{e}^i \hat{e}^i \cdot v^k \hat{e}_k \quad (10)$$

which becomes, upon using $\hat{e}^j \cdot \hat{e}_k = \delta_k^j$,

$$(\nabla \vec{v}) \cdot \vec{v} = v^j v_{i,j} \hat{e}^i. \quad (11)$$

Clearly this reduces to (9) in Cartesian coordinates.

A second remedy is to reject the definition of the gradient of a vector as given by (2) and instead define

$$\nabla \vec{v} = \hat{e}^i \frac{\partial \vec{v}}{\partial x^i} = \hat{e}^i v_{j,i} \hat{e}^j. \quad (12)$$

With (12) we can retain the customary definition of the convective acceleration for we then obtain

$$\vec{v} \cdot \nabla \vec{v} = v^k \hat{e}_k \cdot v_{j,i} \hat{e}^i \hat{e}^j = v^i v_{j,i} \hat{e}^j \quad (13)$$

which is the same as (11) and reduces to (9) in Cartesian coordinates.

CONCLUSION

It is important to notice and remedy the inconsistency between the customary use of $\vec{v} \cdot \nabla \vec{v}$ in fluid mechanics and the (logical) definition for $\nabla \vec{v}$ as given, for example, in Block.¹ Otherwise, one obtains an erroneous momentum equation for fluids.

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DIFFUSION WITH DEPOSITION AT SHORT RANGE

W. G. N. Slinn

Experimental data for the vertical profiles of krypton and a particulate tracer released simultaneously are qualitatively compared with a simple theoretical model of diffusion at short range.

INTRODUCTION

In a recent series of tests, Nickola and Clark¹ compared the vertical concentrations of two tracers released simultaneously on the Hanford diffusion grid. The tracers were the inert gas, ⁸⁵Kr, and a particulate tracer, ZnS. The objective of the tests was to compare the two profiles, for nondepositing Kr and for the depositing particles, and thereby gain information about the deposition flux of the particles. So far, only preliminary results are available; these are illustrated with a figure from Nickola and Clark's paper, shown here as Figure 1. It is hoped that funding will permit future tests to be undertaken in this most informative test series. This note qualitatively compares these experimental results against simple theoretical predictions.

SIMPLIFIED THEORY

As a first attempt to model the test mathematically, we assume that either contaminants' air concentration, χ , is governed by

$$\bar{u} \frac{\partial \chi}{\partial x} = K_y \frac{\partial^2 \chi}{\partial z^2} + K_z \frac{\partial^2 \chi}{\partial z^2} + v_s \frac{\partial \chi}{\partial z} \quad (1)$$

with the diffusivities treated as constants and where v_s is the pollutants' gravitational settling speed (assumed to be zero for Kr since even if its initial temperature was substantially different from the air

temperature at the release height, the quantity released was so small that buoyancy effects would be expected to be negligible). To (1) is added the boundary condition at the ground

$$\left[K_z \frac{\partial \chi}{\partial z} + v_s \chi \right]_{z=0} = v_d \chi \Big|_{z=0} \quad (2)$$

where v_d is Calder's deposition velocity,² expected to be zero for Kr. In later modeling attempts, z -dependencies of the diffusivities and other boundary conditions should be considered. The assumption of x -independent diffusivities and negligible wind shear effects on diffusion should be acceptable at sufficiently short downwind distances. The initial condition for the problem is taken as $\chi(x=0, y, z) = (\bar{Q}/\bar{u}) \delta(y) \delta(z-h)$.

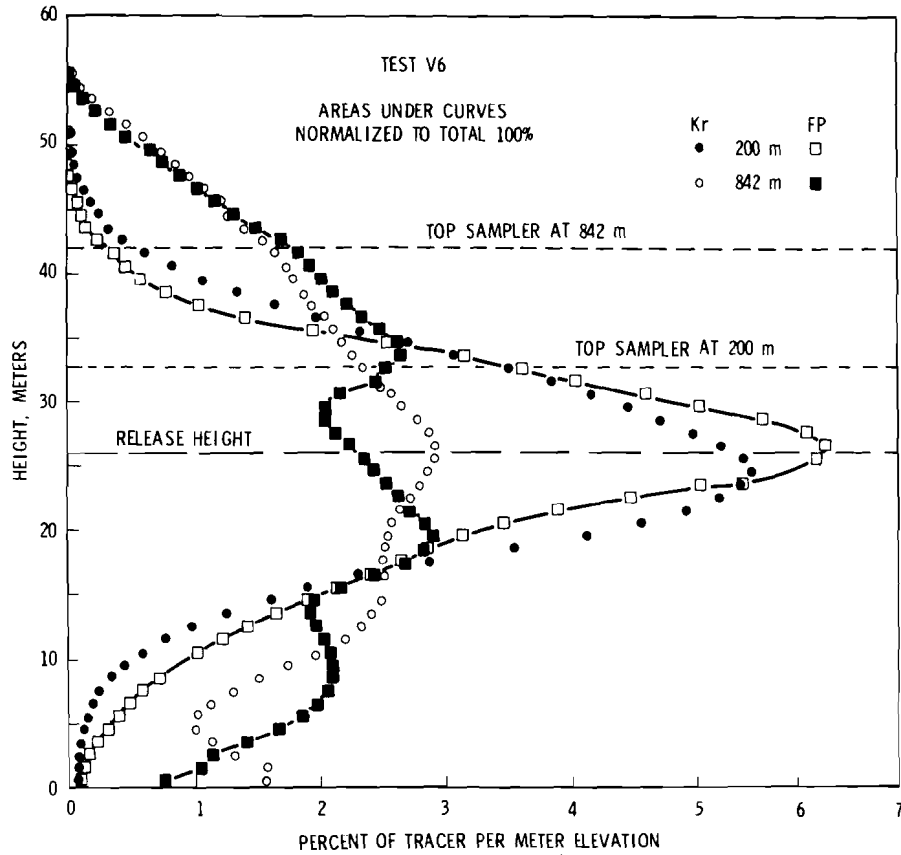
A solution to (1) and (2) can be found perhaps most easily in the following manner. Let

$$\chi(x, y, z) = \psi(x, z) Y(y, z) \quad (3)$$

then the solution to

$$\bar{u} \frac{\partial Y}{\partial x} = K_y \frac{\partial^2 Y}{\partial y^2} \quad (4)$$

subject to $Y(0, y) = (\bar{Q}/\bar{u}) \delta(y)$ and vanishingly small concentrations and fluxes at $y = \pm \infty$ is found to be



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FIGURE 1. Vertical Profiles of Fraction of Tracer Per Unit Height for Nickola and Clark's test V-6. Total amounts of tracers were normalized to 100%.

$$Y(x, y) = \frac{\dot{Q}}{\bar{u}} \frac{1}{(4\pi K_y x/\bar{u})^{1/2}}$$

$$\exp\left\{\frac{-y^2}{4 K_y x/\bar{u}}\right\}. \quad (5)$$

The remaining problem is to solve

$$\bar{u} \frac{\partial \psi}{\partial x} = K_z \frac{\partial^2 \psi}{\partial z^2} + v_s \frac{\partial \psi}{\partial z} \quad (6)$$

subject to

$$\psi(0, z) = \delta(z - h) \quad (7)$$

and

$$\left[K_z \frac{\partial \psi}{\partial z} + v_s \psi\right]_{z=0} = v_d \psi \Big|_{z=0}. \quad (8)$$

Now let

$$\psi(x, z) = \Omega(x, z)$$

$$\exp\left\{-\frac{v_s}{2 K_z} (z - h) - \frac{v_s^2 x}{4 K_z \bar{u}}\right\}. \quad (9)$$

Then (6) becomes

$$\bar{u} \frac{\partial \Omega}{\partial x} = K_z \frac{\partial^2 \Omega}{\partial z^2} \quad (10)$$

subject to $\Omega(0, z) = \delta(z - h)$

and

$$K_z \frac{\partial \Omega}{\partial z} \Big|_{z=0} = (v_d - \frac{1}{2} v_s) \Omega \Big|_{z=0}. \quad (11)$$

This problem is one familiar in heat conduction theory (e.g., see Carslaw and Jaeger,³ p. 115) and has the solution

$$\begin{aligned} \Omega = & \frac{1}{(4 \pi K_z x/\bar{u})^{1/2}} \left[\exp \left\{ -\frac{(z-h)^2}{4 K_z x/\bar{u}} \right\} \right. \\ & + \exp \left\{ -\frac{(z+h)^2}{4 K_z x/\bar{u}} \right\} \\ & - q \exp \left\{ q(z+h + K_z q x/\bar{u}) \right\} \\ & \left. \operatorname{erfc} \left\{ (K_z x/\bar{u})^{1/2} \left[q + \frac{(z+h)}{2 K_z x/\bar{u}} \right] \right\} \right] \end{aligned} \quad (12)$$

where

$$q = (v_d - \frac{v_s}{2}) / K_z. \quad (13)$$

This completes the solution for $\chi = \psi Y$ where Y is given by (5) and ψ is

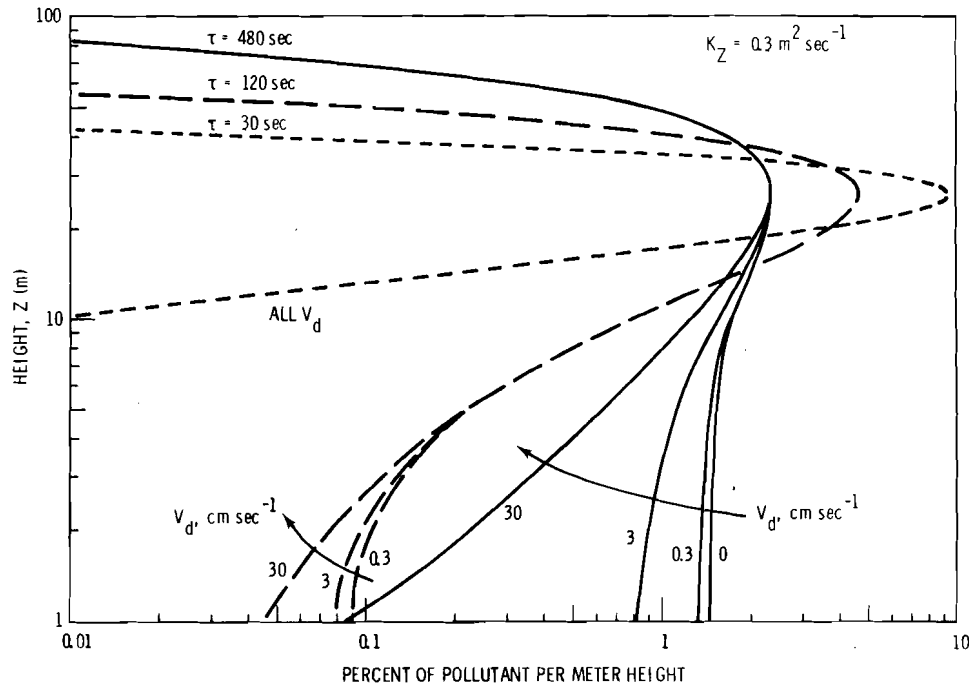
given by (9) with Ω as in (12). One can easily see that when $v_s = v_d = 0$ this result reduces to the familiar result

$$\begin{aligned} \chi(x, y, z; v_s = v_d = 0) \\ = & \frac{\dot{Q}}{4 \pi x (K_y K_z)^{1/2}} \exp \left\{ -\frac{y^2}{4 K_y x/\bar{u}} \right\} \\ & \left[\exp \left\{ -\frac{(z-h)^2}{4 K_z x/\bar{u}} \right\} + \exp \left\{ -\frac{(z+h)^2}{4 K_z x/\bar{u}} \right\} \right]. \end{aligned} \quad (14)$$

It is not so easy to see, but nevertheless can be demonstrated by expanding the complementary error function, that for a perfect sink, i.e., $v_d \rightarrow \infty$, then (12), etc. reduces to the familiar result [which is (14), but with the two exponentials subtracted rather than added].

CONCLUSIONS

Figure 2 shows a plot of these results integrated over all crosswind



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FIGURE 2. A Plot of the Theoretical Results Derived in the Text with K_z , $\tau = x/\bar{u}$ and v_d as Shown and $h = 26 \text{ m}$

distances y and for the particular choices of $K_z = 0.3 \text{ m}^2 \text{ sec}^{-1}$ and $h = 26 \text{ m}$. This figure is to be compared with the experimental data in Figure 1. It should be expected that the theory would not be able to reproduce the fluctuations of the data from a single test, but at best would be able to represent an average profile for many tests. Therefore, the discrepancy between theory and the test results at heights above about 10 m should not cause much concern. There is, however, a curious increase shown in the data for Kr below 5 m which will not follow from the theory

presented. It is too soon to tell whether this discrepancy represents an important physical phenomenon unaccounted by the theory or merely a statistical fluctuation which in the mean will disappear. Further tests would be most welcome. Qualitatively, by comparing Figures 1 and 2, it appears that the deposition velocity for the particulate tracer was 0 (10 cm sec⁻¹).

ACKNOWLEDGEMENTS

The author thanks P. W. Nickola for permission to reproduce Figure 1.

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SOME OBSERVATIONS OF THE RELATIVE CONTRIBUTION OF REAL
PLUME DIFFUSION AND OF MEANDER TO APPARENT PLUME DIFFUSION

P. W. Nickola

The contribution of real plume diffusion to the apparent plume considered in hazards appraisals varies from less than 10% to more than 80%. No dependence on conventional stability measurements is obvious in the 8 field experiments examined.

INTRODUCTION

Most computational formulae and models dealing with diffusion in the lower atmosphere pertain to concentrations averaged over a period of tens of minutes or longer. For instance, the equation recommended in the NRC Regulatory Guides 1.2 and 1.3 for determining "short-term average centerline...concentration" employ the terms σ_y and σ_z representing horizontal and vertical dispersion in the atmosphere. Curves relating σ_y and σ_z to distance from source were originally empirically developed from tracer releases of 10 min duration. These curves--or slightly modified curves--are generally accepted as hourly mean values.¹

However, the mean concentration experienced at a fixed point over, say, an hour results from a real plume (the plume observed by the eye) that meanders back and forth over the point of measurement. Not only should the tolerance of a receptor to the mean concentration be of concern, but also tolerance to the short-term fluctuations above this mean.

Gifford, in his fluctuating plume model,² related the spreading or true diffusion in the real plume and the meander of the real plume to the variance in concentration in the mean or apparent plume. Ramsdell³ presented graphically measurements depicting this relationship. Since Ramsdell's first data, several other field diffusion experiments have been carried

out and/or analyzed at Hanford. These new experiments provide the data base for this note.

The ⁸⁵Kr tracer system⁴ used on the Hanford field diffusion grid presents data in a form suitable for describing characteristics of the real plume. This system simultaneously records concentrations at more than 100 field locations as a series of end-to-end short-period concentrations. The short-period time increment in the current experiments was 38 sec for the "C" tests, and 10 sec for the "V" tests.

MODEL

Variance of the time-integrated or apparent plume crosswind concentration distribution ($\bar{Y}^2$)* can be computed after summing the short-period concentrations for each sampling location. Samplers are located on arcs concentric about the tracer release point.

The variance $\bar{Y}^2$ of the apparent plume can be related to $\bar{D}^2$, the variance in the real plume, and $\bar{M}^2$, the meander or variance of the distribution of the positions of the center of mass of the real plume by the equation

$$\bar{Y}^2 = \bar{D}^2 + \bar{M}^2 .$$

* The nomenclature used for variances in this note is the same as that used by Ramsdell.³

THE EXPERIMENTS

Table 1 lists the field experiments with some meteorology and commonly used descriptors of the atmosphere stability. Although tracer releases lasted from 14 to 30 min, the leading and trailing portions of the plume were not analyzed. The periods analyzed varied from 10 to 24 min.

Figures 1 and 2 give examples of the meteorology and of the general character of plumes during 17 min of plume passage for stable test V6 and unstable test V7. Winds during actual tracer release are missing from the strip chart records. (Charts were driven at 3 in./min during tracer release.) However, the character of the winds can be discerned from the before- and after-release record. The time-history of range of the above-background 10-sec tracer readings is suggested by the outlines at the bottom of the figures. The dots indicate location of peak concentration during each 1-min period.

Above the plume time history plots on Figures 1 and 2 are depicted ground-level crosswind profiles for the 1-min periods with the largest and smallest crosswind integrated concentration. Also note that similar largest and smallest profiles for 10-sec periods within the 1-min periods are shown. The letter "R" on the abscissa is merely a reference azimuth for comparing the depicted distributions.

DISCUSSION AND ANALYSIS

Figures 1 and 2 show that the character of crosswind profile varies a great deal more during test V7 than during test V6. These figures also suggest that the magnitude of Y^2 , D^2 and M^2 would be larger for test V7 than for V6.

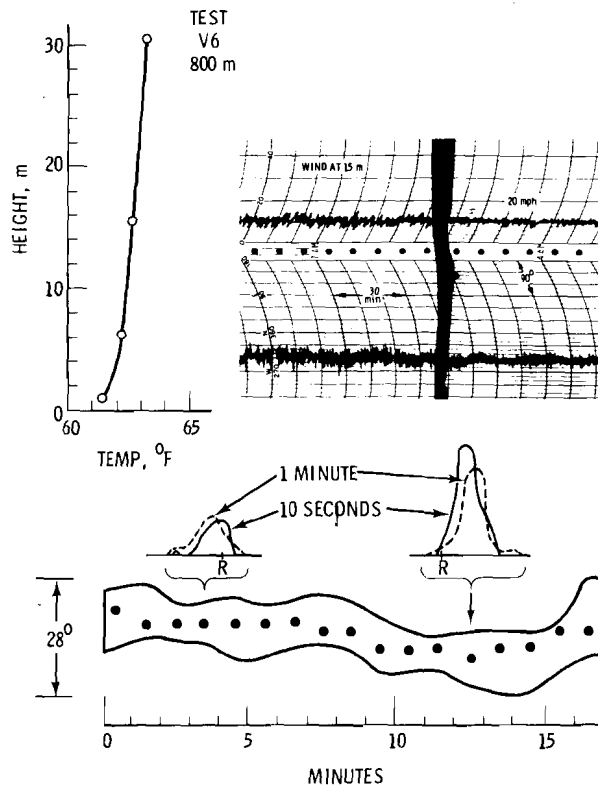
However, the prime point is that a wide range of the relative contribution of real plume diffusion (D^2) to the generally measured apparent plume diffusion (Y^2) is indicated.

TABLE 1. Meteorology and Stability Classification
During Eight ^{85}Kr Releases

Test No.	Release Height (m)	Test Duration (min)	Wind Speed (a) (m/s)	Dir. Range (a) (deg)	Dir. Std. Dev. (b) (deg)	Stability Classification		
						Rich. No. 2 to 15 m	Pasquill Cat. by Reg. Guide 1.23 by σ_θ	by $T_{25}-T_{2m}$
C1	~1	12	2.6	31	4.1	0.13	E	G
C2	~1	13	5.4	62	6.1	-0.04	E	A
C3	~1	10	9.8	77	10.1	-0.03	D	A
C5	~1	17	4.3	52	7.8	0.10	D	E
V2	26	17	2.3	66	~12	-0.03	D	D
V5	26	13	3.8	50	8.2	0.08	D	G
V6	26	24	4.8	47	4.3	0.06	E	F
V7	26	17	4.4	113	~20	-0.29	B	A

a. Aerovane at 15-m elevation. Applies to entire test duration.

b. Beckman and Whitley light-weight vane at 6-m elevation. Derived from 5-sec consecutive readings.



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FIGURE 1. Meteorology and General Character of the Plume During Experiment V6

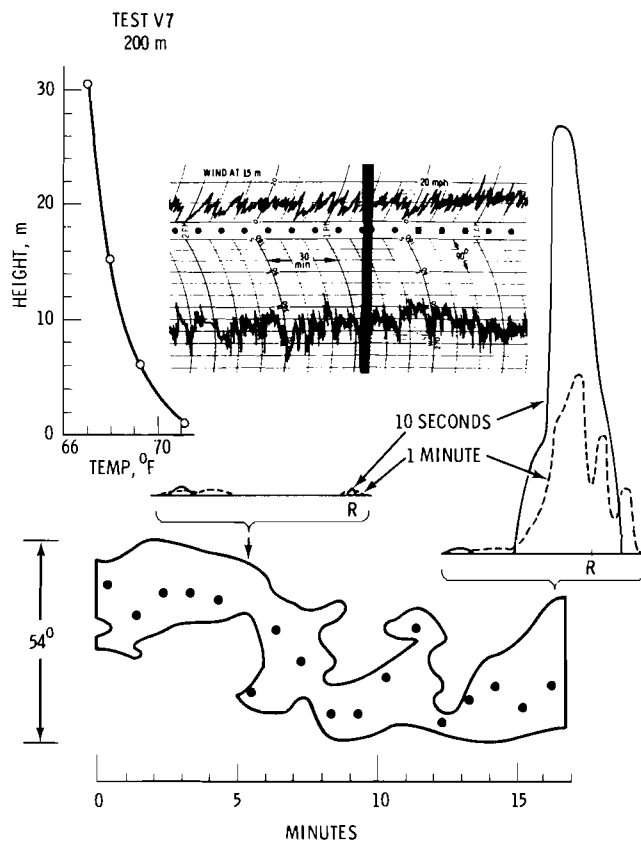
Figure 3 shows the ratio $\overline{D^2}/\overline{Y^2}$ as a function of T/t for measurements made 200 m away from the source. Figure 4 gives the same relationship for a distance 800 m from the source (with one curve for 1600 m also included). The constant T is the total time period during which the apparent plume was examined. The time t is associated with plume segments of shorter duration--segments as short as 38 sec in the "C" tests and 10 sec in the "V" tests. As T/t increases, the ratio $\overline{D^2}/\overline{Y^2}$ is more closely approximated. As seen from Figures 3 and 4, the real plume contributed from about 7% to 83% of the apparent plume diffusion in the current experiments.

Admittedly the relative contribution of the real plume dispersion is enhanced by the fact that these experiments were carried out over periods tending to be meteorologically

stationary. However these data particularly indicate the wide range of effectiveness of the real plume in diffusion at the relatively short end of the time scale.

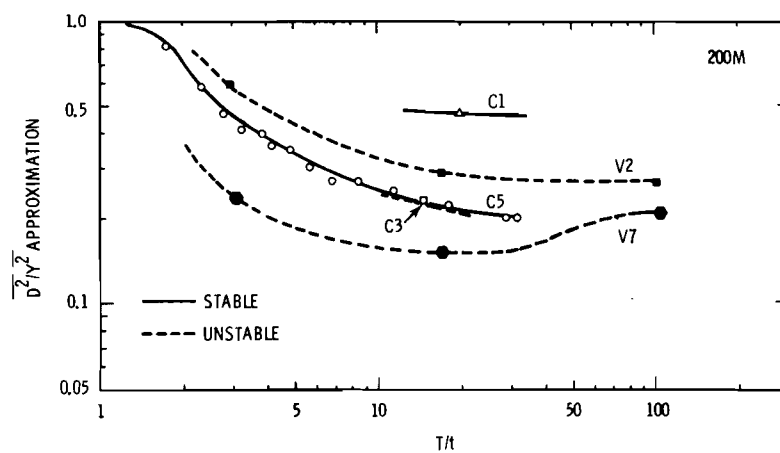
A few comments on specific observations are perhaps in order:

- During four experiments (C1, C3, C5, and V6), $\overline{D^2}/\overline{Y^2}$ could be computed for more than one distance from the source. On three of these tests, the contribution of the real plume dispersion diminished with distance. Surprisingly, on the fourth test (C1), the opposite was true. The contribution of the real plume was greater at 800 m from the source than it had been at 200 m. Meander during this stable-atmosphere release was minimal.



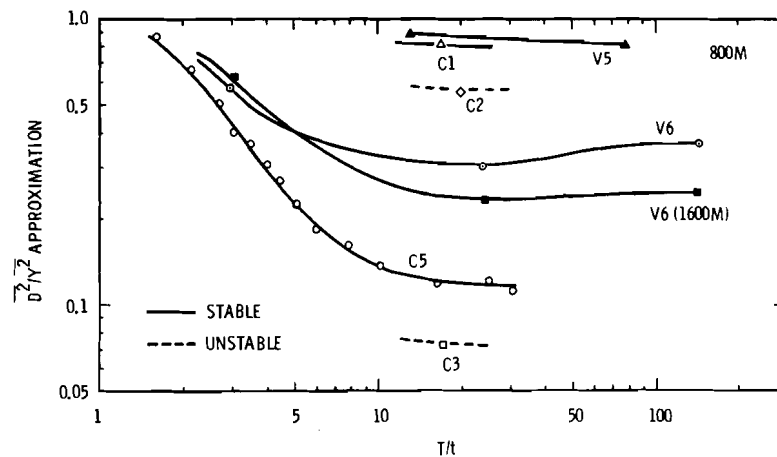
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FIGURE 2. Meteorology and General Character of the Plume During Experiment V7



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FIGURE 3. The Ratio of Diffusion in Short Plume Segments to Diffusion in the Mean or Apparent Plume as a Function of Observation Times at 200 Meters from the Source



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FIGURE 4. The Ratio of Diffusion in Short Plume Segments to Diffusion in the Mean or Apparent Plume as a Function of Observation Times at 800 Meters from the Source

- Stratification by atmospheric stability is not strong. For instance, the minimal meander during neutral to unstable test C2 resulted in a larger $\overline{D^2/Y^2}$ ratio than was observed in stable test V6 despite a larger wind direction range and variance during test C2 (Figure 4 and Table 1).
- The increase in $\overline{D^2/Y^2}$ with increasing T/t displayed on three of the curves in Figures 3 and 4 is very likely a statistical artifact. Only a once-per-minute sampling of the 10-sec duration concentrations was used to compute the largest T/t values in the "V" experiments.

CONCLUSIONS

Although hazards appraisal is generally based on parameters associated with the apparent plume, significant deviations from these mean parameters are caused by the meander and real plume diffusion components of the apparent plume.

The contribution of real plume diffusion to apparent plume diffusion at a distance 800 m from the source varies from less than 10% to more than 80%. No dependence on conventional stability measurements is obvious.

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VARIATION OF PLUME CENTERLINE CONCENTRATION WITH TIME

P. W. Nickola

Real time measurements of plume concentration are used to investigate various plume centerline statistics. Included are the contribution of plume meander to apparent plume centerline concentration, peak-to-mean concentration ratios in the real and apparent plume, and the deviation in interim plume centerline exposure from the exposure finally experienced at the end of plume passage.

INTRODUCTION

The dependence of air pollutant concentrations on the duration of sampling is a recognized concept. Pasquill¹ points out that "...the time over which the plume is 'sampled' changes the spatial distribution of the mean concentration." He further states that "...because of the complex dependence (of wind fluctuation data) on sampling time..., no simple formula or factor can be expected to be universally applicable" in deriving concentration estimates.

A considerable amount of discussion, theory and data are available in the literature relating sampling time to wind direction fluctuations. These wind fluctuations in turn can be related to spread parameters which in turn can be related to predicted or observed concentration measurements.

However, due primarily to the difficulties involved in field measurement, minimal data are available directly relating observed concentrations to sampling time. The ⁸⁵Kr tracer system measurements, alluded to in the immediately preceding contribution* in this report, are trac-

table to investigation of variation of concentration with time.

DATA

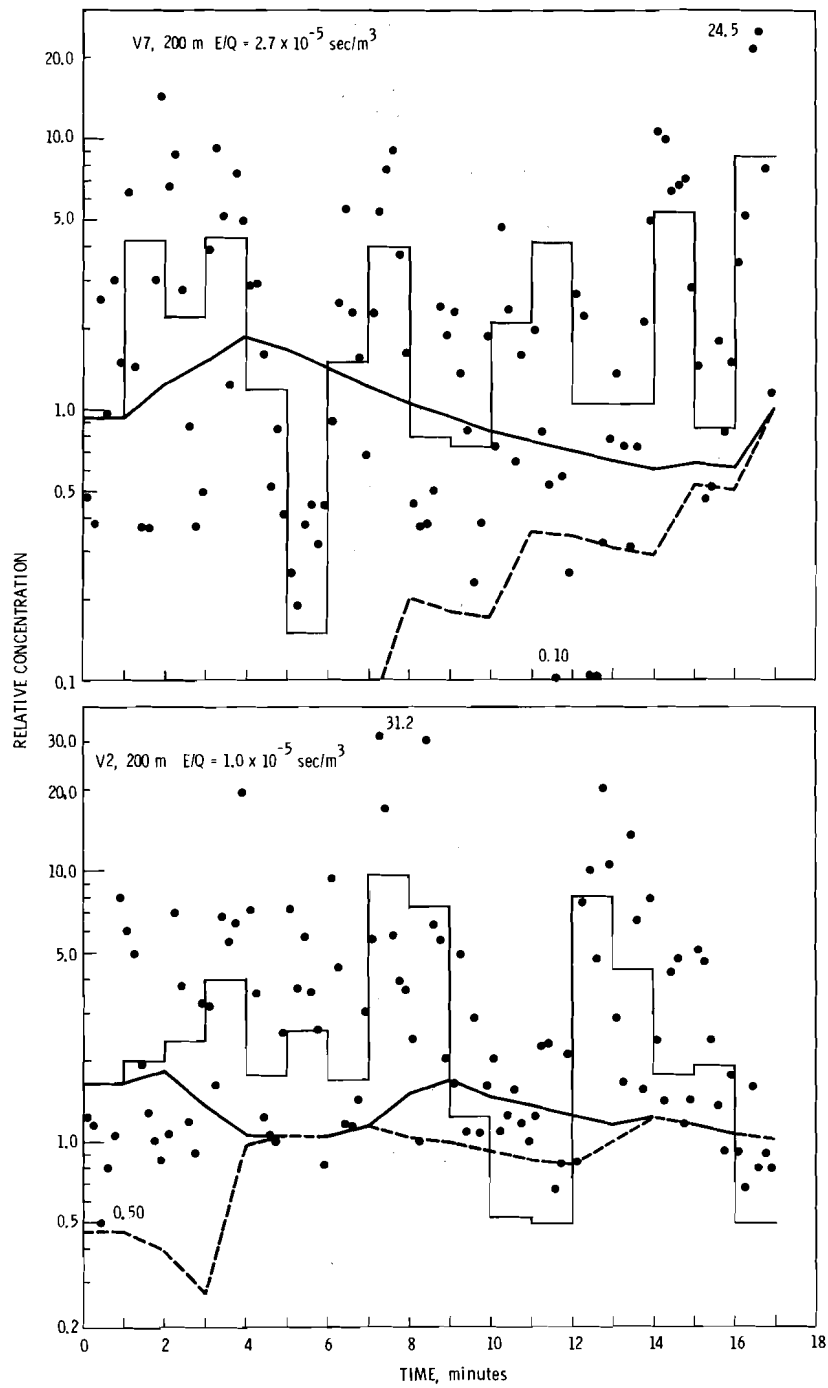
The four "V" tests of the preceding contribution* are examined here. In these field experiments, concentration was measured in a series of 10-sec end-to-end increments. The number of samplers along an arc ranged from 16 to 30. Pertinent meteorological and dispersion information is given in Table 1 and Figures 1 and 2 of the preceding contribution.

Figures 1 and 2 here present concentration measurements versus time for the four experiments. Experiments are presented in order of increasing atmospheric stability. All data plotted on a specific figure result from measurements made at a given distance (listed on each figure) from the source. Tracers were released from an elevation of 26 m, during nearly stationary periods as far as the meteorology was concerned.

Concentrations are specified relative to the final apparent plume centerline exposure (E_p). This time-integrated peak concentration, normalized to amount of tracer emitted, is the value most commonly desired and predicted in hazards appraisals.

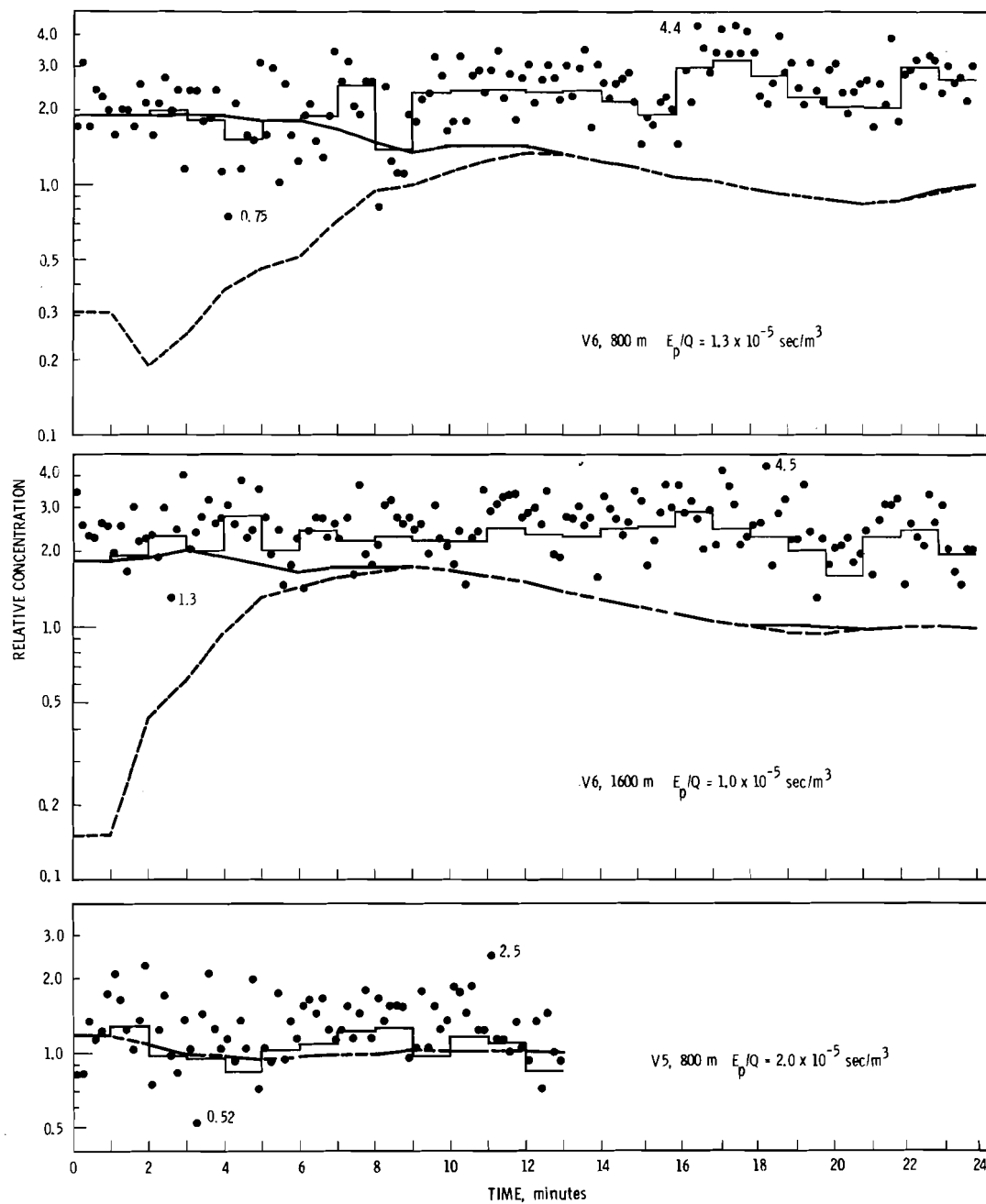
Dots on the figures represent x_p , measured 10-sec end-to-end mean concentrations normalized to emission

* P. W. Nickola, "Some Observations of the Relative Contribution of Real Plume Diffusion and of Meander to Apparent Plume Diffusion," pp.



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FIGURE 1. Concentration Versus Time During Tracer Release in Unstable Atmospheres



Neg 72203-2

FIGURE 2. Concentration Versus Time During Tracer Release in Stable Atmospheres

rate. The values plotted are the highest observed during each time increment--irrespective of azimuth. These concentrations apply to the centerline of the meandering real plume. The bar graphs give similar end-to-end peak concentrations, but for plumes averaged over 1-min periods.

Each broken continuous curve follows an exposure that would have been experienced by a specific "fence post" starting from time zero to the end of any minute. This specific fence post is at E_p , a fixed location on the arc of samplers. Each solid continuous curve gives a history of exposures based on a variety of locations. The momentary exposure selected for plotting is that associated with the fence post (irrespective of location) which, up to a given point in time, has experienced the highest exposure. The curve follows peak exposure as time accumulates. Although the exposures depicted by the broken and by the solid curves could have been derived by summation of 10-sec mean concentrations, they were derived from 1-min means. A perfect correspondence would be observed at the end of each minute in any event.

DISCUSSION

The graphics presented with this report suggest several areas of possible study. (For instance, relationships between stability and variance in real plume centerline could be investigated. Note the much greater variance in the 10-sec peak concentrations plotted for unstable tests V7 and V2 than for stable tests V6 and V5. The ratio of largest to smallest 10-sec centerline concentration observed during unstable V7 was about 250; during stable V5 the corresponding ratio was less than five.) However, a study of relatively wide scope and detail is reserved for a future journal publication. In the brief treatment appropriate for this report, only a few points will be stressed.

The dots on Figures 1 and 2 give essentially the real-time history of concentration of the real plume centerline. The mean of these concentrations ($\bar{x}_p$) gives the E_p that would have been observed had there been no plume meander. The observed

E_p can be compared to $\bar{x}_p$ to prescribe the contribution of meander in the reduction of real plume mean to the time-integrated mean. Table 1 shows that the reduction due to meander, $E_p/\bar{x}_p$, varies from 0.24 to 0.76.

Various "peak-to-mean" ratios can be defined and investigated. Two are defined by the nomenclature given in Table 1 column headings. Perhaps the most interesting is the peak-to-mean observed at the real plume centerline, $x_{pmax}/\bar{x}_p$. In the tests examined, this value varied from more than eight to less than two. The more conventional ratio x_{pmax}/E_p , the highest short-term concentration observed anywhere along the arc compared to the apparent plume centerline concentration, varied from 2.5 to more than 30.

The final discussion here is concerned with the accumulating peak exposure (E_a) depicted by the solid curves in Figures 1 and 2. In a non-meandering plume, a plot of E_a versus time would be congruent with the broken curve plotted for exposure at the location of E_p . In the meandering case, E_a equals x_p at the end of the first averaging increment and equals E_p after the last increment. If it is presumed that E_p is accurately predicted for the entire period of tracer sampling, then the curve E_a indicates the deviation from that prediction that would have been observed had tracer release ceased (or had fence posts been removed) prior to the end of the entire period.

TABLE 1. Peak-to-Mean Concentration Ratios and Reduction in Apparent Plume Centerline Due to Meander ($E_p/\bar{x}_p$)

Test	Distance from Source	Peak-to-Mean Ratios		
		$E_p/\bar{x}_p$	$x_{pmax}/\bar{x}_p$	x_{pmax}/E_p
V7	200 m	0.34	8.3	24.5
V2	200	0.24	7.6	31.2
V6	800	0.42	1.8	4.4
V6	1600	0.38	1.7	4.5
V5	800	0.76	1.9	2.5

After the passage of only 1 min of time, E_a never exceeded $2.1 \times E_p$. In view of the fact that 10-sec and 1-min peak concentrations reached, respectively, magnitudes 31.2 and 9.7 times the E_p value, this relatively low E_a/E_p ratio may be surprising. It also may be comforting to those engaged in forecasting the ~1-hr means commonly required in hazards appraisals.

It is true that E_a is strongly dependent on x_p values during the initial moments of exposure. However, if during test V2 on Figure 1, it is presumed that exposure had started at minute 7--just prior to maximum short period concentrations-- E_a would have been less than 2 at the end of minute 12.

CONCLUSIONS

Based on four field experiments conducted during a wide range of atmospheric stabilities:

- Meander reduces the centerline concentration of the apparent plume to values ranging from 24% to 76% of the centerline concentration of the real plume.
- The peak-to-mean concentration ratio observed at real plume centerline varied from 8.3 during an unstable atmosphere to 1.7 under a reasonably stable atmosphere.
- Although short period fluctuations in concentration are much higher, the exposure experienced between onset of plume passage and any selected time (greater than 1 min) after onset, did not exceed 2.1 times the final apparent plume centerline exposure.

Reference

1. F. Pasquill and F. B. Smith, "The Physical and Meteorological Basis for the Estimation of the Dispersion of Windborne Material," ME-20A, presented at the Second International Union of Air Pollution Prevention Association, Washington, DC, December 6-11, 1970.

FIELD MEASUREMENT OF THE EFFECT OF FILTER
ORIENTATION ON SAMPLING OF A PARTICULATE TRACER

P. W. Nickola

A particulate tracer was sampled by filters facing upward and filters facing toward and away from the mean wind direction. The toward-source samplers proved most efficient while the upward-facing filters displayed lowest efficiencies. Irrespective of efficiency, aerosol sampling under conditions departing significantly from isokinetic may be invalid.

INTRODUCTION

In exposing a filter in the atmosphere to obtain samples of a diffusing tracer or aerosol, the filter usually faces the direction from which the pollutant is expected to arrive. However, there can be advantages to filters facing in other directions. For instance, facing a filter downward could avoid deleterious effects associated with precipitation or large windborne particles of extraneous matter, and be independent of wind direction.

Unfortunately, a truly representative sample of an aerosol-laden air will only be obtained on a filter under isokinetic sampling conditions. These conditions imply that the sampled air is withdrawn parallel to stream flow, that the velocity through the filter is identical to ambient air stream velocity, and that filter holder edge effects do not disturb the aerodynamic streamlines of the approaching air.

Many field experiments conducted at Hanford have entailed the release of the particulate tracer FP 2210*

* FP 2210 is a fluorescent zinc sulfide powder manufactured by U.S. Radium Corp. It has a mass median diameter of about 5 μ m and a specific gravity of 4.1.

to the atmosphere, and subsequent collection of this tracer on filters facing the release point. In this system, embracing hundreds of field sampling locations, it was practical to supply a fixed flow rate to each filter. Isokinetic sampling was a very fortuitous circumstance. Consequently, Sehmel,¹ considering both theory and empirical data, determined corrections to be made following use of "standard" Hanford filters* for collecting FP 2210 under nonisokinetic conditions.

Early experimental work at Hanford had qualitatively confirmed the need for corrections when conditions did not approach the ideal isokinetic flow. Included were comparisons of the masses collected upon field filters which were simultaneously exposed side-by-side during tracer release, but which were oriented in different directions. A more recent quantitative examination of the earlier experiments serves as the basis of this report.

* The standard Hanford assembly employs a 47-mm diameter membrane filter supported by a porous crepe paper backing wound on a central wooden core. The filter surface exposed for tracer collection is about 12 cm².

Since the data presented are from field observations rather than wind tunnel simulation, they are particularly instructive.

FIELD APPROACH

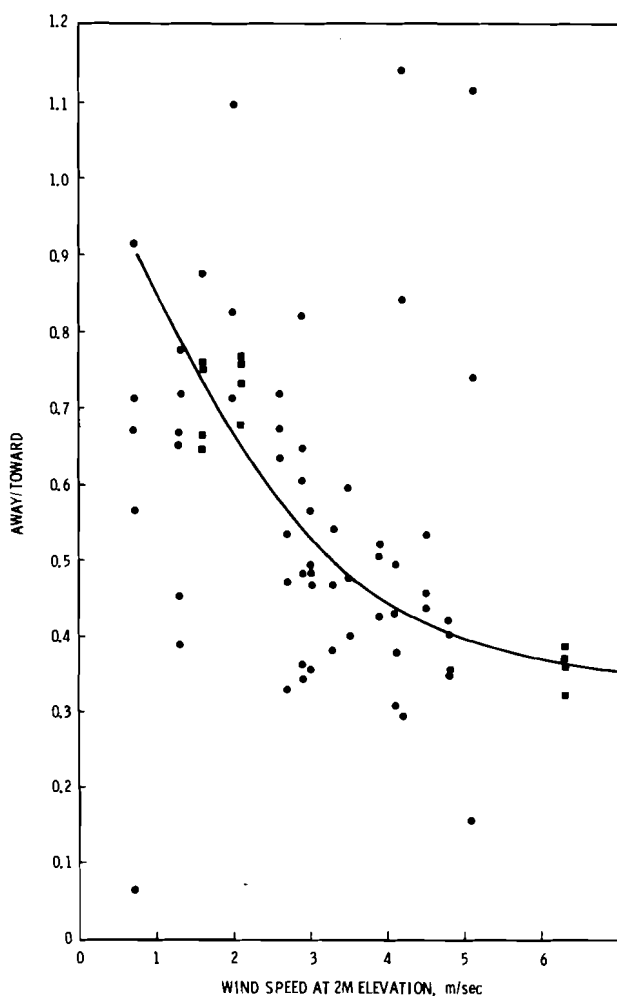
During routine field exposure, Hanford filter samplers on the diffusion grid face the source point. However, during 25 ground-level tracer releases, six additional field locations were fitted with filters that faced vertically upward and filters that faced directly away from the source point. The volumetric flow rate of the additional filters was the same as the routine filter at each location. The "triple-header" sampling stations located 200 m from the source had flow rates of 0.3 cfm. Similarly, the samplers at 800 m sampled at 0.3 cfm; the two triple-headers at 1600 m sampled at 0.5 cfm; and the two at 3200 m sampled at 1 cfm. With the standard Hanford filter, a wind speed of 0.35 m/sec results in isokinetic sampling at a 1-cfm rate. Isokinetic sampling occurs at correspondingly lower wind speeds for the lower flow rates.

During three experiments filters were faced alternately TOWARD and AWAY from the source at arcs 200 m, 800 m, 1600 m and 3200 m from the source. Flow rates remained as indicated in the previous paragraph. This second approach presented an opportunity to examine crosswind profiles based on both the TOWARD and AWAY filters. Although this alternate sampling approach did not offer multiple sampling at any given location, its statistical significance was likely greater due to the larger number of filters involved (up to 26 pairs for one sampling arc).

Figures 1 and 2 show the results of the field exposures. Figure 1 gives, as a function of wind speed, the ratio of mass collected on filters facing AWAY from the source, to mass collected on those facing TOWARD the source; Figure 2 presents the same ratios for the UP to TOWARD filters. Tracer mass was determined by the Rankin counting method.² Dots on these figures represent results of individual filter pairings; the crosses

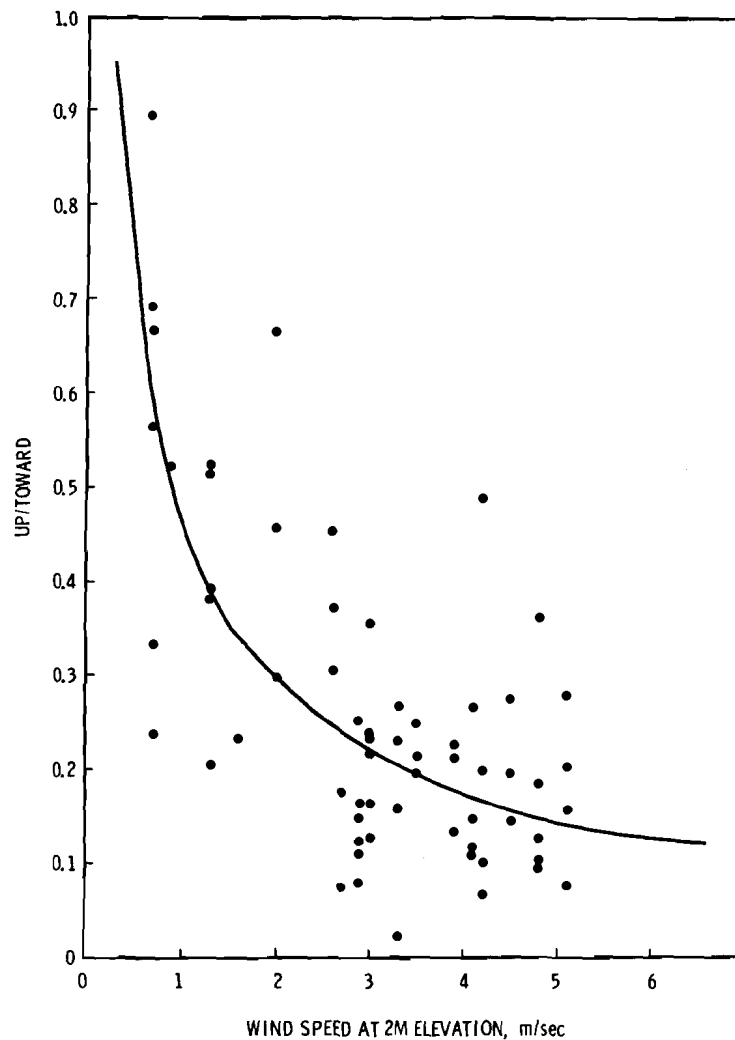
on Figure 1 represent ratios determined from the crosswind profile approach of the immediately preceding paragraph. The curves sketched in are "eyeball" estimates.

There is little doubt that the ratios graphed in Figures 1 and 2 are a function of wind speed. This observation is expected since the TOWARD filters should be most affected by impaction as the wind speed increases above the 0.35 m/sec (or lower) required for isokinetic flow.



Neg 72182-3

FIGURE 1. The Ratio of Tracer Mass on Filters Facing Away from Source to Those Facing Toward Source as a Function of Wind Speed



Neg 72182-4

FIGURE 2. The Ratio of Tracer Mass on Filters Facing Upward to Those Facing Toward Source as a Function of Wind Speed

The figures indicate that at wind speeds above about 0.5 m/sec, the TOWARD filter is the most efficient collector and the UP filter is the least efficient collector. For instance, at a wind speed of 5 m/sec, the AWAY filter collects about 40% of the mass collected by the TOWARD filter, and the UP filter collects only 15% of the TOWARD filter mass.

CONCLUSIONS

Data presented here strictly apply only to FP 2210 collected by the stan-

dard Hanford filter-filter holder assembly. However, for aerosols, the results indicate the large sampling differences possible due merely to filter orientation.

Although the TOWARD filter sampler is a more efficient sampler than the AWAY or UP samplers, it is not necessarily more accurate. As suggested in the introduction, corrections to sampled masses are needed when aerodynamically large aerosols are sampled in any nonisokinetic configuration.

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VARIATION OF SAMPLE WIND COMPONENT SPECTRA AND IMPLICATIONS FOR DIFFUSION MODELING IN THE ATMOSPHERIC BOUNDARY LAYER

D. C. Powell

Comparison of sigma values computed by the Hay-Pasquill equation using 1) sample spectra from analysis of Hanford turbulence time series of not more than 55-min duration, and 2) typical model boundary layer turbulence spectra, shows greater values by a factor of about 1.2 to 1.8 when the sample spectra are used and when the downwind distance of integration is on the order of hundreds of meters. There is also considerable unparameterized variation in the estimates of sigma y from the sample spectra.

INTRODUCTION

In previous Battelle reports^{1,2,3,4} predictions for σ_z , the vertical diffusion of a plume from an elevated source, have been made by integrating the Hay-Pasquill equation⁵ adapted for vertical diffusion. Writing this equation we use the symbol $\sigma_{y,z}$ indicating that it can be used in the same form to formulate either σ_y (the lateral diffusion) or σ_z . The equation may be written as follows:

$$\frac{\sigma_{y,z}}{X} = \frac{u_*}{\bar{U}} \left\{ \int_{-\infty}^{\infty} \left[\frac{n S_{v,w}(n)}{u_*^2} (F) \right] d \left[\ln(F) \right] \right\}^{1/2}, \quad (1)$$

where

$$\theta = \frac{\pi F / (f_m)_{v,w} \beta}{z/X}.$$

In Eq. (1), z is the height of release, X is the downwind travel distance, $\bar{U}$ is the mean wind speed, u_* is the friction velocity (assumed to be the characteristic turbulence velocity), n is frequency, $n S_{v,w}(n)$ refers to the logarithmic power spectrum for either the lateral or the vertical turbulence component, E indicates that the bracketed dimensionless spectrum is in the Eulerian frame of reference, and β is the ratio of the integral scales of turbulence in the Lagrangian and Eulerian frames of reference. The F is a dimensionless frequency best defined by

first introducing another dimensionless frequency of boundary layer turbulence

$$f = nz/\bar{U}. \quad (2)$$

Then $(f_m)_{v,w}$ is the value of f for which $nS_{v,w}(n)$ reaches its maximum value, and

$$F = f/(f_m)_{v,w}. \quad (3)$$

Placing (F) with the bracketed spectrum indicates that the dimensionless spectrum $nS_{v,w}(n)/u_*^2$ will be considered as a universal function of F.

Equation (1) may also be written

$$\frac{\sigma_{y,z}}{X} = \phi \left(\frac{z}{X}, \frac{z}{L}, \frac{z}{z_0} \right) \quad (4)$$

since, according to atmospheric boundary layer theory, all the dimensionless meteorological parameters therein must be functions of z/L and z/z_0 , where L is the Monin-Obukhov stability length and z_0 is the roughness length.

In principal, Eq. (1) exhibits one advantage over the many empirical formulae or graphic sets for evaluating the sigmas in that their dependence on stability and roughness may be shown. But the advantage in practice is open to question. Two reasons are lack of known specification of β and f_m as functions of stability and roughness^{5,6} and restricted application for vertical diffusion due to the vertical shear of the mean wind and the change of turbulence scale in the vertical. A third reason is the unparameterized variation of the sigmas from any ensemble prediction, caused by physical conditions not included in usual boundary layer specification; they may change with time at a given location. An example of unparameterized variation would be terrain effects that affect the meso-scale features of the wind differently when the mean wind blows from different directions. The purpose of this paper is to show the magnitude of unparameterized variation as indicated by integration of sample

power spectra and model power spectra according to Eq. (1), and comparing the results.

DATA AND FORMULATIONS

Among the turbulence tests analyzed from the Hanford measurements made in July 1970, four cases are neutral to slightly stable, i.e., the calculated L was positive and greater than 250 m.⁷ Using data segments of 41 or 55 min from the three heights instrumented by sonic anemometers (7.5 m, 15 m and 60 m) the sample values of u_* , $\bar{U}$, and $nS_{v,w}(n)$ for the numerical integration of Eq. (1) were derived.

The modeled parameters are as follows. The modeled spectra used were^{8,9}

$$\frac{nS_v(n)}{u_*^2} = \frac{2.7F}{(1 + 1.5F)^{5/3}} \quad (5)$$

$$\frac{nS_w(n)}{u_*^2} = \frac{1.1F}{1 + 1.5(F)^{5/3}} \quad (6)$$

with f_m in F [Eq. (3)] given by

$$(f_m)_v = 0.16(1 + 3.0z/L) \quad (7)$$

$$(f_m)_w = 0.47(1 + 2.0z/L) \quad (8)$$

All these formulae and f_m values for neutral conditions were derived either from equations by Kaimal, et al.⁸ or by Busch and Panofsky.⁹ The dependence on stability for f_m is based on the results of Kaimal, et al.⁸ for small positive z/L . Analysis was also made without the stability correction in f_m , and the results were amenable to the same interpretation.

The value of β was calculated according to

$$\beta = 0.35 \bar{U}/u_* \quad (9)$$

For each test, at each height, ratios R_y and R_z were calculated for σ_y and σ_z , respectively, by dividing the results of integration of Eq. (1) using the sample spectrum, into that obtained by integration of the modeled spectrum from Eq. (5) or (6). This can be written

$$R_{y,z} = \frac{\sigma_{y,z} \text{ (sample spectrum)}}{\sigma_{y,z} \text{ (modeled spectrum)}} \quad (10)$$

$z = 7.5, 15, 60 \text{ m},$

$X = 10, 30, 100, 300, 1000 \text{ m}.$

GRAPHIC RESULTS

The average values of R_y and R_z are shown by the broken lines in Figure 1. They are given separately for release heights of 7.5, 15, and 60 m, and continuously for travel distances of 10 to 1000 m for each release height. The standard deviations are shown by the dotted lines. All these ratios apply for neutral stability and for the Hanford site roughness value, $z_0 = 0.03\text{m}.$ ¹⁰

The positions of the s and r designations and the lengths of the lines connecting them to the central broken lines show the change predicted in the ratios when the stability and roughness changes (that we shall call unit changes) are fed into the model. The unit change in stability we shall define as the change from neutral to moderately stable--the change from Pasquill D to Pasquill E.⁵ This change is effected by reading L into the model as 30 m rather than as infinite. The unit change of roughness is defined as half an order of magnitude in z_0 --from 0.03 m to 0.01 m or 0.10 m.

The positions of the s and r designations in Figure 1 indicate that

1. the standard deviation of R_y is well in excess of the change of

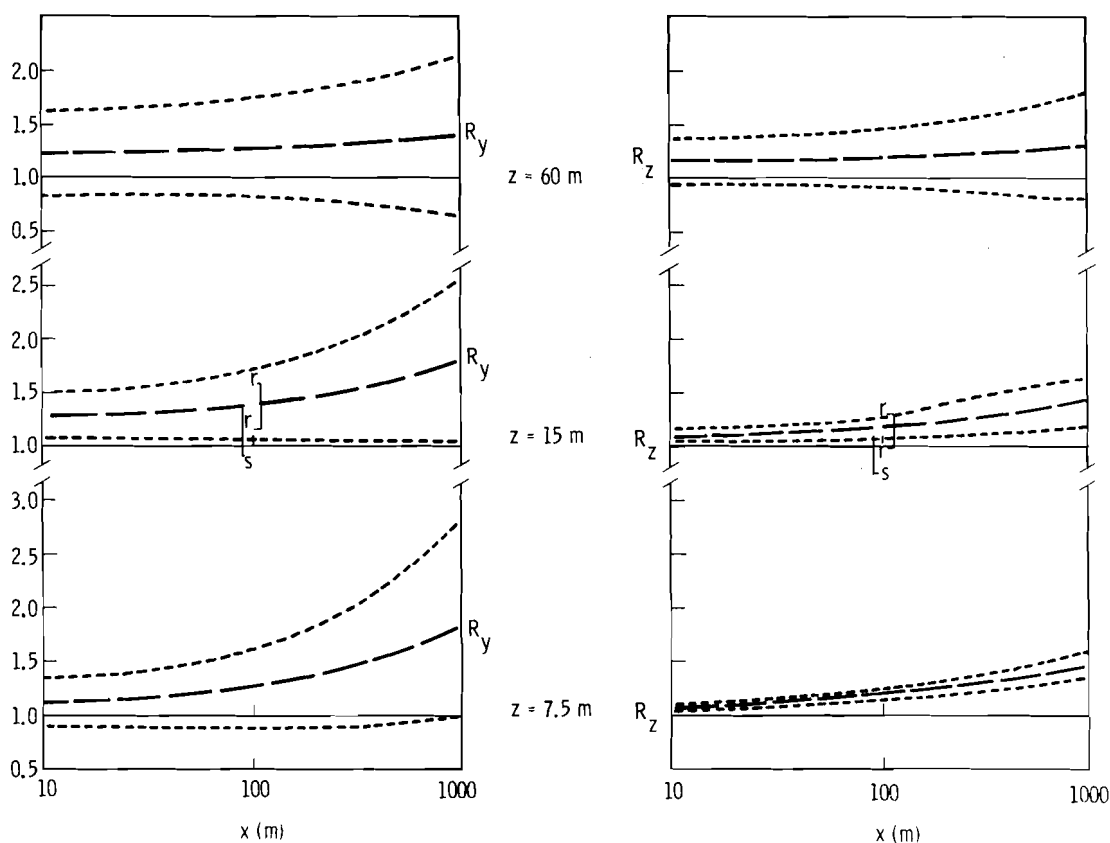
σ_y predicted by the model for a unit roughness change, and roughly equals the change predicted by the model for a unit stability change.

2. the standard deviation of R_z is about equal to the change of σ_z predicted by the model for a unit change of roughness and much less than the change of σ_z predicted for a unit change of stability.

CONCLUSIONS

The following conclusions are offered from inspection of the graphs.

1. The average values of σ_y and σ_z predicted by integration of the sample spectra are in all cases greater than those predicted by integration of the modeled spectra. Moreover the ratios of prediction from sample spectra to prediction from modeled spectra, R_y and R_z , all increase with increasing separation distance to values not greater than two for separation distances not greater than 1000 m. The reason is that when the models are aligned with the sample spectra in the high and middle frequency ranges, the low frequency estimates of the sample spectra (the parts the contributions of which are not filtered out by the trigonometric term in the Hay-Pasquill equation) are generally above the model. Thus the low frequencies are improperly modeled.
2. The uncertainty, i.e., the unparameterized variation of σ_y is much greater than that for σ_z .
3. Tentatively we state that the unparameterized variation of σ_y is large enough to mask the dependence of σ_y on roughness and to some extent on stability. However, the unparameterized variation of σ_z , while not large enough to mask the dependence of σ_z on stability, may do so in the case of small roughness changes.



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FIGURE 1. Average Values for $R_y R_z$ Given at Release Heights of 7.5, 15, and 60 m Plus Standard Deviations

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FURTHER STUDIES ON A LOW LEVEL MODEL TO COMPUTE
MONTGOMERY STREAM FUNCTIONS

W. E. Davis

A previously developed model which computed geostrophic trajectories for Washington and Oregon has been changed, improved, and generalized. The model has been changed to compute more accurate stream functions. Improved to compute gradient wind trajectories, the model can provide more accurately estimate air flow in areas of frontal storms.

The model has been generalized to compute trajectories anywhere sufficient data are available; i.e., data from four radiosonde sites with twelve hourly observations.

MODEL

In order that the model reported by Davis¹ may be used at various locations, a number of changes were made. One of the changes made was that the latitude and longitude of the radiosonde stations were used as input. The maximum and minimum latitude along with the maximum and minimum longitude were used as four corners of a rectangular grid. The assumption was then made that the data at the radiosonde sites had been bilinearly interpolated from the four grid corners. This yielded a set of four equations easily solved for the four grid corners. The data was then linearly interpolated in time to produce a sounding for the hourly reporting stations. The model was run to produce Montgomery stream functions (ψ) as a function of potential temperature at each of the hourly sites. This data can then be used to compute trajectories.

Since the trajectories to be calculated will be near fronts, a decision was made to include in the model a program to compute gradient winds. These winds can then be used for transporting the air parcels. One concern though, is with the error generated in the gradient winds through erroneous stream functions.

Since the model has been changed, it was decided that another error study was necessary.

A series of comparisons were made with the pressures of potential temperature surfaces as derived from generalized model to the values derived from the old model. The result was that the new pressures were usually within a few millibars of the pressures calculated by the old model. While making these comparisons, two problems were found in the model. The first problem was that superadiabatic lapse rates were being artificially produced in the soundings. This was caused in cases where the potential temperature of the first level was less than the surface potential temperature. To correct this a level was found where the potential temperature was greater than surface potential temperature. A linear change was assumed in the potential temperature between the surface and the new level.

The second problem in the model was the maintaining of relative spacing between potential temperature surfaces. (This was used previously when the surface potential temperature minus the potential temperature of the first level was greater than 1° A.) The model now assumes a linear change in potential temperature

between the surface and the first level above surface. With these changes the model was rerun for August and November, 1972. The model's stream functions were compared with those computed from radiosonde data.

RESULTS

As can be seen in Table 1, the data produced by the revised model

are generally more accurate than that from the old model. The main beneficial effect was seen in the 0700 PST data in August for SEA and in the 1300 PST data in November for SEA and for HMS. A decrease in accuracy occurred in the 1300 PST data in November for PDX and in August for SEA. More work will be required to take into account the surface heating effects for 1300 PST data. Since

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

November								
A 0700 PST								
$\Delta\theta$	PDX				SEA			
	New		Old		New		Old	
	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ
1	0	.01	0	.02	.01	.01	.01	.01
2	0	.01	0	.02	0	.01	.01	.01
3	.01	.02	0	.02	0	.02	0	.02
4	.02	.02	.01	.02	0	.03	.01	.03
5	.03	.03	.02	.02	.01	.04	.02	.04
6	.04	.04	.03	.04	.02	.04	.03	.03
B 1300 PST								
$\Delta\theta$	PDX				HMS			
	New		Old		New		Old	
	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ
1	.01	.03	.01	.03	0	.01	.01	.02
2	.01	.02	.01	.01	0	.02	.02	.02
3	.02	.04	.02	.02	.01	.02	.03	.03
4	.03	.05	.02	.03	.02	.02	.03	.04
5	.04	.05	.02	.04	.03	.03	.05	.05
6	.03	.05	.02	.04	.04	.04	.06	.06
November								
B 1300 PST								
$\Delta\theta$	SEA				August			
	New		Old		A 0700 PST		SEA	
	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ
1	0	.02	.02	.04	0	.01	0	.01
2	.01	.02	.03	.04	0	.01	.02	.01
3	.02	.02	.04	.04	.01	.01	.04	.01
4	.03	.03	.05	.04	.02	.02	.06	.02
5	.03	.03	.05	.04	.03	.02	.07	.02
6	.03	.03	.06	.05	.04	.03	.09	.05
August								
B 1300 PST								
$\Delta\theta$	SEA				August			
	New		Old		A 0700 PST		SEA	
	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ	$\bar{x}$	σ
1	.01	.02	0	.02				
2	.03	.02	.02	.03				
3	.04	.03	.02	.03				
4	.06	.03	.02	.04				
5	.08	.04	.02	.05				
6	.11	.03	.01	.05				

changing the model, we are now able to lift the restriction of the computation of stream functions within 100 mb of the surface pressure,¹ while still using the $\Delta\theta^*$ (see Table 1) restriction of 5° A for the 0700 PST data and $2-3^\circ \text{ A}$ for the 1300 PST data.

CONCLUSIONS

The model reported by Davis has been generalized to compute hourly

* $\Delta\theta$ = Potential temperature of a level-surface potential temperature.

values of stream functions anywhere there are four radiosonde stations with at least twelve hourly reports. Also, the output of the model has been gridded and a gradient wind routine developed for use in frontal storms. The model also has been altered to yield generally more accurate values for 0700 PST with a slight decrease in accuracy at 1300 PST. The main result is the lifting of the previous year's restriction on computation of the stream functions within 100 mb of the surface pressure. With the $\Delta\theta$ restrictions listed above, we are able to produce stream functions with errors within $\pm 0.05 \times 10^7 \text{ cm}^2 \text{ sec}^{-2}$.

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DEPOSITION AND RESUSPENSION

Because deposition and resuspension are so important in determining the environmental consequences to man of controlled or inadvertent releases of effluents from nuclear and conventional energy production and from manufacturing, agriculture, transportation, and other industries, PNL has undertaken studies of these processes. The principal objectives of this work are to identify the significant controlling parameters to develop theoretical models, justify theory with laboratory and field experiments, then perform the sensitivity analysis to determine the relative importance of the many independent variables. These studies provide vital input into environmental impact assessment of many sources of released contaminants.

Significant contributions were made in 1975 in both the theoretical and the more practical experimental measurements of deposition and resuspension. Solutions of theoretical deposition-resuspension equations were formulated and nondimensionalized air and ground concentrations were predicted as a function of distance. In other theoretical studies assumptions and analyses regarding surface boundary conditions were investigated and methods presented whereby they can be fitted together within a single theoretical framework. Deposition in vegetation canopies was considered; formulations were developed and conclusions drawn regarding canopy "filtration efficiency." Dry deposition of gases was shown to be rate-limited by many processes, and experiments and equipment were designed to measure gradients of SO₂ and deposition fluxes. A computer model was improved and used to predict downwind concentrations for a generalized area source. A dimensional analysis correlation was formulated from experimental particle deposition velocity data, but was found to show insignificant improvement when compared statistically with an earlier derived correlation. Wind tunnel measurements of deposition velocities to gravel beds and scaled trees showed that particles will penetrate very significantly to underlying surfaces. Initial field experiments measured deposition velocity to sagebrush canopies. Other controlled field studies were initiated for measuring resuspension, including resuspension from truck traffic. Suspension of soil and the size distribution of particles airborne under various air regimes was studied. In the large METROMEX study done near St. Louis, several pollutants were sampled and analyzed as a function of distance. These studies gave insight into the relative importance of dry deposition and atmospheric dispersion as mechanisms for reducing air concentrations.

In the following section an account of these and other studies is given in more detail. These studies have contributed to the understanding of deposition and resuspension, yet much remains to be learned about the fundamental physical processes governing these mechanisms.

- **THEORETICAL STUDIES IN ATMOSPHERIC SCIENCES**
- **PARTICLE RESUSPENSION AND TRANSLOCATION**
- **RADIOISOTOPES AS PARTICLES AND VOLATILES**
- **ATLANTIC RICHFIELD HANFORD COMPANY**

MODELING PRELIMINARIES FOR DRY DEPOSITION TO A CANOPY

W. G. N. Slinn

Approximations are introduced to evaluate a canopy's filtration efficiency. The results are illustrated by plotting the deposition velocity as a function of particle size for various values of the parameter $\gamma = HB/(\lambda\bar{p})$, where $B/\bar{p}$ is the canopy packing density, H is the canopy height, and λ is a characteristic length of the collecting fibers.

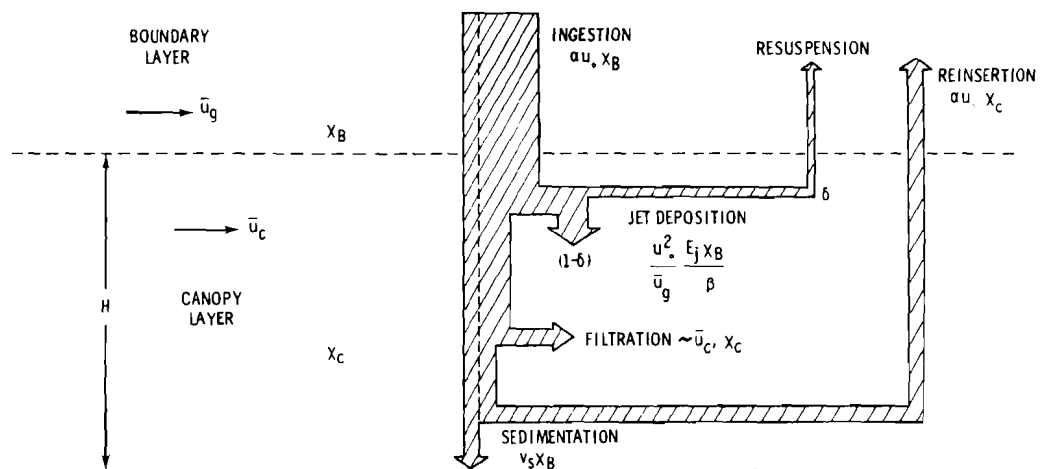
INTRODUCTION

Dry deposition is one mechanism by which pollution is removed from the atmosphere. Typically dry deposition is comparable in effectiveness to precipitation scavenging, although for some pollution sources (e.g., low-level releases) dry deposition is more effective and for others (e.g., stratospheric bomb debris) it is less. Yet the mathematical models of dry deposition lag substantially behind

the wet deposition models. In general, the dry deposition models available apply only to dry deposition on smooth surfaces, or are empirical fits to data. This report demonstrates recent progress¹ to extend the author's dry deposition model² to the case of dry deposition to a canopy.

POLLUTION FLUXES IN A CANOPY

Figure 1 shows the assumed pollution fluxes to, within, and from a



DRY FLUXES OF POLLUTION IN A CANOPY

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FIGURE 1. Assumed Pollutant Fluxes in a Canopy

canopy. From this picture and with obvious assumptions, a steady-state continuity equation can be written as

$$\frac{\partial \chi_C}{\partial x} + \frac{(\alpha u_* + CH)}{\bar{u}_C H} \chi_C = \left(\alpha u_* - \frac{u_*^2 E_j}{\bar{u}_g \beta} \right) \chi_B \quad (1)$$

where α and β are unknown parameters $\mathcal{O}(1)$; u_* is the friction velocity; C is the fraction of the pollutant of a specific (e.g., size) class filtered by the canopy per second; and E_j is the jet collection efficiency for which the author elsewhere² gave the approximation

$$E_j = 10^{-3/St} + \frac{\beta}{\gamma} (Sc)^{-0.6} \quad (2)$$

in which St is the Stokes number based on the viscous length ν/u_* : $St = \tau u_*^2/\nu$ and $Sc = \nu/D$ is the Schmidt number. In these expressions ν is the kinematic viscosity, τ is the particle relaxation time and D is the pollutant's molecular diffusivity.

DEPOSITION VELOCITY

If the concentration of the pollutant in the boundary layer above the canopy, χ_B , is a constant, then Eq. (1) predicts an x -independent concentration in the canopy, χ_C , in distance $\mathcal{O}[\bar{u}_C H/(\alpha u_* + CH)]$, which is typically about 10 canopy heights. Then for x -independent conditions, (1) can easily be solved to give χ_C in terms of χ_B . From this result, the deposition velocity, the net flux to the canopy divided by χ_B , becomes

$$v_d = v_d - \frac{\delta u_*^2 E_j}{\bar{u}_g \beta} + \frac{\alpha u_*}{\alpha u_* + CH} \left[CH + \frac{u_*^2 E_j}{\bar{u}_g \beta} \right] \quad (3)$$

where δ is the fraction of jet-deposited pollutant which is resuspended. An estimate for δ , derived from data obtained by Chamberlain,³ is given elsewhere.²

FILTRATION EFFICIENCY

The major thrust of this report is to present an estimate for the canopy filtration efficiency, C . To do this, it is noted that during a time interval dt the amount of contaminant removed by a single collector of cross-sectional area A is $\bar{u}_C dt A \Xi \chi_C$ where Ξ is a collection efficiency. If the number of collectors, per unit volume, and of cross-sectional Area A to $A + dA$ is $N(A)dA$, the total removal per unit volume during dt is

$$C \chi_C dt = \chi_C \int_{\text{collectors}} \bar{u}_C dt \Xi A N(A) dA. \quad (4)$$

Obviously this integral would be extremely difficult to evaluate for real canopies, the integral being, for example, over all pine needles, leaves, tree trunks, grass blades, etc.

Because of these complexities we propose an approximation for the integral in Eq. (4). For vegetative canopies, the total biomass per unit volume is essentially

$$B = \bar{\rho} \int_{\text{collectors}} \lambda A N(A) dA \quad (5)$$

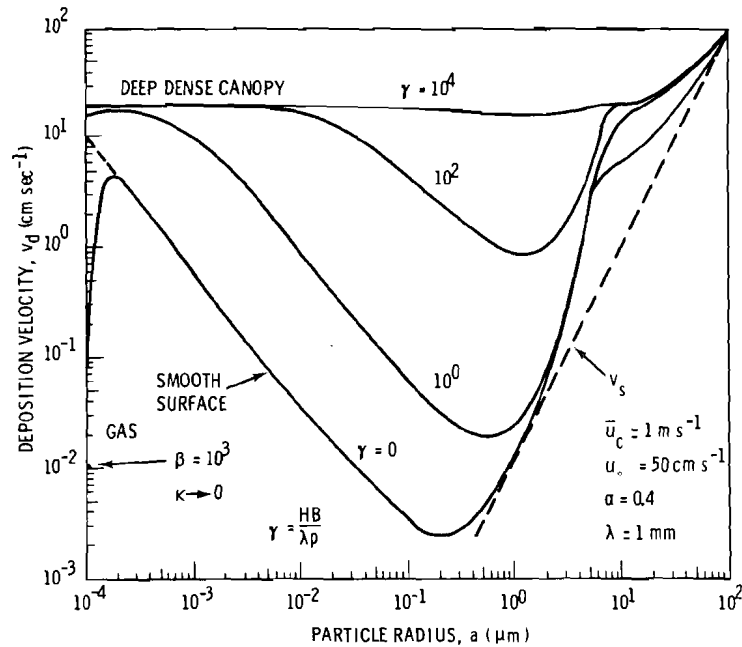
where $\bar{\rho}$ is an average mass density of the foliage and λ is a typical length scale (e.g., radius) of individual fibers. $B/\bar{\rho}$ might be called the canopy filter's packing density. Upon comparing (4) and (5), the removal rate may be approximated by

$$C = \frac{\bar{u}_C B}{\lambda \bar{\rho}} \Xi(a, \lambda) \quad (6)$$

which is to be used in Eq. (3), with a the pollutant particle's radius.

ILLUSTRATION

Figure 2 shows a plot of Eq. (3) with C given by Eq. (6) for the specific set of parameters shown. To evaluate (6) the collection efficiency Ξ was taken to be the same as



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FIGURE 2. A Plot of Eq. (3) with the Removal Rate Given by (6), Demonstrating a Significant Increase in Deposition Velocity for Particles Smaller than About 10 μm , with Increases in Canopy Height, H or Biomass, B . There is a similar increase of v_d with increasing wind speed within the canopy, $\bar{u}_c$. The increase in v_d with decreasing characteristic dimension of the collectors, λ , is even more dramatic because of the concomitant increase in the collection efficiency. At the left-hand side of the plot is qualitatively indicated the possible reduction in v_d for gases because of nonatmospheric effects. This reduction can be significantly less in a canopy (compare the dashed and solid portions of the $\gamma = 10^0$ curve) because of the increased collector area.

for snowflakes,* with the characteristic length $\lambda = 1 \text{ mm}$. The basis for this choice of collection efficiency is the following two considerations: 1) the physical processes governing the collection in both cases (viz., Brownian diffusion, interception, inertial capture, etc.) are the same and, therefore, the collection efficiencies will be similar; and 2) even though the analogy almost certainly

fails in detail, still the general accuracy of this model is so crude as to tolerate inaccuracies in the specification of λ .

RESULTS AND CONCLUSIONS

Sufficient data are not yet available to test this model and to evaluate the parameters α , β , γ and δ . Nevertheless, it can easily be seen from (3) and (6) that the theory is consistent with the following experimental results:

- A linear increase and then saturation of v_d with increasing $\bar{u}_c$ and $H^{3,4}$

* See the contribution by the author entitled "Precipitation Scavenging of Monodisperse Aerosol Particles" in this volume.

- An increase of v_d with roughness height⁵
- An increase of v_d with biomass⁶
- The reduction in v_d caused by resuspension.³

To account for the observed variations⁶ of v_d for gases as a function of humidity and biological activity, a reintroduction of the surface, rate-

limiting arguments is needed.* Such considerations lead to the reduction in v_d for a gas, qualitatively as shown in Figure 2, using the results shown elsewhere* with the β of that theory = 10^3 and $\kappa = 0$.

* See the report by the author entitled "Rate Limiting Aspects of the Dry Deposition of Gases" in this volume, pp. 69-72.

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RATE LIMITING ASPECTS OF THE DRY DEPOSITION OF GASES

W. G. N. Slinn

Results from a simple model are presented which demonstrate that the flux of a pollutant gas from the atmosphere to a surface can be rate limited by a number of processes other than atmospheric diffusion.

INTRODUCTION

As is discussed elsewhere,¹ a neutral atmosphere is capable of delivering the flux of gas to a surface of about $(u_*^2/\bar{U}_1) \chi_1$ where u_* is the friction velocity, $\bar{U}_1$ is the wind speed at 1 m, and χ_1 is the concentration of the gas at this height. However, experimental results indicate that this deposition flux is rarely attained except for very reactive gases such as I_2 . This report presents a simple model which illuminates some of the other possible rate-limiting aspects of the dry deposition process for gases.

CANDIDATE PROCESSES

Many other processes could conceivably be the rate-limiting step in the dry deposition process for gases (if, indeed, a single process dominates). An extreme example of nonatmospheric rate limitation is the case of dry deposition of the noble gases, whose deposition velocity is essentially zero. For most gases, the flux to the ground or to vegetation is rarely limited by the conversion of the gas to a less volatile compound, by diffusion into the ground water or the ground-water's motion, or by passage of the gas through plant membranes. In the case of gas deposition to lakes or oceans for reasonably reactive gases, the atmosphere may be rate-limiting since turbulent mixing in the water body's surface layer may promote the transfer of the gas in these sinks. Liss and Slater's² estimates lead them to conclude that the transport to the ocean of SO_2 , NH_3 , NO_2 , SO_3 and HCl is limited by atmospheric processes, whereas, even to the ocean, the trans-

port of gases such as N_2O , CO , CH_4 , CCl_4 , CCl_3F , MeI and Me_2S is rate-limited by transport in the ocean.

Simple Model

A simple model for dry deposition of gases to a stationary water body (which could be a simulation for soil or plant moisture) can assist toward quantifying the above comments. With obvious approximations and assumptions, the problem is to solve:

$$\frac{\partial \chi}{\partial t} = K \frac{\partial^2 \chi}{\partial z^2} \quad (z \geq 0) \quad (1)$$

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial z^2} - kc \quad (z \leq 0) \quad (2)$$

$$K \frac{\partial \chi}{\partial z} = D \frac{\partial c}{\partial z} \quad (z = 0) \quad (3)$$

$$c(0, t) = H_0 \chi(0, t) \quad (4)$$

where χ is the concentration of the gas in the atmosphere and c its concentration in the water; K and D are diffusivities in the respective regions; k is the rate at which the dissolved gas is irreversibly converted to a nonvolatile product; and H_0 is the overall partition coefficient, the ratio of the total gas in solution (including any ionized component) to the equilibrium air concentration (e.g., see Postma).³

The solution to equations (1)-(4) with the initial conditions $\chi(z, 0) = \chi_0$, $c(z, 0) = 0$ can easily be found using Laplace transform techniques. It is

$$\frac{v_d}{(K/\pi t)^{1/2}} = \frac{\beta^{1/2} e^{-\kappa} - 1}{\beta - 1} + \frac{\beta (\pi \kappa)^{1/2}}{(\beta - 1)^{3/2}}$$

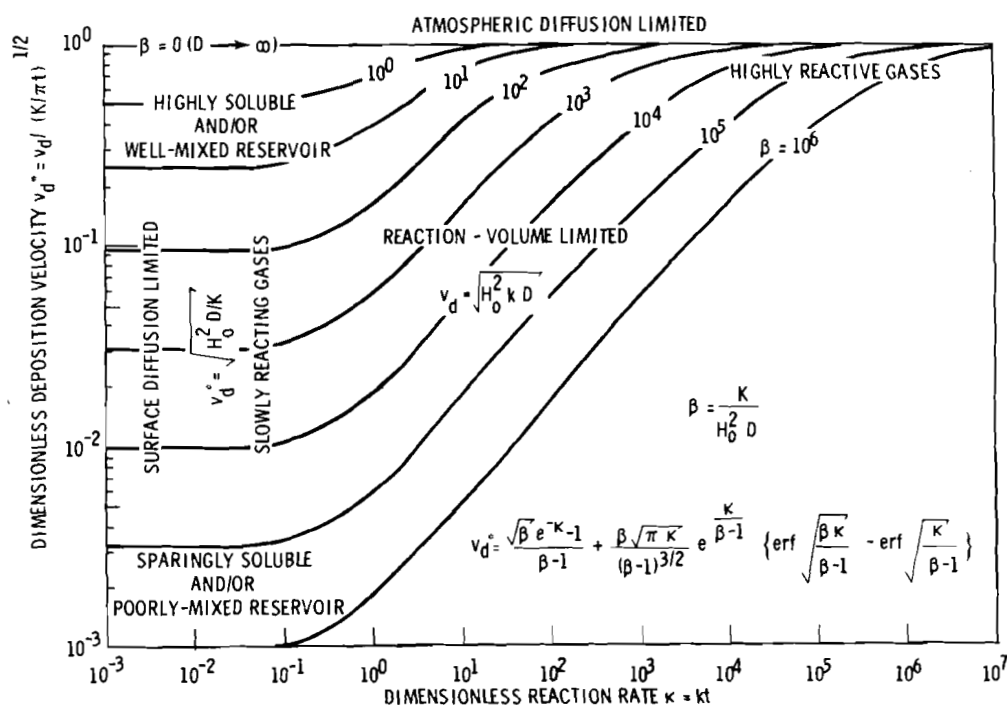
$$\exp \left\{ \frac{\kappa}{(\beta - 1)} \right\} \cdot \left[\operatorname{erf} \left(\frac{\beta \kappa}{\beta - 1} \right)^{1/2} - \operatorname{erf} \left(\frac{\kappa}{\beta - 1} \right)^{1/2} \right] \quad (5)$$

where $\kappa = kt$, $\beta = K/(H_0^2 D)$ and v_d is the ratio of magnitude of the flux to the air concentration x_0 . For $\beta = 1$ the rhs of (5) reduces to $[1 - \{1 -$

$\exp(-\kappa)\}/(2\kappa)]$. For $\beta < 1$, (5) can be written in terms of Dawson's integral.

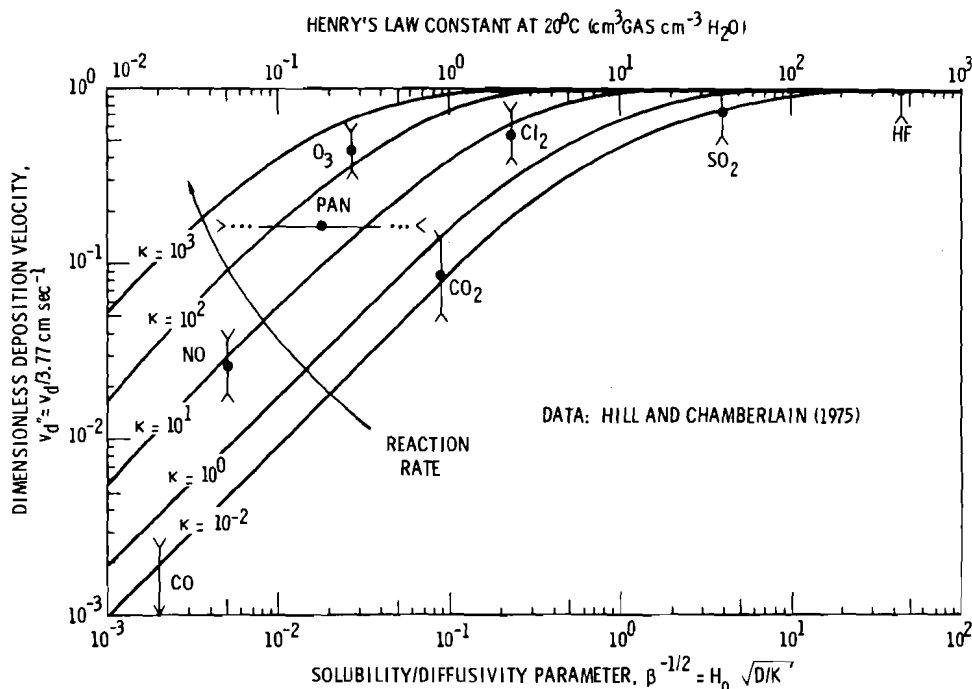
RESULTS AND CONCLUSIONS

Equation (5) is plotted in Figure 1 and demonstrates that the dry deposition velocity for gases can be rate-limited by slow mixing in the ground water [large $\beta = K/(H_0^2 D)$], low solubility (again large β) or by slow conversion to a nonvolatile compound (small κ). The plot of Equation (5) in Figure 2 suggests that the model may have some merit for the interpretation of dry deposition to vegetation.



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FIGURE 1. A Plot of Equation (5) Which Demonstrates that the Dry Deposition Velocity for Gases is Frequently Dictated by Other than Atmospheric Phenomena



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FIGURE 2. A Second Plot of Equation (5), in this Case with $\beta^{-1/2}$ on the Lower Abscissa and the Reaction Rate as a Parameter. For a given diffusivity in the sink, D , and atmospheric diffusivity, K , then $\beta^{-1/2}$ is a constant multiplied by the overall partition coefficient, H_0 , which in turn is proportional to the Henry's law constant. Consequently, by conveniently shifting the upper abscissa and normalizing the deposition velocities as measured by Hill and Chamberlain (4) by their measured value for HF, it can be seen that the theory is capable of reflecting the measured data for the dry deposition of gases to alfalfa. The error bars on the data are subjectively estimated by the author.

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SOME COMMENTS ON THE SURFACE BOUNDARY CONDITIONS FOR THE DIFFUSION-DEPOSITION-RESUSPENSION PROBLEM

W. G. N. Slinn

It is shown how previous analyses and assumptions for the surface boundary condition for the diffusion-deposition-resuspension problem can be fitted together within a single theoretical framework.

INTRODUCTION

If the implied assumptions are acceptable, a pollutant's air concentration, x , is governed by

$$\frac{\partial x}{\partial t} + \vec{v} \cdot \nabla x = \nabla \cdot (K \cdot \nabla x) + v_s \frac{\partial x}{\partial z} \quad (1)$$

where the notation is standard. This note discusses the boundary conditions for (1) (and approximations to it) for the description of deposition and resuspension.

PREVIOUS STUDIES

First, relevant portions of previous studies will be mentioned. For depositing particles, Calder¹ suggested what in thermal physics is familiarly called the "radiation boundary condition":

$$\left[K_z \frac{\partial x}{\partial z} + v_s x \right]_{z=0} = v_d^C x|_{z=0} = v_d^C x_0 \quad (2)$$

which defines Calder's deposition velocity, v_d^C . One of the unsatisfactory features of (2) is that the (other-

wise unspecified) deposition velocity ranges from zero (no flux to the surface and $x_0 \neq 0$) to infinity (perfect sink: finite flux and $x_0 = 0$).

For gases, Heines and Peters² have included the possibility of desorption of a gas from the surface (or, in the terminology to be emphasized here, of "resuspension" from the surface). For the case that Henry's law applies in the absorbing surface medium, their boundary condition simplifies to:

$$K_z \frac{\partial x}{\partial z} \Big|_{z=0} = \frac{k_\lambda}{H} [x_0 - x_{eq}] \quad (3)$$

where k_λ is the mass transfer coefficient in the absorbing medium, H is Henry's law constant and $x_{eq} = Hc_{1b}$ is the air concentration which would be in equilibrium with the actual concentration c_{1b} in the bulk of the absorbing medium.

Recently, for the case of deposition and possible resuspension of particles, Horst et al.³ suggested that in addition to (1) an equation be written for the pollutant's concentration on the surface, G (units m^{-2}). Their proposed equation is

$$\frac{\partial G}{\partial t} = v_d x_o - \Lambda G - \alpha G \quad (4)$$

where Λ is the resuspension rate and α is the rate at which G becomes fixed to the surface and unavailable for resuspension. Concomitantly, (2) is modified to

$$K_z \left. \frac{\partial x}{\partial z} \right|_{z=0} + v_s x_o = v_d x_o - \Lambda G. \quad (5)$$

Notice that the superscript C has been removed from v_d .

This approach does have some desirable features. For example, if the ground concentration reaches a steady state value, then from (4)

$$G_{ss} = v_d x_o / (\alpha + \Lambda) \quad (6)$$

and this in (5) gives

$$K_z \left. \frac{\partial x}{\partial z} \right|_{z=0} + v_s x_o = \frac{\alpha}{\alpha + \Lambda} v_d x_o. \quad (7)$$

Thus in this case the deposition velocity need not have the range from zero to infinity: no flux to the surface can be interpreted as zero fixation rate ($\alpha \rightarrow 0$), infinitely rapid resuspension rate ($\Lambda \rightarrow \infty$) or, see (5), simply as a balance between deposition and resuspension; the surface acting as a perfect sink can be interpreted as $\alpha \rightarrow \infty$. However, there is some difficulty in reconciling these interpretations with time independent G .

But, two other features of this approach urge reconsideration. One is that with negligible deposition, (5) predicts that the surface concentration decays exponentially:

$$G(t) = G(0) \exp \left\{ - \int_0^t [\Lambda(\tau) + \alpha(\tau)] d\tau \right\} \quad (8)$$

in which incidentally, Λ and α are usually taken as constants. Yet data (e.g., Anspaugh et al.⁴) can be fit better with a simple power-law dependence of G on time⁵ suggesting that the governing physical process

is more an increase of volume (in which the "surface concentration" is distributed) rather than just resuspension and fixation. A second phenomenon which may be important is suggested by the experimental results of Rosinski et al.⁶ and Gillette.⁷ It appears that the deposition-resuspension process can frequently change the physical/chemical characteristics of the pollutant: submicron particles can deposit with their characteristic settling speeds, become attached to larger host particles, and then during the resuspension process may or may not dislodge from these host particles whose settling speeds would typically be larger than those of the depositing submicron particles. In the case of some chemicals (e.g., SO_2) one could expect that a substantial portion could be chemically converted (e.g., $SO_2 \rightarrow SO_4^-$) before being resuspended.

THEORY

When phenomena such as these are important, it would appear necessary to discard the above approximations and formulate the problem in a more general manner which, nevertheless, is quite customary. That is, a convective diffusion equation is needed also for the pollutant's concentration, c , in the surface layer:

$$\frac{\partial c}{\partial t} + \vec{w} \cdot \nabla c = \nabla \cdot (D \cdot \nabla c) - L(c) + G(c) + w_s \left. \frac{\partial c}{\partial z} \right|_{z=0} \quad (9)$$

in obvious notation, with L and G some rate of loss and gain of c , respectively, by processes such as conversion to other species, coagulation, breakup, etc. An inviolable interface condition is equality of fluxes:

$$K_z \left. \frac{\partial x}{\partial z} \right|_{z=0} + v_s x_o = D_z \left. \frac{\partial c}{\partial z} \right|_{z=0} + w_s c_o \quad (10)$$

It is less certain how to specify the necessary second boundary condition at the surface; for gases it seems acceptable to assume equilibrium is established at the interface, leading to

$$x_0 = Hc_0 \quad (11)$$

where H is Henry's law constant (a typical example which illustrates this case is given elsewhere*); for particles the concentrations may be continuous (for example $x_0 = C_0$) but more likely are discontinuous (for example, $x_0 = Jc_0$), to ascertain the discontinuity, analysis of the governing physical processes would be necessary.

Alternatively, one can not only use the inviolable equality of fluxes, (10), but also assume an expression for this common value. For example, analogous to Calder's approach, the net flux to the surface could be set equal to $v_d x_0 - v_r C_0$ where v_r is a "resuspension velocity." Of course, this method only postpones the inevitable analysis of the controlling physical phenomena.

To relate this proposal to those made by others, (9) is integrated over all z (< 0). If $\bar{w}$, w_s , D_x , D_y and $G(c)$ are ignored and if $L(c) = \alpha c$, then (9) becomes

$$\frac{\partial G}{\partial t} = \int_{-\infty}^0 \frac{\partial}{\partial z} \left(D_z \frac{\partial c}{\partial z} \right) dz - \alpha G \quad (12)$$

where the average concentration in the surface is

$$G(x, y, t) = \int_{-\infty}^0 c(x, y, z, t) dz \quad (13)$$

The integral in (12) can be evaluated as $D_z (\partial c / \partial z) |_{z=0}$, assuming no flux at $z = -\infty$, and with (10) it is seen how this is related to the flux from the atmosphere. To obtain Horst et al.'s³ expression, (4), this common flux is assumed to be $v_d x_0 - \Lambda G$. To obtain Heines and Peters's⁴ expression, (3), the common flux is set equal to $k_\ell [c_0 - c_\ell b]$. One could also set it equal to $k_a [x_{ab} - x_0]$ or $k_o (x_{ab} - Hc_\ell b)$ where the overall transfer coefficient k_o can easily be seen to be given by $k_o^{-1} = k_a^{-1} + k_\ell^{-1}$.

If c were fairly uniformly mixed to a depth δ , the relation between Λ and the resuspension velocity, v_r , would be approximately, $\Lambda \sim v_r / \delta$. For $\delta = 1$ cm and $\Lambda = 10^{-3} \text{ sec}^{-1}$ during a dust storm, then $v_r \sim 10^{-3} \text{ cm sec}^{-1}$. Similarly, in this case, the resuspension fraction $K = x_0 / G$ could be written as $x_0 / (c\delta)$ and, therefore, if K is determined experimentally, this provides an estimate of the jump discontinuity discussed earlier:

$$x_0 = K\delta c_0.$$

CONCLUSIONS

This analysis has shown how different boundary conditions used by various authors for the diffusion-deposition-resuspension problem can be fit within a single framework; a summary is given in Table 1. Applications of these results are shown elsewhere in this volume.**

* See the report by author entitled: "Rate Limiting Aspects of the Dry Deposition of Gases," in this volume.

** See the report by author, "Approximations and Solutions for the Deposition-Resuspension Problem," this volume, pp. 76-79.

TABLE 1. Parameterization of the Second Interfacial Boundary Condition (the First Boundary Condition Being the Inviolable Equality of Fluxes)

OPTIONS	INTERRELATIONS AND COMMENTS
1. Specification of the jump discontinuity;	
(a) Gases (liquid absorber)	
$x_o = Hc_o$	$H = \text{Henry's law constant}$
(b) Particles	
$x_o = Jc_o$	
$x_o = KG^{(a)}$	$K \approx J/\delta$, resuspension factor ^(b)
2. Specification of the magnitude of the common Flux, F:	
(a) Gases (liquid absorber)	
$F = k_\ell(c_o - c_{\ell b})$	$k_\ell \approx D_z/\delta$
$F = k_a(x_{ab} - x_o)$	$k_a \approx K_z/H$
$F = k_o(x_{ab} - Hc_{\ell b})$	$k_o^{-1} = k_a^{-1} + Hk_\ell^{-1}$
(b) Particles	
$F = v_d^c x_o$	$\infty > v_d^c > ?$
$F = v_d x_o - v_r c_o$	$v_r = \text{resuspension velocity}$
$F = v_d x_o - \Lambda G$	$\Lambda \approx v_r/\delta$, resuspension rate
$F = \Omega A - \Lambda G^{(a)}$	$\Omega \approx v_d/H$, deposition rate ^(b)
a. A and G are z-averaged x and c , respectively.	
b. δ and H are approximate mixed depths in the surface and atmosphere, respectively.	

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APPROXIMATIONS AND SOLUTIONS FOR THE DEPOSITION-
RESUSPENSION PROBLEM

W. G. N. Slinn

Some simple solutions of the deposition-resuspension equations are obtained for the z-averaged contaminant concentration in the ground and in the atmosphere.

INTRODUCTION

The complete formulation of the diffusion-deposition-resuspension problem is rather formidable:

$$\frac{\partial \chi}{\partial t} + \vec{v} \cdot \nabla \chi = \nabla \cdot (K \nabla \chi) + v_s \frac{\partial \chi}{\partial z} \quad (1)$$

$$\begin{aligned} \frac{\partial c}{\partial t} + \vec{w} \cdot \nabla c &= \nabla \cdot (D \nabla c) + G(c) - L(c) \\ &+ w_s \frac{\partial c}{\partial z} \end{aligned} \quad (2)$$

where χ is the pollutant's concentration in the atmosphere and c its concentration in the surface. Other notation is standard. To complete the specification of the problem, suitable initial and boundary conditions are needed and these will be introduced later. This report discusses solutions to approximate versions of (1) and (2), which may be used to assist analyses of the complete problem.

AVERAGES

To obtain an approximate description of the physical processes, (1) and (2) are averaged over the crosswind (y) and vertical (z) directions. Further, if x -diffusion in the atmosphere is ignored as well as both x -diffusion and all transport in the surface, then (1) and (2) become

$$\begin{aligned} \frac{\partial A}{\partial t} + \bar{u} \frac{\partial A}{\partial x} &= - \int_{-\infty}^{+\infty} dy \\ \left[K_z \frac{\partial \chi}{\partial x} + v_s \chi \right]_{z=0} & \end{aligned} \quad (3)$$

$$\frac{\partial G}{\partial t} = \int_{-\infty}^{+\infty} dy D_z \frac{\partial c}{\partial z} \Big|_{z=0} - \alpha G \quad (4)$$

where A is the crosswind and vertical integral of the contaminant in the atmosphere,

$$\begin{aligned} A(x, t) &= \int_{-\infty}^{+\infty} dy \int_0^{\infty} dz \chi \\ (x, y, z, t) & \end{aligned} \quad (5)$$

and similarly for G , the ground concentration. To obtain (4) it has been assumed that $G(c) = 0$ and the loss term, $L(c) = -\alpha c$ where α is the rate at which c is attached to the surface, thereby becoming unavailable for resuspension.

BOUNDARY CONDITIONS

The boundary conditions can be used to obtain some information about the integral terms in (3) and (4). Equality of fluxes states

$$\begin{aligned} - \left[K_z \frac{\partial \chi}{\partial z} + v_s \chi \right]_{z=0} &= - \left[D_z \frac{\partial c}{\partial z} \right]_{z=0} \end{aligned} \quad (6)$$

which can be integrated over y . Now, extending the method of Horst et al.,¹ we assume that these y -averaged fluxes are given by $\Lambda G - \Omega A$ where Λ is a

resuspension rate and, similarly, Ω is a deposition rate. Then (3) and (4) become

$$\frac{\partial A}{\partial t} + \bar{u} \frac{\partial A}{\partial x} = \Lambda G - \Omega A \quad (7)$$

$$\frac{\partial G}{\partial \tau} = \Omega A - \Lambda G - \alpha G \quad (8)$$

PARAMETER ESTIMATES

Horst et al.¹ reference experimental data for the resuspension rate, Λ , in the range of 10^{-7} to 5×10^{-9} sec^{-1} and for the fixation rate, α , they suggest a value of about 10^{-7} sec^{-1} . However, this author questions if such values adequately reflect reality. Even a casual observation of the resuspension that occurs during a dust storm suggests that the actual process has a time constant on the order of hours (rather than years). The experimental data that lead to $\Lambda \sim 10^{-8}$ sec^{-1} may better reflect the time constant of the sampling process ($\sim$ months) rather than the time constant of the physical process. Of course if only a few high wind situations occur per year, then a long-time average of Λ will lead to a value such as $\Lambda \sim 10^{-8}$ sec . Here, however, an approximate analysis will be performed to estimate the air concentration during time periods comparable to the duration of an individual dust storm and, therefore, we take (with only casual observations as the experimental basis) $\Lambda \sim \Omega$. In turn, the deposition rate can be estimated to be $\Omega \approx v_d/H$, where H is the height of the mixed layer. Typical values for the deposition velocity are $1 \lesssim \Omega \lesssim 10$ days. The rate α is assumed to be slow enough to be ignored during the time scale of interest.

SOLUTIONS

Equations (7) and (8) are vastly simplified versions of (1) and (2) but still are difficult to solve analytically. The steady-state solution

$$A(x) = A(0) - \frac{\alpha}{\bar{u}} \int_0^x G(\xi) d\xi \quad (9)$$

does not seem to be of much practical value. If $G(x)$ can be considered time independent then the steady-state solution for A is

$$A(x) = \exp \left\{ - \frac{\Omega x}{\bar{u}} \right\} \left[A(0) + \frac{\Lambda}{\bar{u}} \int_0^x e^{\Omega \xi / \bar{u}} G(\xi) d\xi \right] \quad (10)$$

which is similar to the case studied by Horst² who, however, includes x-diffusion.

To solve (7) and (8) in the general case, subject to the boundary and initial conditions $A(x=0, t) = 0$, $A(x, t=0) = 0$ and $G(x, 0) = Q_0 \delta(x - x_0)$, Laplace transforms in t (parameter s) and x (parameter p) are taken of (7) and (8). This leads to

$$\bar{A}(p, s) = \frac{\Lambda Q_0 e^{-px_0}}{(s + \Lambda + \alpha)(s + p\bar{u} + \Omega - \Omega\Lambda)} \quad (11)$$

Inverting the p-Laplace transform gives

$$\bar{A}(x, s) = \frac{\Lambda Q_0}{\bar{u}} \frac{1}{(s + \Lambda + \alpha)} \exp \left\{ - \frac{(x - x_0)}{\bar{u}} \frac{[s^2 + s(\Lambda + \alpha + \Omega) + \alpha\Omega]}{(s + \Lambda + \alpha)} \right\} \quad (12)$$

This last expression is difficult to invert because of the essential singularity at $s = -(\Lambda + \alpha)$. Evaluating the residue and recognizing the sum as the expansion of a zeroth-order, modified Bessel function finally yields the solution

$$A(x, t; x_0, t=0) = \frac{\Lambda Q_0}{\bar{u}} e^{-\Omega \tau} \exp \left\{ - (\Omega + \alpha)(t - \tau) \right\} \cdot I_0 \left[2 \{ \Omega \Lambda \tau (t - \tau) \}^{1/2} \right] \quad (13)$$

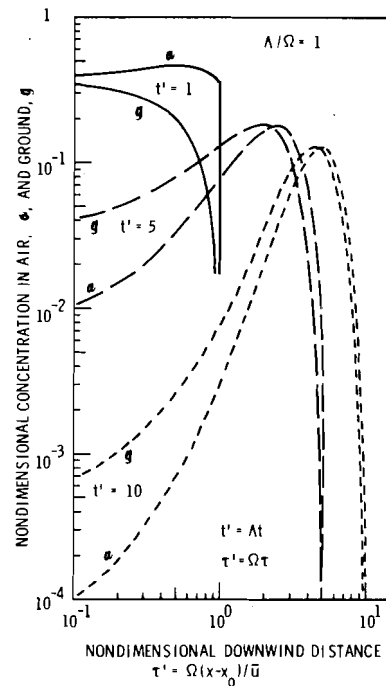
in which $\tau = (x - x_0)/\bar{u}$ and it is required that $\tau < t$ or $(x - x_0) < \bar{u}t$, whose physical significance is obvious. From (13), which is essentially the Green's function of the problem, the solution for an arbitrary initial distribution of G can be found easily. The solution for $G(x, t)$ is given elsewhere.³

CONCLUSIONS

Figures 1-3 illustrate the solution to the z -averaged equations for nondimensionalized A and G . The complexity of this solution for this case foretells of serious analytical barriers to obtaining analytical solutions to Eq. (1) and (2). Numerical explorations would appear to be warranted. If a numerical approach is taken, then we hope the analytical results obtained here will provide a useful check on the numerical techniques used. Further, the formalism presented here may strengthen the foundations of future analyses.

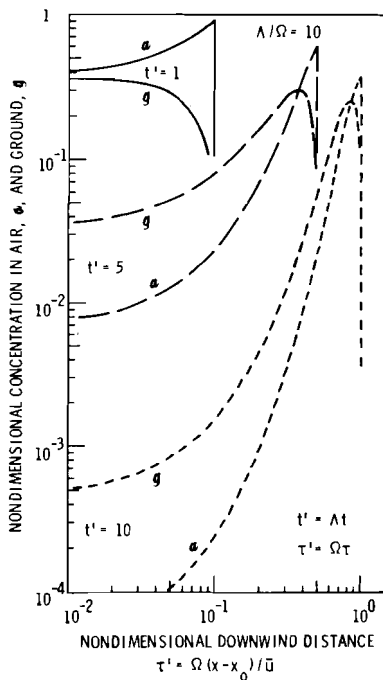
ACKNOWLEDGEMENTS

The author thanks Tom Horst for stimulating discussions.



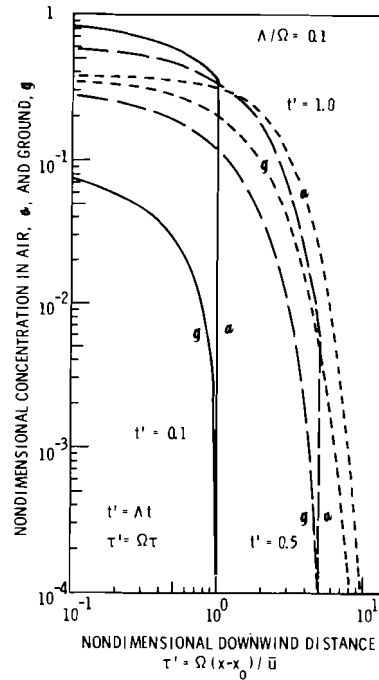
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FIGURE 2. A Plot of the Nondimensionalized Air and Ground Concentrations for Equal Rates of Resuspension and Deposition



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FIGURE 1. A Plot of the Nondimensionalized Air and Ground Concentrations for Resuspension Rapid Compared with Deposition



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FIGURE 3. A Plot of the Nondimensionalized Air and Ground Concentrations for Deposition Rapid Compared with Resuspension

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RESUSPENSION FROM A GENERALIZED AREA SOURCE

T. W. Horst

Approximate equations are given for the dependence of the crosswind-integrated resuspended air concentration on resuspension rate, wind speed, deposition velocity and source geometry. These are shown to compare very well with numerical solutions of the problem and are used to discuss the usefulness of the resuspension factor.

INTRODUCTION

The air concentration of material resuspended from an area source is a complicated function of the current meteorology, source geometry and source history. An investigation of the first two variables was begun¹ by studying a uniform field of resuspendable material of infinite extent in the crosswind direction. These results have now been extended to the case of a general distribution in the downwind direction.

A distribution of surface contamination $G(x)$ which depends only on the downwind coordinate x and extends infinitely in the crosswind direction is assumed to resuspend at a rate Λ and redeposit with a deposition velocity v_d from the resulting air concentration C . The relevant coordinates are the height z above the ground and

the downwind distance x from the upwind edge of the contaminated area.

UNIFORM AREA SOURCE

Using the surface flux model,² the air concentration is

$$C(x, z) = \int_0^x \left[\Lambda G(\xi) - v_d C(\xi, z_d) \right] D(x - \xi, z) d\xi \quad (1)$$

Here z_d is the reference height for deposition and $D(x, z)$ describes the vertical diffusion from a ground-level, crosswind infinite line source of nondepositing material,

$$D(x, z) = \frac{2}{\sqrt{2\pi} \sigma_z u} \exp \left(-z^2 / 2\sigma_z^2 \right) \quad (2)$$

where u is the mean wind speed and the dispersion parameter $\sigma_z(x)$ is calculated from the formulas of Briggs.³

If for resuspension from a uniform area source without deposition we define

$$\psi_0(x, z) = u C(x, z) / AG, \quad (4)$$

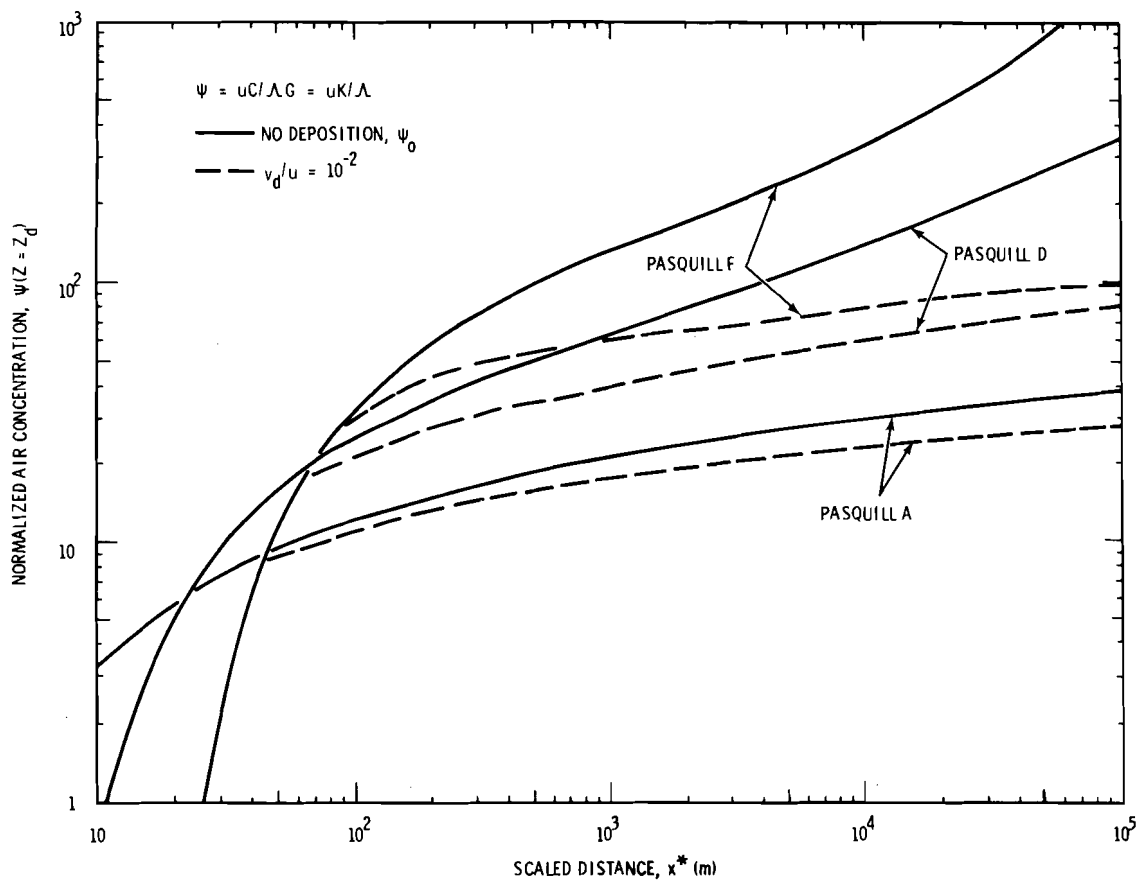
then Horst has shown¹ that with deposition

$$\psi(x, z) \approx \frac{\psi_0(x, z)}{1 + \frac{v_d}{u} \psi_0(x, z_d)} \quad (5)$$

This equation follows from an analysis of (1), utilizing the basic properties of the diffusion function D , and is verified by numerical solutions of (1). The approximation is best for small z since D then has its peak value near the source. Figure 1 shows ψ , as calculated numerically at $z = z_d = 1$ m, for three different atmospheric stability categories, for no deposition and for $v_d/u = 10^{-2}$.

INCREASING SOURCE

If we consider now a surface contamination $G(x)$ which increases in the downwind direction, it can be shown from (1) that the above results can be generalized by introducing a scaling distance



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FIGURE 1. Resuspended Air Concentration for a Uniform Area Source

$$x^* \equiv \frac{1}{G(x)} \int_0^x G(\xi) d\xi. \quad (6)$$

Then we have

$$\psi(x, z) \equiv \frac{u C(x, z)}{\Lambda G(x)} \approx \frac{\psi_0(x^*, z)}{1 + \frac{v_d}{u} \psi_0(x^*, z_d)} \quad (7)$$

This approximation has been verified by numerical solution of (1) for several different $G(x)$ distributions and is also best for small z .

GENERAL SOURCE

Finally, for a real source which increases to a maximum at $x = x_0$ and then decreases, these results may be applied by defining

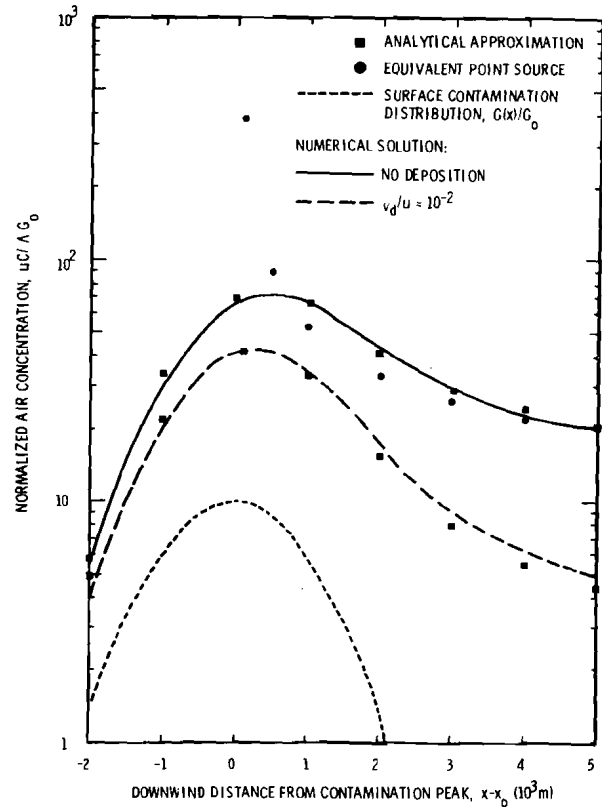
$$G_1(x) = \begin{cases} G(x), & x < x_0 \\ G(x_0), & x > x_0 \end{cases}$$

$$G_2(x) = \begin{cases} 0, & x < x_0 \\ G(x_0) - G(x), & x > x_0 \end{cases} \quad (8)$$

These distributions are each monotonically increasing in the downwind direction and their difference is equal to $G(x)$. Hence, applying (7),

$$C(x, z) \approx \frac{\Lambda}{u} \left\{ \frac{G_1(x) \psi_0(x_1^*, z)}{1 + \frac{v_d}{u} \psi_0(x_1^*, z_d)} - \frac{G_2(x) \psi_0(x_2^*, z)}{1 + \frac{v_d}{u} \psi_0(x_2^*, z_d)} \right\}. \quad (9)$$

Figure 2 is an example of the application of (9) to a Gaussian distribution of surface contamination which is peaked at x_0 and which has a standard deviation of 1 km. The air concentration, normalized by the peak surface contamination, is shown as calculated exactly with (1) for the



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FIGURE 2. Resuspended Air Concentration for a Gaussian Surface Source

case of no deposition (solid curve) and for the case of $v_d/u = 10^{-2}$ (broken curve). The dotted line simply gives the shape of the surface contamination distribution. The predictions of (9) are plotted as squares and agree with the exact solution within 10% to 20%. Also shown, as dotted data points, are the predicted air concentrations for an equivalent point source of nondepositing material located at $x = x_0$. Note the close match with the Gaussian source several standard deviations downwind of the peak.

HORIZONTAL FLUX

The total horizontal flux of material in the downwind direction,

$$F_H = \int_0^\infty u C(x, z) dz \quad (10)$$

can be approximated with (7) to be

$$F_H \approx \frac{\Lambda G(x) \int_0^\infty \psi_0(x^*, z) dz}{1 + \frac{v_d}{u} \psi_0(x^*, z_d)} \quad (11)$$

Since the numerator applies also for the case of no deposition, conservation of material requires that

$$F_H \approx \frac{\Lambda G(x) x^*}{1 + \frac{v_d}{u} \psi_0(x^*, z_d)} \quad (12)$$

This also follows from a direct application of (1). The extension of (12) to the general source as evaluated with (8) should be obvious.

In obtaining (11), (7) was assumed to be applicable at all heights, whereas in fact it is best for small z and becomes less valid as z increases. However, due to the vertical distribution of ψ_0 , the major contribution to F_H is near $z = 0$, i.e., the bulk of the flux is carried near the surface, and (11) is a fairly good approximation. This is again verified by numerical calculations of (10).

THE RESUSPENSION FACTOR

In order to estimate resuspended air concentrations, much use has been made in the past of the resuspension factor $K \equiv C/G$. This concept does not account for the variations in C due to source configuration or the dependence of diffusion on source-receptor separation. Hence measured values of K can be used with confidence to estimate C in new situations only when K is known to vary little with location relative to the contamination distribution $G(x)$. The results outlined above may be used to comment more specifically on the variation of K .

The vertical separation of the air concentration curves from the surface contamination curve in Figure 2 is proportional to the resuspension fac-

tor. K increases slowly with increasing x upwind of the peak and then increases very rapidly downwind of the peak. In general the resuspension factor has little use downwind of a contaminated area because C is controlled by the upwind contamination rather than the local contamination. Note that the inclusion of deposition increases somewhat the linkage of C with the local value of G . A large deposition velocity decreases the downwind influence of the surface contamination on the air concentration.

Equation (7) directly predicts the value of K upwind of the contamination peak. Figure 1 shows ψ , or K in units of Λ/u , at the deposition reference height of 1 m. For $x^* \gtrsim 100$ m, ψ increases rapidly with x^* ; between 100 m and 10 km, ψ_0 increases least rapidly; and beyond 10 km ψ_0 begins to increase more rapidly again as the vertical mixing reaches a limiting value, especially for the stable Pasquill F. The unstable Pasquill A increases least rapidly because the strong vertical mixing of the contaminant limits its downwind influence. As observed previously, deposition also limits the downwind influence of the contaminant, reducing the variation of the resuspension factor with location. From (7) it can be seen that at large x^* , and hence also ψ_0 , K with deposition approaches a limiting value Λ/v_d .

The influence on K of the rate of change of G with x can be investigated by representing the surface distribution as a power function, $G(x) \sim x^n$. Then the scaling distance is simply $x^* = x/(n+1)$, and ψ can be plotted as a function of x in Figure 1 by shifting the curves to the right by the factor $(n+1)$. It is then apparent that, at a given value of x , the G with the largest n also has the least constant resuspension factor. The only exception is for the very large values of x^* where the slope of ψ in Figure 1 increases with x^* .

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COMPARISON OF DEPOSITION VELOCITIES PREDICTED FROM DIMENSIONLESS ANALYSIS CORRELATIONS

G. A. Sehmel

Experimental particle deposition velocity data used for determining the initial surface mass transfer resistance correlation were used to develop a second correlation. Statistical tests suggest both correlations fit equally well. Additional data are needed as a data base since extrapolations from the two correlations can show wide variations in the predicted deposition velocities.

INTRODUCTION

Particle deposition velocities are a function of many variables including surface, particle, and meteorological parameters. Limited deposition velocity measurements in the field have never been correlated into any predictive model. Possibly no field-based predictive model exists because experimental conditions in the field cannot be controlled. However, deposition velocities can be measured in controlled experiments in wind tunnels. Results from wind tunnel experiments have been correlated into an initial predictive model. The present objective was to determine if an improved predictive model could be developed based upon a second modeling approach.

MODELS

A particle deposition velocity, K , can be described¹⁻³ by the equation,

$$K = \frac{v_t}{1 - (1/\alpha)}, \quad (1)$$

where v_t is the settling velocity for a monodispersed particle. This equation indicates the lower limit of deposition velocities to be the gravity settling velocity. However, as v_t approaches zero, mass transfer is controlled by Brownian and eddy diffusion. The term α is a model-derived expression containing a grouping of mass transfer resistance:

$$\alpha = \exp(-v_t \text{Int}/u_*), \quad (2)$$

where u_* is the friction velocity and Int is an integral mass transfer resistance term. Obviously, Int is related to a simple resistance R by,

$$R = \frac{1}{K} = \frac{1 - \exp\left(\frac{v_t}{u_*} \text{Int}\right)}{v_t} \quad (3)$$

The initial modeling approach² was to correlate Int as a function of dimensionless groupings. In this case, Int included only the mass transfer resistance within the closest 1 cm adjacent to the deposition surface. Methods for predicting the meteorological contributions to Int above 1 cm have been described.²

The initial or 1974 correlation for Int was,

$$\begin{aligned} \text{Int}_{1974} = & -\exp \left\{ -23.667 + 5.555 \ln \frac{d}{z_0} \right. \\ & - 0.07681 \left(\ln \frac{d}{z_0} \right)^2 + 0.9722 \ln \frac{u_*}{v_t} \\ & + 0.03799 \left(\ln \frac{u_*}{v_t} \right)^2 - 2.254 \ln \frac{D}{u_* z_0} \\ & \left. - 3.724 \ln \frac{\rho_p u_* d^2}{18 \mu z_0} \right\} \quad (4) \end{aligned}$$

in which d is particle diameter, z_0 is roughness height, D is the Brownian diffusion coefficient, ρ_p is particle density of 1.5 g/cm³, and μ is viscosity. Since particle density was not varied in the development of this correlation, Int is assumed to be independent of particle density changes.

In the new modeling approach, the important parameters for final mass transfer are principally controlled by the particle relaxation time τ^+ (particle inertia) and the Schmidt number Sc . These terms are

$$\tau^+ = \frac{\rho_p d^2}{18 \mu} \frac{u_*^2}{v_t} \quad (5)$$

and

$$Sc = \nu/D. \quad (6)$$

A new correlation, based upon τ^+ , Sc , and statistically significant interactions, was developed using least squares techniques to fit experimental values for Int . The correlation is

$$\begin{aligned} \text{Int}_{1975} = & -\exp \left\{ -66.392 \right. \\ & + \ln Sc [11.42 - 0.4438 \ln Sc \\ & + 9.562 \times 10^{-3} \ln \left(\frac{d}{z_0} \right)] \\ & + \ln \tau^+ [-5.216 - 0.3247 \ln \tau^+ \\ & \left. + 0.3337 \ln \left(\frac{d}{z_0} \right) - 0.3910 \ln \left(\frac{D}{z_0 u_*} \right)] \right\}. \quad (7) \end{aligned}$$

In both correlations all terms are statistically significant at the 99% level and the multiple correlation coefficients are 0.92. There appears to be no statistically significant difference between fitting the data with either Eq. (5) or (6).

PREDICTED DEPOSITION VELOCITIES

Deposition velocities were predicted using the mass transfer resistances from both correlations combined with the meteorological mass transfer resistances from 1 cm to 1 m heights. These deposition velocities, K_{1-M} , are referenced to the airborne concentration 1 m above the surface. Deposition velocities were predicted as a function of particle diameter, friction velocity, surface roughness, and particle density. Particle density changes effect only v_t and not Int in Eq. (1) through (3).

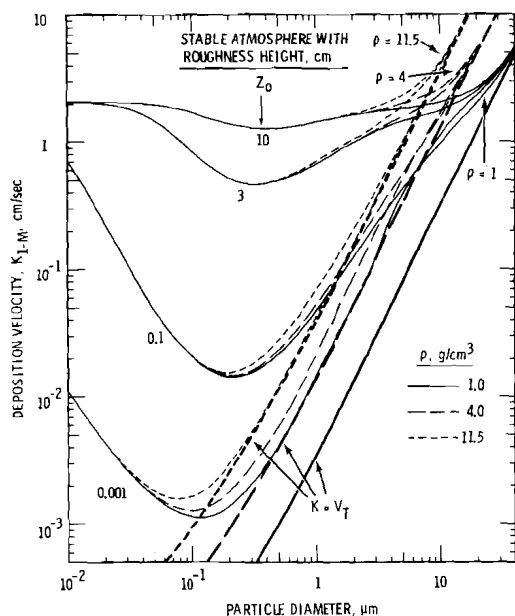
Comparisons for predictions between the two models are shown in Figures 1 and 2 for a friction velocity of 30 cm/sec. Deposition velocities are always greater than the terminal settling velocity v_t . The main difference between predictions is the minimum deposition velocity value for each roughness height. For other friction velocities, the minimum values from the two models show both less and greater deviation than this

example for the two correlations. In general, the new correlation does not predict as large a dependency of K_1-M upon roughness as the older correlation.

CONCLUSIONS

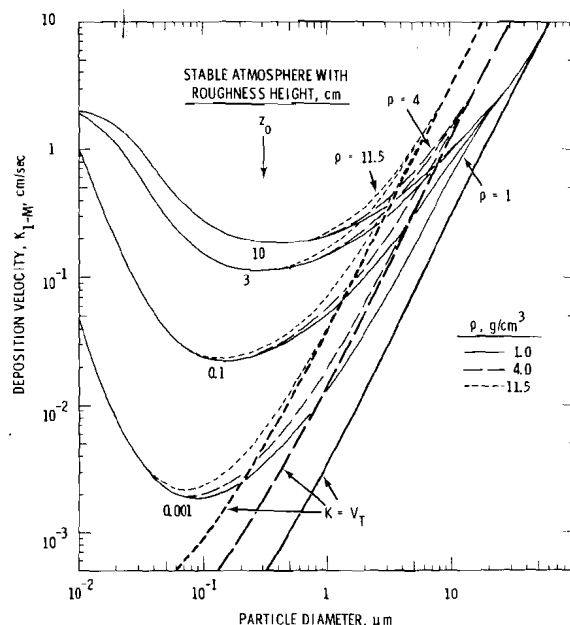
Predicted deposition velocities depend on which correlation is used

to predict the mass transfer adjacent to the deposition surface. As the data base for developing correlations is increased, predicted deposition velocities should become more accurate. Nevertheless, these are the only experimentally-based correlations to predict deposition velocities for rough surfaces.



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FIGURE 1. 1974 Model Predicted Deposition Velocities at 1 m for $u_* = 30$ cm/sec and Particle Densities of 1, 4 and 11.5 g/cm³



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FIGURE 2. 1975 Model Predicted Deposition Velocities at 1 m for $u_* = 30$ cm/sec and Particle Densities of 1, 4 and 11.5 g/cm³

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PARTICLE DEPOSITION AND PENETRATION THROUGH VEGETATION
CANOPIES FROM WIND TUNNEL EXPERIMENTS

G. A. Sehmel and W. H. Hodgson

Particle deposition velocities of monodispersed 0.8 to 29 μm diameter particles were measured during deposition onto 3.8 to 5.1 cm diameter crushed gravel and 9 cm tall artificial trees. At higher wind speeds increased foliage area for trees appears to significantly alter deposition velocity curves, (as a function of particle diameter). Particle penetration through the canopies to the underlying surface ranged up to 83%.

INTRODUCTION

Particle deposition velocities describe mass transfer rates at the air-surface interface. Predicted deposition velocities can vary from about 10^{-4} to 10^2 cm/sec as a function of particle diameter,^{1,3} particle density, friction velocity, and aerodynamic surface roughness. However, prediction procedures can not yet describe the effects of foliage density on deposition velocities, nor particle penetration through the foliage to the underlying surface. Since initial results⁴ indicated significant particle penetration through the top layer of a crushed gravel deposition surface, particle penetration could also be significant for vegetation canopies. Consequently, penetration results and foliar deposition velocities are needed to improve deposition velocity models. Here we report large particle penetration results to a crushed gravel surface and also penetration and deposition velocities for an artificial tree foliage fetch 9 cm high.

EXPERIMENTAL

Deposition velocities and particle penetration were measured in a 61 cm by 61 cm cross section wind tunnel, using monodispersed uranine particles. Experimental techniques have been described.⁵ In brief, air concentration is measured by isokinetic sampling and deposition is measured by dissolving the deposited particles in

water. The mass of uranine is determined fluorimetrically from the wash solution.

The crushed gravel deposition surface covered the entire 7.3 m by 61 cm wind tunnel floor to a depth of approximately 5 cm. Average gravel sieve diameters were between 3.8 and 5.1 cm. Deposition was measured on test sections 22 cm wide by 30 cm long, located between 4.7 and 6.3 m along the wind tunnel midplane.

The artificial tree foliage section was mounted on a 23 cm by 30 cm long plastic plate laid on top of the gravel surface at the 6.9 m length of the wind tunnel. Trees were mounted in the plate in a rectangular array with eight downwind rows of six trees. Tree spacing was 3.8 cm. The polyethylene trees were 7 to 9 cm high with a maximum crown width of 4 to 6 cm. Each crown had eight branches located around the central trunk, and the tree trunk extended ~2 cm below the crown. Particles depositing on the trees and plate were washed off and analyzed separately.

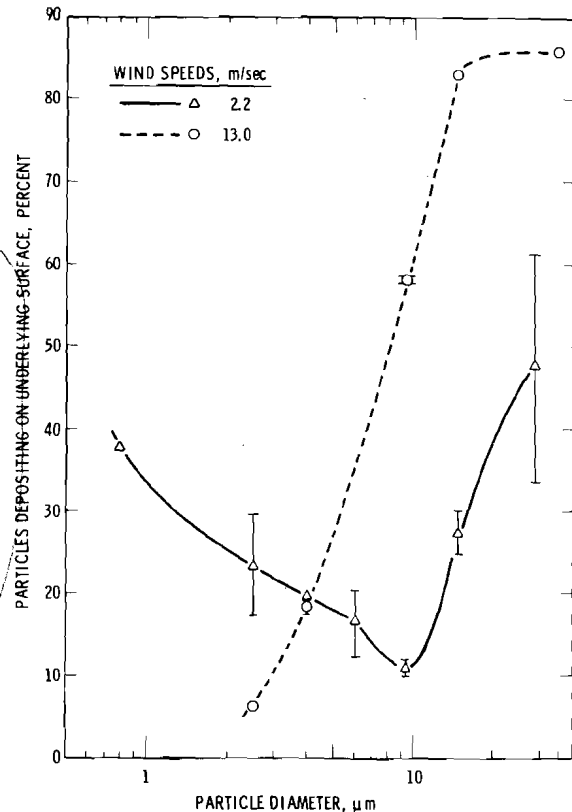
RESULTS

Nonuniform particle deposition in the tree fetch indicated edge effects. Depending upon particle diameter and wind speed, the front row of trees usually had either more or less deposition than downwind rows. These edge effects extended for three to

five rows. Since particle penetration to the entire plate was significant, deposition velocities for each row could not be calculated. Average deposition velocities for all trees and the plate are shown in Figure 1. The two wind speeds were measured upwind and at 6 cm above the gravel surface. This 6 cm height is approximately the height of the tree crown mid-plane.

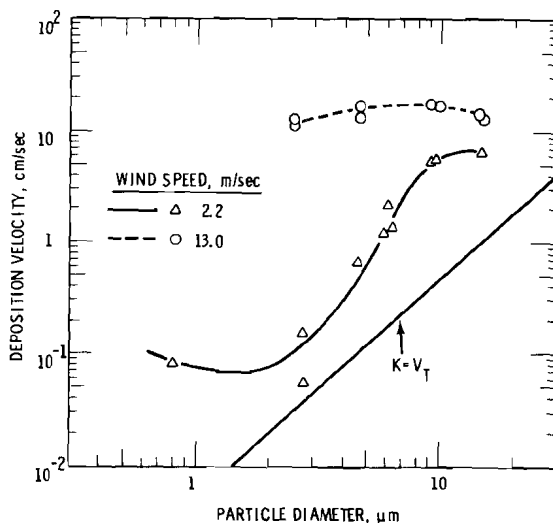
Deposition velocity curves show different patterns than those for simpler surfaces.¹⁻³ At a wind speed of 2 m/sec, a minimum deposition velocity occurs at about 1 to 2 μm for the trees. In contrast, simpler surfaces have minima in the particle diameter range from 0.1 to 1 μm . For a 13 m/sec wind speed, deposition velocities are nearly constant for all particle diameters studied. Again, in contrast, deposition velocities for 2 μm compared to larger particles would be significantly less for simpler surfaces.

Particle penetration to the plastic mounting base for the trees is shown in Figure 2. Penetration is the deposition to the plate as a percent of the deposition to the plate and trees. Penetration was significant with a maximum value of 83%. Penetration is a function of both wind speed and particle diameter. A cross-over diameter is about 4 μm .



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FIGURE 2. Penetration of Depositing Particles Through a Canopy of Plastic Trees (trees 9 cm high and 3.8 cm wide, in 8 downwind rows of 6 trees each)



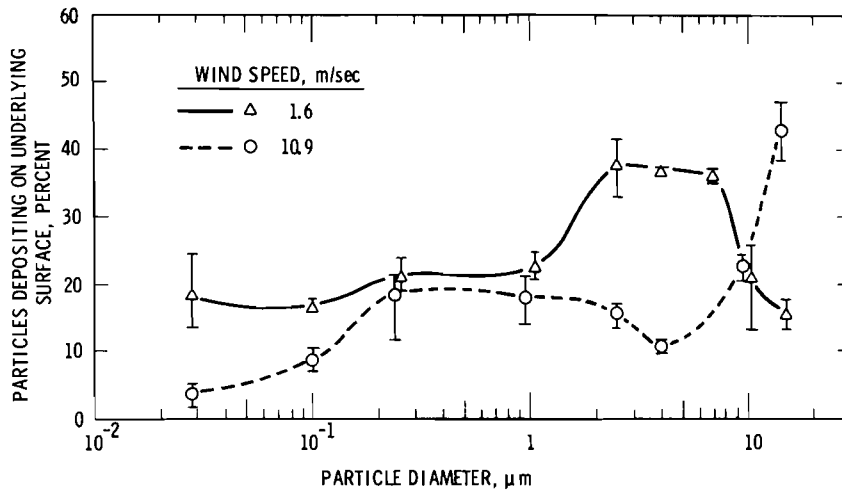
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FIGURE 1. Deposition Velocities to a Canopy of Plastic Trees 9 cm High

For particles smaller in diameter, penetration is greatest at the low wind speed. For larger particles, penetration is greatest at the high wind speed.

Penetration through crushed gravel is shown in Figure 3. In this case, the two wind speeds were measured 3 cm above the gravel surface. The particle diameter range is larger in this figure. Penetration is greater at the slower (1.6 m/sec) wind speed, for particle diameters from 3×10^{-2} μm up to the 9 μm cross-over diameter.

The penetration curves for crushed gravel and trees are similar. Penetration is larger for the slow wind speeds for particle diameters below the cross-over point. However, the particle diameter at the cross-over point is a factor of two (4 versus



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FIGURE 3. Penetration of Depositing Particles Through 3.8 to 5.1 cm Diameter Crushed Rock to an Underlying Surface

9 μm) less for trees than for crushed gravel. Apparently the nonrigid tree motion and open tree structure tend to enhance resuspension at the smaller 4 μm particle diameter as compared to the rigid, more dense gravel structure. For particles larger than the cross-over point, penetration was always greater at the higher wind speed.

An explanation was sought for these penetration similarities. One possibility is that above the cross-over point deposition and resuspension occurred simultaneously at the higher wind speed. If particles resuspended from the upper surfaces of the gravel and trees while the flux of penetrating particles remained constant, the observed penetration would increase since fewer particles remained deposited on the upper surfaces. For particles smaller than the cross-over diameter, a satisfactory suggestion is yet needed to explain higher penetration at lower wind speeds. Possibly for these par-

ticle relaxation times the air turbulence scale and intensity or relative wind profiles through the canopies are sufficiently different for the two wind speeds.

CONCLUSIONS

Predictive deposition velocity models must eventually consider both deposition within vegetative canopies as well as deposition penetration to the underlying surface. However, this penetration data base is very limited and consequently penetration cannot yet be generalized in predictive models. Deposition velocities to vegetation (9 cm trees) do show edge effects at the entrance to the "grove" of vegetation. Similar edge effects may be very important within forests since most forests contain many clearings from small to large size. Indications are that resuspension could probably occur simultaneously with deposition on the upper surfaces of vegetation.

References

1. G. A. Sehmel, "Comparison of Deposition Velocities Predicted from Dimensionless Analysis Correlations," see this report.
2. G. A. Sehmel and W. H. Hodgson, "Particle Dry Deposition Velocities," Pacific Northwest Laboratory Annual Report for 1974 to the USAEC Division of Biomedical and Environmental Research, Part 3, Atmospheric Sciences, BNWL-1950 PT3, Battelle-Northwest, Richland, Washington, pp. 175-180, 1975.
3. G. A. Sehmel and W. H. Hodgson, "Predicted Dry Deposition Velocities," Proc. of the Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants--1974 Symposium, Richland, Washington, Sept. 4-6, 1974, ERDA Symposium Series CONF-740921 in press, November 1975.
4. G. A. Sehmel and W. H. Hodgson, "Penetration of Depositing Particles through Crushed Gravel," Pacific Northwest Laboratory Annual Report for 1974 to the USAEC Div. of Biomedical and Environmental Research, Part 3, Atmospheric Sciences, BNWL-1950 PT3, Battelle-Northwest, Richland, Washington, pp. 181-183, 1975.
5. G. A. Sehmel, W. H. Hodgson, and S. L. Sutter. "Dry Deposition of Particles," Pacific Northwest Laboratory Annual Report for 1973: Part 3, Atmospheric Sciences, BNWL-1850 PT3, Battelle-Northwest, Richland, Washington, pp. 157-162, 1974.

FIELD DEPOSITION VELOCITY MEASUREMENTS TO A SAGEBRUSH CANOPY

G. A. Sehmel and W. H. Hodgson

Experimental upwind and downwind air sampling arrays were set up and an initial mass budget experiment conducted to measure particle deposition velocities across a 136 m fetch of sagebrush. Particle diameters in the range of 0.7 μm (where deposition velocities should be nearly independent of particle diameter) were used. Plume depletion within the canopy was 65%.

INTRODUCTION

Particulate air pollution removal by dry deposition has been predicted from controlled wind tunnel experiments to be a function of many variables including particle diameter, surface roughness, and air friction velocity. Predicted¹ deposition velocities increase as both surface roughness and friction velocity increase, but any effects of the canopy vegetation density on deposition velocities are unknown and are only now being studied.² Predicted deposition

velocities as a function of particle diameter are a minimum and are almost constant in the particle diameter range of ~ 0.1 to 1 μm . In contrast, deposition velocities reported for field measurements have not been correlated and generalized into predictive models. The inability to correlate field data may have been caused by inaccuracies in the experimental techniques. The principal experimental difficulty probably has been poor particle size control. Reported field deposition velocity measurements may be accurate, but these

deposition velocities cannot be generalized. The results are masked by the particle size dependency and the use of polydispersed particle sizes.

The objective of this research is to measure deposition velocities in the field with particle sizes for which deposition velocities are nearly independent of particle size. The initial deposition site is a reasonably dense sagebrush area.

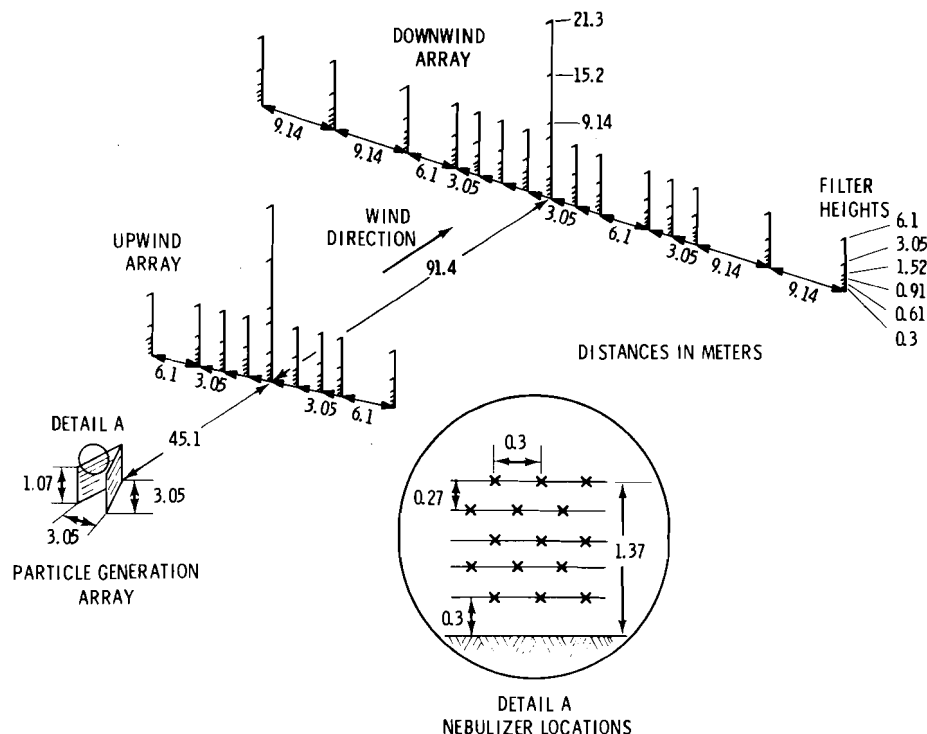
EXPERIMENTAL

An initial deposition velocity mass budget measurement was made with the field deposition arrays shown in Figure 1. The arrays are used to determine a mass budget between the airborne particles entering the upwind array and exiting through the downwind array. The experiment was conducted for a 11-min time interval during which the wind was blowing from the particle generation array into the upwind and downwind tower arrays.

The particle generation array consisted of 100 Retec nebulizers* mounted in a V-shaped rack. As shown, particles were generated at five heights from 0.3 to 1.37 m. Particles were generated from a 1 g/l uranine solution of 80% ethanol--20% water. After solution droplet drying in air, the mass average particle diameter was $0.7 \mu\text{m}$ with a standard deviation of 1.9. After generation, the particles had 30 sec to dry at a 1.5 m/sec average wind speed before passing through the upwind tower array.

All air sampling towers were 6.1 m high except the central 21.3 m high towers. Distances between towers were 3.05 to 9.14 m. Airborne particles were sampled onto 47 mm fiber-glass filters mounted on each tower at heights from 0.3 m to 21.3 m. The average sample flow rate was $0.03 \text{ m}^3/\text{min}$. After sampling, the uranine on

* Retec model PN7002, Retec Development Laboratory, Portland, Oregon



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FIGURE 1. Field Deposition Arrays

the filters was dissolved and each solution was analyzed fluorimetrically to determine the mass of uranine collected.

CALCULATIONS

Airborne particle fluxes entering the upwind array and downwind array were determined from the uranine exposure at each filter location. The exposure at each filter location was calculated from:

$$\text{Exposure} = \frac{\left[\frac{\text{(uranine mass on filter)}}{\text{(wind velocity at filter height)}} \right]}{\text{(filter face velocity)}}$$

The assumption was that the wind velocity profile at the central upwind tower was representative of all tower sites. Total exposure at each height was calculated by graphically integrating as a function of crosswind distance the exposures at each filter location. Since the wind direction and subsequently the maximum of the airborne plume had shifted from the central tower during the experiment, crosswind integration was from the plume maximum to one-half the tower spacing distance beyond the outermost tower. The total plume was assumed to be symmetrical around this maximum plume location. The total vertical airborne exposure was calcu-

lated by integrating with height the total exposure at each height.

RESULTS

Plume depletion in the 1.2 to 1.5 m high sagebrush was 65% between the upwind and downwind air sampling arrays for a 1.5 m/sec air velocity at a 1.5 m height. Uncertainties in the mass budget suggest the depletion uncertainty is between 57 and 73% due to the noncentering of the plume on the central tower. However, uncertainties could be greater since deposition was only 22% from the particle generators to the upwind sampling array. For equivalent deposition/length, this 22% corresponds to only 45% deposition between upwind and downwind sampling towers. The experiment will be repeated to minimize uncertainties. A deposition velocity is yet to be calculated from the observed plume disposition.

CONCLUSIONS

Plume depletion was much more rapid than the few percent expected. The experiment will be repeated at different meteorological and surface conditions to determine reproducibility. If results are reproducible, dense vegetation is an excellent dry deposition surface.

References

1. G. A. Sehmel, "Comparison of Deposition Velocities Predicted from Dimensionless Analysis Correlations," see this annual report, pp. 83-89.
2. G. A. Sehmel and W. H. Hodgson, "Particle Deposition and Penetration through Vegetation Canopies from Wind Tunnel Experiments," see this annual report, pp. 86-89.

RESUSPENSION RATES FROM A CIRCULAR FIELD SOURCE

G. A. Sehmel and F. D. Lloyd

Experiments are continuing in a lightly vegetated area to measure wind-caused particle resuspension rates of a controlled submicrometer inert tracer. Resuspension rates continue to reproducibly increase with wind speed for this aged source. The apparent availability for resuspension has not decreased for times up to 8 months after deposition.

INTRODUCTION

Wind surface stresses can resuspend surface-deposited materials into the air. Prior to this research, concentrations of resuspended plutonium and other radioactive materials were measured over contaminated areas, and these results were reported as resuspension factors [defined as the ratio of airborne concentration to the surface (areal) concentrations]. The resuspension factors reportedly decreased with a half-life of about 30 days.

This research sought to determine, using controlled sources, wind-caused particle resuspension rates as a function of wind speed, and also to measure the weathering half-life.

EXPERIMENTAL

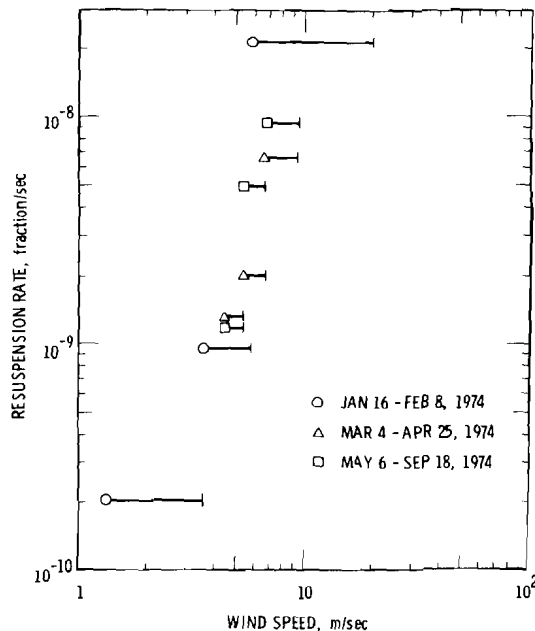
Wind resuspension experiments are continuing at the original controlled source resuspension site. Briefly, the site¹ is a lightly vegetated desert soil with a roughness height, z_0 , of 3.4 cm. On October 2, 1973, a known quantity of a tracer suspension of submicrometer calcium molybdate was sprayed on the ground in a circle of 22.9 m radius. Resuspended Mo tracer was sampled from a 6.1 m tower erected at the center of the tracer area. Self-orienting impactor-cowl² air samplers directed into the wind were mounted at tower heights of 0.3, 0.9, 1.8, 3.0 and 6.1 m. Three air samplers were at each height. Thus, there were three sets of six air samplers. Each set of six air

samplers was turned on automatically when the wind speed was within selected wind-speed increments. Resuspension rates were calculated for each wind speed increment from the vertical airborne concentration profiles of Mo.

Analytical results from the air filters have been received for two additional air sampling time periods. Resuspension rates were calculated from airborne Mo fluxes, ground surface concentration, and sampling times. Average resuspension rates for "respirable" particles were calculated for all particles entering each particle impactor. A resuspension rate for larger "nonrespirable" particles was also determined from particles settling within each cowl.

RESULTS

Resuspension rates for respirable particles increased rapidly with increase in wind speed. Resuspension rates are shown in Figure 1. Data points are plotted at the lower limit of each wind speed interval. A horizontal line through each data point extends to the upper limit of that wind speed interval. Resuspension rates increased two orders of magnitude, from 2×10^{-10} fraction resuspended/sec to 2×10^{-8} fraction resuspended/sec, as wind speed increased from 1.3 to 6.6 m/sec. The overall increase in resuspension rates is a nonlinear function of wind speed. However, between 4.4 to 6.6 m/sec wind speeds, resuspension rates appear to increase with the 4th



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FIGURE 1. Particle Resuspension Rates as a Function of Wind Speed

to 5th power of wind speed. The experiment is continuing and is directed toward determining the exponential relationship.

Resuspension rates for tracer particles used at this location do not appear to be decreasing as a function of time during the period the tracer

has been on the ground. The resuspension rate is so small that the total source concentration on the ground is diminishing rather slowly. Since there are 3.15×10^7 sec/year, only 0.1 of the source would be resuspended even if resuspension rates were the highest measured for an entire year.

Resuspension rates for "nonrespirable" particles were 1.3×10^{-11} fraction resuspended/sec during January 16 to February 8, 1974, and 8.9×10^{-12} fraction resuspended/sec during May 6 to September 18, 1974. Wind speed dependency could not be determined for these larger particles due to the sampling technique. Large particles could be blown into the sampling cowl even if the high-volume air sampler were not operating.

CONCLUSIONS

Particle resuspension rates caused by wind stresses are of the order of 10^{-10} to 10^{-8} fraction resuspended/sec for a lightly vegetated desert soil and increase rapidly with an increase in wind speed. Since these rates are nearly constant over an 8-month time period, the concept of a 30-day weathering half-time is questioned. Results from additional experiments should determine when and if resuspension rates decrease. Other sites must be studied to determine both resuspension rates and weathering as a function of soil, surface,³ particle, and wind parameters.

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2. G. A. Sehmel, "An Evaluation of a High-Volume Cascade Particle Impactor System," Proc. of the Second Joint Conference on Sensing of Environmental Pollutants, Instrument Society of America, Pittsburgh, PA, pp. 109-115, 1973.
3. G. A. Sehmel, "Experimental Measurements and Predictions of Particle Deposition and Resuspension Rates," paper 75-25.2 presented at the 68th Annual Meeting of the Air Pollution Control Association, Boston, MA, 1975, BNWL-SA-5228, Battelle-Northwest, Richland, WA, 1975.

AN EXPERIMENT TO MEASURE PARTICLE RESUSPENSION
FROM A LARGE ANNULAR AREA

G. A. Sehmel and F. D. Lloyd

Equipment installation was completed and the first experiment begun to measure inert particle resuspension rates from a 1031 m² annular source. Mass budgets are being made with 108 air filter samplers mounted on six towers. Air is sampled as a function of wind direction and six wind speed increments.

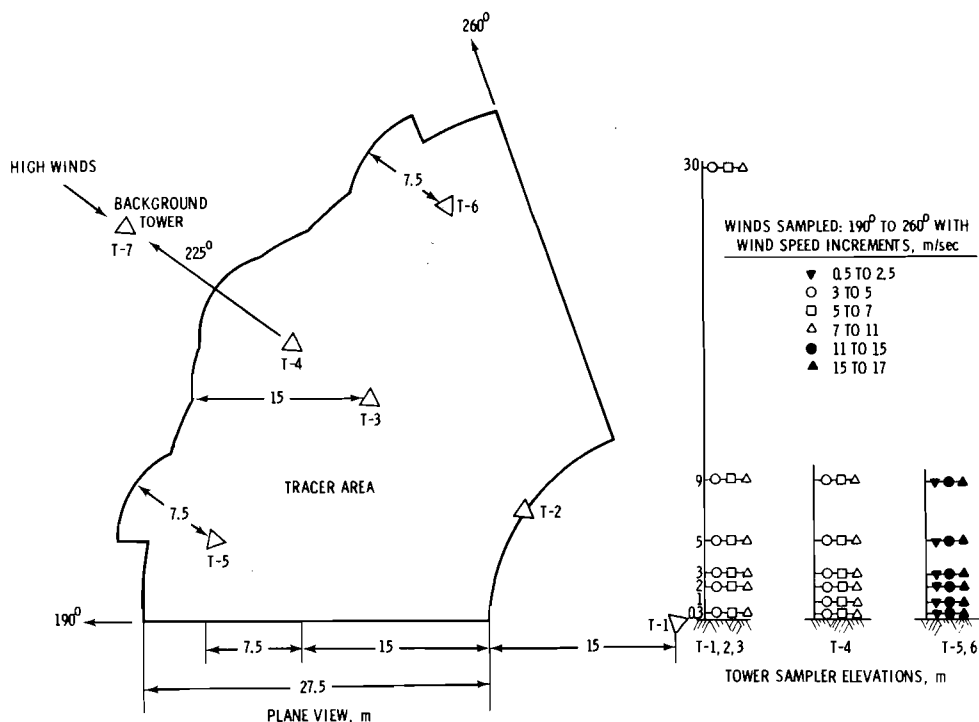
INTRODUCTION

Wind-caused particle resuspension rates of tracer particles are being measured using one sampling tower in a lightly vegetated area.¹ To determine reproducibility, resuspension rate measurements were needed at a second site. The objectives of this second experiment are to determine resuspension rates as a function of

wind speed increments and atmospheric stability as well as the upward diffusion coefficients for resuspended particles.

EXPERIMENTAL

Equipment installation was completed and a resuspension experiment begun at the site shown schematically in Figure 1. The figure shows the



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FIGURE 1. Resuspension Experiment Sampling Only Selected Wind Direction and Wind Speed Increments

tracer area; one upwind background sampling tower, T-7; and six downwind sampling towers. Open-faced 8 x 10 in. air filter samplers are mounted on these towers. Sampler orientation is toward a 225° wind direction.

The tracer area is a sector bounded by radial lines on 190° and 260° with a center on tower 1. The inner part of the sector to a distance of 15 m (arc passes through tower 2) is not seeded with tracer. Tracer is also seeded in 70° arcs upwind of towers 4, 5, 6. These areas extend somewhat beyond the arc centered on tower 1. This geometry of the source resulted from tower placement and the desire to sample a near-uniform tracer area from 190° to 260°, generally upwind of each tower. The tracer was sprayed in 30 equal segments to give an average surface loading of 0.87 g Mo/ft. The tower area is flat and was reasonably unvegetated when sprayed. Originally, cheat grass covered the area, but was burned over a few weeks before spraying. Vegetation is now growing back in the area.

The sampler locations on each tower are schematically shown on the right side of the figure. There are 18 samplers on each tower at heights from 0.2 to 30 m. These samplers turn on automatically when both the wind direction is from 190° to 260°

and the wind speed is within six indicated increments extending from 0.5 to 17 m/sec. Three wind speed increments are used for towers 5 and 6 and three other wind speed increments are used for towers 1, 2, 3, and 4. Airborne concentrations measured with towers 1, 2, 3 and 4 will be used to determine resuspension rate reproducibility and upward diffusion coefficients. Results from all towers will be used to determine the resuspension rate depending upon wind speed.

This initial experiment is being run for all atmospheric stabilities due to the short time between tracer spraying and expected winter conditions. Future experiments will also utilize the temperature lapse rate along with wind direction and speed to activate the air samplers.

CONCLUSIONS

Resuspension rates from this experiment will determine the wind speed dependence of resuspension rates. In an earlier experiment the dependency of resuspension rate on wind speed was poorly defined since only three wind speed increments were used. Succeeding experiments will determine decreased resuspension rates caused by growing vegetation and effects of atmospheric stability on resuspension rates.

Reference

1. G. A. Sehmel and F. D. Lloyd, "Resuspension Rates from a Circular Field Source," this volume, pp. 92-93.

PARTICLE RESUSPENSION FROM TRUCK TRAFFIC
IN A CHEAT GRASS AREA

G. A. Sehmel

Resuspension rates caused by truck traffic were determined using ZnS tracer particles and downwind air sampling towers in a flat cheat grass area. Resuspension rates were initially high until most of the tracer readily available on the upper portions of the grass was removed. Resuspension rates were one to two orders of magnitude lower than similar rates for resuspension from an asphalt road.

INTRODUCTION

Plutonium or other hazardous materials deposited on environmental surfaces are resuspended into the air by surface stresses produced from vehicular traffic. These resuspension stresses can be greater than simple wind stresses. Resuspension rates for car and truck traffic on asphalt roads have been determined using a ZnS tracer.¹ Resuspension rates increased with vehicular speed. At 60 mph, approximately 1% of the tracer became airborne when the vehicle was driven through the tracer.

Most contaminated areas, however, are not asphalt roads. Usually contaminated areas are highly vegetated, controlled areas which are restricted to human activity. The vegetation cover helps to decrease wind resuspension; this is desirable to decrease potential downwind transport. Nevertheless, human activity including vehicular traffic can occur in these contaminated areas. Consequently, controlled experiments were needed to estimate resuspension rates caused by vehicular traffic in contaminated areas. The objective of these experiments was to determine truck-caused resuspension rates as a function of truck speed in a cheat grass area.

EXPERIMENTAL

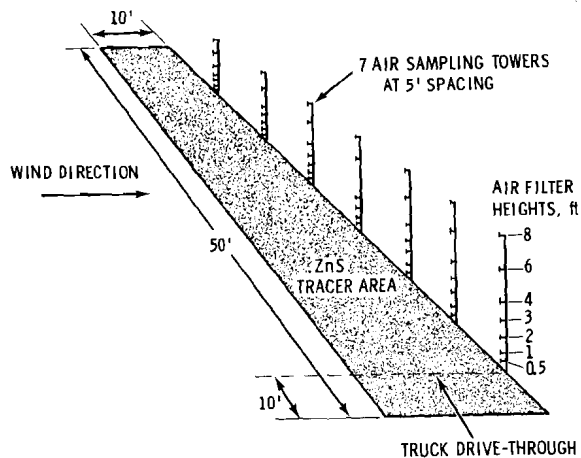
Particle resuspension rates were determined on two dry June days using the experimental arrangement shown schematically in Figure 1.

The important features are the 10 x 50 ft ZnS tracer area defining the road, the wind blowing perpendicular to the road, and the seven air sampling towers downwind and adjacent to the road. The average airborne exposure of ZnS* tracer resuspended from the tracer area road was determined by horizontally and vertically integrating the ZnS amounts collected on the eight air filters on each tower. Wind-caused resuspension was measured immediately after the tracer was deposited on the ground and also after 3/4 ton truck resuspension runs had been completed. In these wind cases, average wind speeds were determined with integrating Casella cup anemometers located at heights of 20, 43, and 88 in.

The ZnS particles were deposited onto the ground by rapping containers on the ground. Deposition was through 25 μ m screens on the bottom of the containers. The mass mean particle diameter passing through the screens was 8 μ m. An average of 0.25 g/ft² was deposited on the tracer area.

Airborne particles were collected on the towers with membrane filters at a flow rate of 1 cfm. Exposed filter diameter was 1.6 in. Particles collected were analyzed¹ for total ZnS.

* ZnS type 2210, United States Radium Corporation.



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FIGURE 1. Schematic Showing Experimental Design for Particle Resuspension from a Cheat Grass Area Caused by Truck Passage

RESULTS

Six resuspension experiments were conducted in a 2-day time period rather than 1 day since the wind direction was not always into the air sampling towers. On the first day tests were for initial wind resuspension and for a truck speed of 5 mph. On the second day, experiments were for truck speeds of 15, 30 and 40 mph, and a final wind resuspension experiment.

Wind resuspension rates are shown in Table 1. On the first day, the resuspension rate was 3.38×10^{-8} fraction resuspended/sec for a 4.7 m/sec average wind speed. This is a relatively high resuspension rate and probably reflects increased resuspension of ZnS particles from the upper portions of the cheat grass.

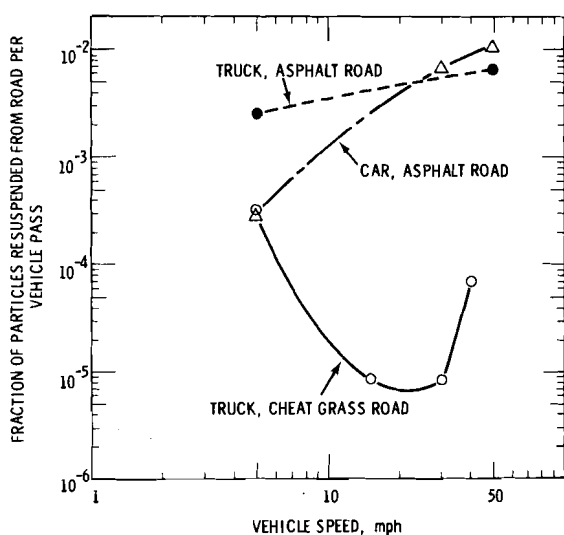
After 3.5×10^{-3} of the ZnS has been removed by truck traffic, the wind resuspension rate decreased to 1.25×10^{-9} fraction resuspended/sec. This last rate is almost one order of magnitude greater than the 2×10^{-10} fraction/sec resuspension rate² of an inert submicron tracer from a somewhat comparable resuspension site and average wind speed.

Truck resuspension rates are defined as the fraction of particles removed each time the truck was driven at a constant speed through the tracer area. The units are fraction resuspended per vehicle pass through the area. In using this resuspension/pass concept, the total airborne material is the product: (fraction resuspended/pass) (length of pass) (average surface contamination/length of pass). Fraction resuspension per pass = airborne/initial deposit.

Truck resuspension rates are shown in Figure 2 for the cheat grass experiments and are compared to vehicle resuspension¹ from an asphalt road. Truck resuspension from the cheat grass is always less than for the asphalt road. This decrease is attributed to the protective action of the cheat grass in hindering truck generated air turbulence from reaching the ground. For the asphalt, resuspension/pass increased with vehicle speed. This increased resuspension was caused by greater surface wind stresses from the increased vehicle speed. In contrast, the resuspension/pass decreased for the cheat grass as the truck speed was increased from 5 to 30 mph. This decrease is attributed to the truck speed sequence used in the experiments. The initial truck resuspension was at 5 mph. Apparently, the large resuspension/pass at 5 mph was caused by removing the most readily resuspended particles

TABLE 1. Wind Caused Resuspension Rates

Time	Wind Speed at 1.5 m, m/sec	u_* cm/sec	Z_0 cm	Resuspension Rate, Fraction/sec
Immediately after ZnS deposited.	4.7	52	4	3.38×10^{-8}
After 3.5×10^{-3} of ZnS removed by truck traffic.	2.7	51	2	1.25×10^{-9}



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FIGURE 2. Particle Resuspension Rates from Asphalt and Cheat Grass Roads Caused by Vehicle Passage

from the cheat grass itself. In succeeding experiments at increasing truck speeds up to 15 mph (and possibly 30 mph) all the readily resuspended material was removed from the cheat grass foliage. When truck speed was increased from 30 to 40 mph, resuspension/pass also increased. At this time, increased air turbulence at the base of the cheat grass is suggested as the source of increased resuspension.

CONCLUSIONS

Particle resuspension rates from a cheat grass area is a function of both vehicle speed and the relative location of the material being resuspended. The results suggest that material at the base of the cheat grass is more protected from resuspension forces and is less readily resuspended than material on the upper portions of the cheat grass.

References

1. G. A. Sehmel, "Particle Resuspension from an Asphalt Road Caused by Car and Truck Traffic," Atmospheric Environment, vol. 7, pp. 291-301, 1973.
2. G. A. Sehmel and F. D. Lloyd, "Resuspension Rates from a Circular Field Source," this volume pp. 92-93.

THE INFLUENCE OF SOIL INSERTION ON ATMOSPHERIC PARTICLE SIZE DISTRIBUTIONS

G. A. Sehmel

Measured volume distributions indicate airborne concentrations from soil resuspension are different from bimodal distributions reported near cities with significant air pollution. Fine and coarse soil insertion modes are suggested as two new modes which should be included with air pollution modes near cities. The fine soil insertion mode can fill the reported valley at 1 to 3 μm reported near cities.

INTRODUCTION

Atmospheric aerosol size distributions in both urban and nonurban areas have been measured by various investigators. Results in general show size distributions can be multimodal. A mode is a predominant concentration of a size range between valleys of lower concentration for larger and smaller size ranges. Whitby, et al.,^{1,2,3} define the three volume modes as:

- 1) A transient nuclei or Aitken nuclei mode for particles less than about 0.08 μm .
- 2) An accumulation mode between about 0.08 μm and 2 μm .
- 3) A mechanically-generated aerosol or coarse mode for particles greater than 2 μm .

The accumulation mode is often caused by anthropogenic sources while the coarse mode is often mainly airborne soil. Modes are being considered important⁴ in identifying and monitoring air pollution since chemical composition is often different in each mode. Similar modes below 2 μm might also occur for resuspended soil. Consequently, the objective was to determine airborne soil concentrations and possible modes in a region with low anthropogenic air pollution.

EXPERIMENTAL

Airborne particle concentrations were measured with a laser optical particle counter.⁵ Measurements were usually 0.9 m above the ground and were for both dust storm and nondust storm conditions. The range of repeated measurements will be compared with previously reported⁶ results for the Hanford area. These latter results were determined with particle cascade impactor-cowl systems. The cowl is an entry passage to the impactor where larger particles deposit by gravity settling. Impactor data are for three particle diameters between 1.1 to 3.6 μm while the cowl particle data are for larger particles up to 230 μm diameter.

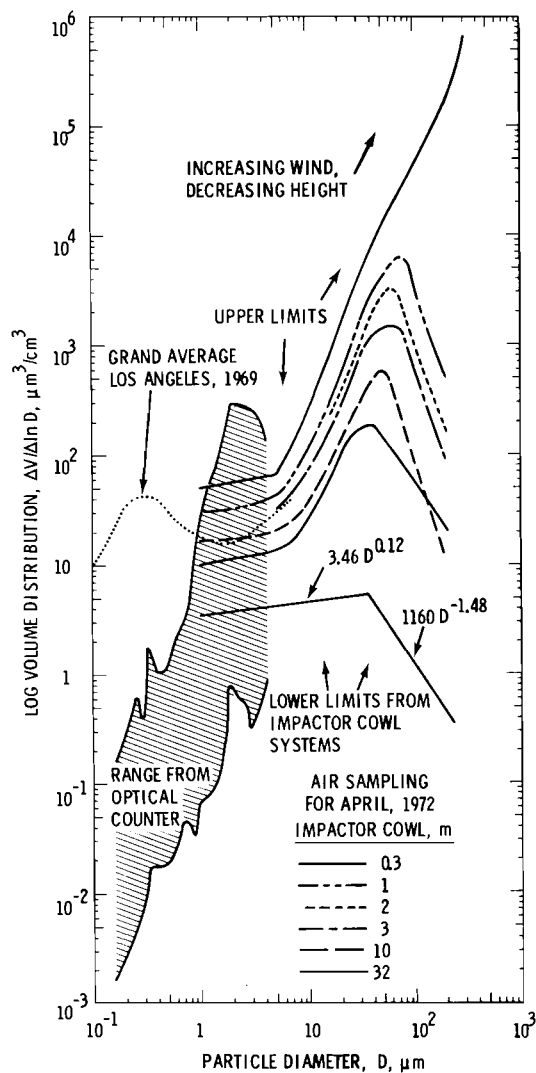
Results are reported as the volume distribution:

$$\frac{\Delta V}{\Delta \ln D} = \frac{\pi D^4}{6} \frac{\Delta N}{\Delta D} \quad (1)$$

which has units of $\mu\text{m}^3/\text{cm}^3$. The total airborne particle volume is the integral of Eq. (1). The total dust loading in $\mu\text{g}/\text{m}^3$ is calculated from the volume once a density is assumed. For this study a 2 g/cm^3 density is assumed.

RESULTS

Volume distributions are shown in Figure 1 for the optical counter,



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FIGURE 1. Airborne Particle Volume Distribution

impactor-cowl systems, and also for Los Angeles airborne particles in 1969. The range of optical counter results is shown by the lined area. In general, these volume distributions increase with approximately the square of particle diameter. There are no significant modes indicated in the ragged edges. The impactor-cowl data are shown for April 1972, during a dust storm. Concentrations were maximum near ground and decreased with increasing sampling heights. For nondust storm conditions, lower limits were indicated by the impactor-cowl system. These limits are:

$$\frac{\Delta V}{\Delta \ln D} = 3.46 D^{0.12} \text{ and}$$

$$\frac{\Delta V}{\Delta \ln D} = 1160 D^{-1.48}.$$

No significance should be attributed to their intersection at 38 μm.

There is reasonable consistency between the optical counter and impactor-cowl data. The data overlap between 1 and 4 μm. Differences between results for the two counting systems can probably be attributed to differences in soil moisture content and meteorology during sampling periods.

The lower and upper limit curves were integrated to determine the range of mass loading indicated by these volume distributions. Results are shown in Table 1 for three diameter ranges as well as the total mass from 0.16 to 230 μm. The maximum

TABLE 1. Apparent Limits of Airborne Soil Concentrations

Particle Diameter Range, μm	Total Volume, μm³/cm³		Soil Mass Loading	
			Lower Limit, μg/m³	Upper Limit, mg/m³
0.16 - 1	0.036	4.58	0.072	0.00916
1 - 10	3.83	3.5 x 10²	7.66	0.704
10 - 100	9.39	2.22 x 10⁴	18.78	44.4
100 - 230	0.609	9.37 x 10⁴	1.218	187.4
Total From 0.16 - 230	13.87	1.163 x 10⁵	27.74	232.6

airborne loading is 232.6 mg/m^3 with a $162 \text{ }\mu\text{m}$ mass-average particle diameter. The lowest airborne loading is $27.7 \text{ }\mu\text{g/m}^3$ with a $19 \text{ }\mu\text{m}$ mass-average particle diameter.

DISCUSSION

Airborne soil concentrations are also compared in Figure 1 with the 1969 average Los Angeles distribution between 0.1 and $7 \text{ }\mu\text{m}$. This curve between 0.1 and $2 \text{ }\mu\text{m}$ illustrates the accumulation mode. Most of the accumulation mode is over one order of magnitude greater than for local airborne soil concentrations. However, in the particle size range of around $2 \text{ }\mu\text{m}$ (a "valley" in the Los Angeles size distribution) the concentration of airborne soil could be greater. Indeed, a fine soil insertion mode could be defined as maximizing at about $2 \text{ }\mu\text{m}$. Similarly, coarse soil

insertion modes could be defined as maximizing at about 50 to $80 \text{ }\mu\text{m}$.

CONCLUSIONS

Volume distributions of airborne soil do not fit the bimodal distributions reported near cities with significant air pollution. There are different modes which reflect differences in the nature of the particle source.

Airborne soil concentrations should depend upon many variables including sampling height, wind speed, soil, and soil moisture. Some dependency upon wind speed⁸ has been determined with impactor-cowl systems, but additional data from the optical counter is needed to determine the influence of wind speed on the concentrations of the smaller soil particles.

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INITIAL OBSERVATIONS OF AIRBORNE CONCENTRATIONS OF SMALL
PARTICLE SIZE RANGES DURING TWO DUST STORMS

G. A. Sehmel

Airborne respirable soil concentrations were measured in small diameter increments with a laser optical counter. Concentrations were measured during two dust storms. Particles from 0.13 to 2 μm diameter tend to remain airborne longer than 0.3 to 2 μm diameter particles.

INTRODUCTION

Wind will resuspend plutonium or other hazardous contaminants deposited on soil. These contaminants will resuspend as particles the same size as originally deposited, or attached to host soil particles. Which ever the resuspension mode, it is important to know how wind stresses remove particles as a function of particle size. Soil particle resuspension is rapid during dust storms. Consequently, measurements of airborne respirable particle concentrations as a function of particle diameter in dust storms should suggest if all particle sizes are resuspended similarly. This paper reports initial history observations of airborne respirable size increments during the progress of two dust storms.

EXPERIMENTAL

The dust storms sampled were on May 23 (SW winds) and on September 11, 1975 (N winds) on the Hanford reservation. Particles were sampled with a laser optical counter* located 93 cm above the surface. The counter sizes and accumulates counts in multiple channels, with channel widths as small as 0.07 μm . These channels sized particles into the following size intervals, in μm : 0.13 to 0.2, 0.2 to 0.3, 0.37 to 0.45, 0.65 to 0.75, 0.95 to 1.05, 1.45 to 1.55, 1.60 to 1.85, and 1.85 to 2.10.

* Active Scattering Aerosol Spectrometer, Particle Measuring Systems, Inc., Boulder, Colorado.

Results are presented as the volume density function

$$\frac{\Delta V}{\Delta \ln D} = \frac{\pi D^4}{6} \frac{\Delta N}{\Delta D},$$

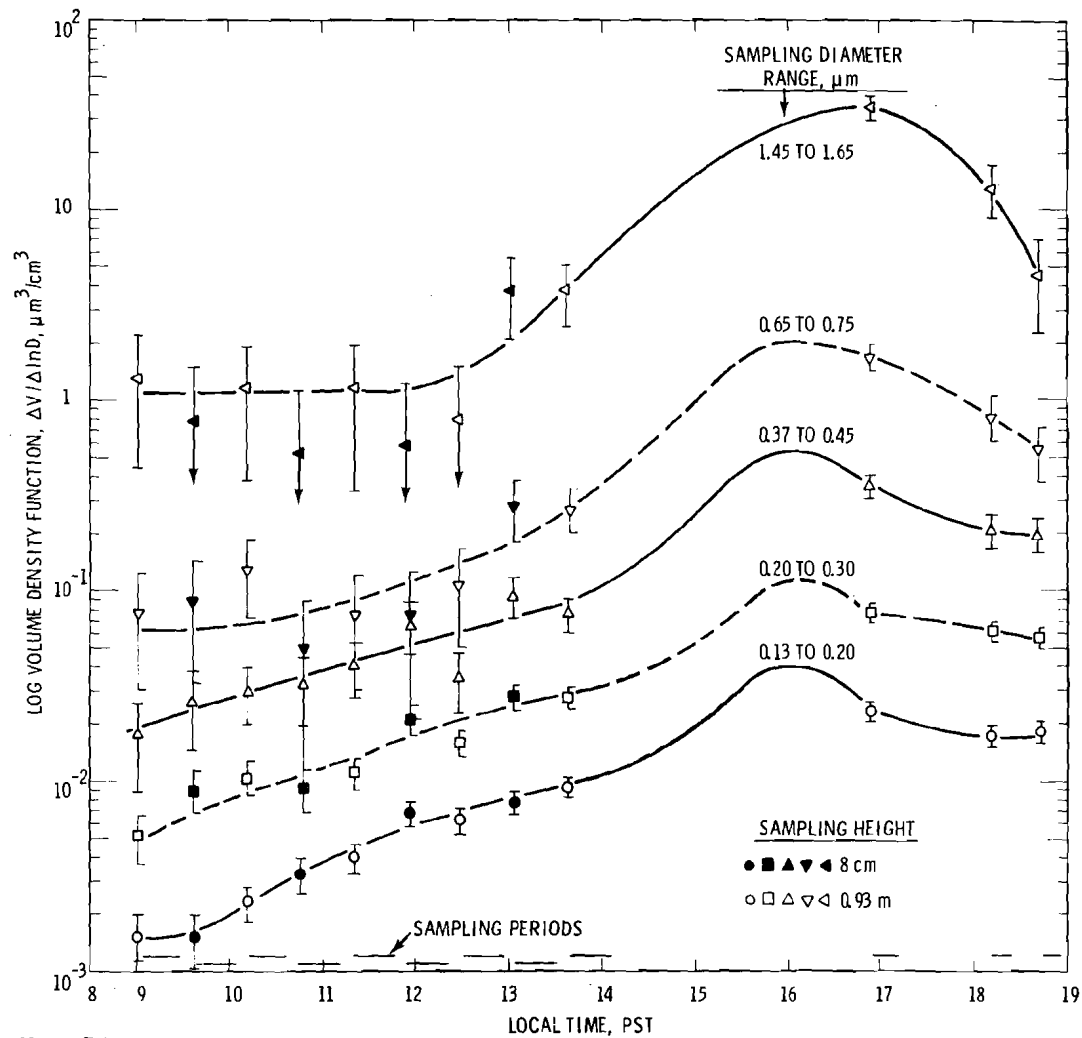
evaluated at the average diameter, D , for the increment. N is the number concentration. The units of $\Delta V/\Delta \ln D$ are $\mu\text{m}^3/\text{cm}^3$.

RESULTS

Volume density functions are shown in Figures 1 and 2 as a function of local standard time. Data points are plotted at the start of each sampling time increment indicated by horizontal lines at the bottom of each figure. Error limits for each data point are the 95% confidence limits from counting statistics. Standard deviations were calculated from the square root of the counts in each increment.

Airborne particle sizes and concentrations from the beginning to the end of a dust storm have yet to be measured. Particle sizes and concentrations were measured during a period following storm initiation, as it built up to a maximum, and the dust storm decay.

Discrete particle size data prior to the first dust storm and during the dust storm decay are shown in Figure 1. Measurements in the dust storm began at 5:00 PM, and continued until the dust storm was nearly dissipated at 7:00 PM. Visibility was



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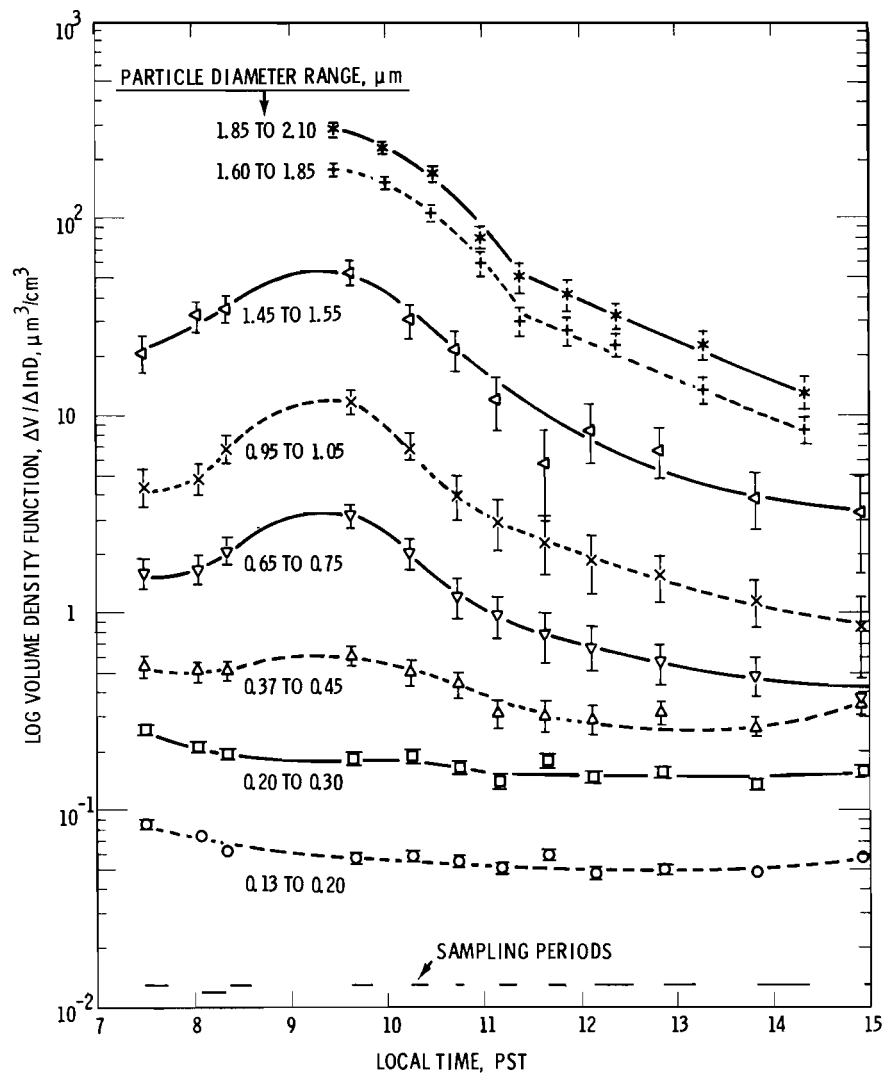
FIGURE 1. Airborne Particle Volume Density Function
During Dust Storm Day on May 23, 1975

less than 10 km at 5:00 PM and subsequently improved. Maximum measured concentrations occurred at 5:00 PM and subsequently decreased. The decrease was more rapid for the larger particles.

Figure 1 also shows ambient particle size data taken earlier during the day of the dust storm, but at 32 km from the dust storm measurement site. Ambient concentrations increased during the day, approaching the higher dust storm concentrations. This increase may signal an imminent dust storm. Too few measurements have been made to confirm such a generalization, but additional measurements may support our theory that

early increase in dust loading may indicate a dust storm.

A more complete dust storm history was taken for the second dust storm. Size data are shown in Figure 2. Concentrations in general are greater than for the storm represented in Figure 1. For particle diameters greater than about 0.37 μm , concentrations maximized about 9:30 AM. At that time, maximum visibility was 1.4 km. Subsequently, concentrations of greater than 0.37 μm particles decreased and visibility increased. In contrast, concentrations of 0.3 μm and smaller particles did not significantly decrease with time.



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FIGURE 2. Airborne Particle Volume Density Function
During Dust Storm Day on September 11, 1975

Only tentative model insights are currently suggested to explain the observed airborne particle concentrations as a function of particle diameter and time. In the simplistic case, a dust storm is a volume of air transporting dust at an average air velocity. Within the dust storm it is unknown if airborne concentrations change by either deagglomeration or agglomeration. Nevertheless, airborne soil concentrations must maximize at some point in the dust storm. Subsequently, larger diameter airborne soil particles are removed by dry deposition more rapidly than the smaller $0.3 \mu\text{m}$ particles. Obviously,

measured concentrations are larger if the location of the maximum dust storm concentration is near the sample site. In comparison to the larger particles, the smaller particles appear to be airborne longer at the sampling site. The reason for this increased residency time is not obvious if one thinks of a dust storm as a single air mass moving at an average velocity. Possibly, a dust storm is caused by a series of air eddies penetrating the relatively stagnant boundary layer. After re-suspension, the eddies lose their identity and mix with the relatively stagnant air. In this case, the

longer residency time of the stagnant air could explain the increased residency time of the small particles. However, some eddies must both penetrate the boundary layer and retain their identity in order to raise particles to large heights observed in dust storms.

CONCLUSIONS

Air motions and particle transport in dust storms are not understood.

Particle airborne residency times in the dust storm are a function of particle diameter. Some of this change can be attributed to increased dry deposition velocities for the larger particles. Although dust storm physics is not understood, the air concentration data are indicative that respirable sized particles can be resuspended.

FREQUENCY OF AIRBORNE DUST IN THE CONTINENTAL UNITED STATES

M. M. Orgill and G. A. Sehmel

Frequency of airborne dust periods having visibility less than 11 km were determined by analyzing existing weather data from 343 stations throughout the continental United States. Seasonal and regional distributions of dust frequency show that the majority of the country is subjected to suspended dust.

INTRODUCTION

The purpose of this study was to evaluate and quantify wind translocation and natural wind-caused airborne dust loadings throughout the contiguous United States. Initial results of this study were presented in last year's Annual Report.¹ The study is now completed and will appear as a separate report.² In this summary, some results from the final report will be presented on regional and seasonal distribution of suspended dust.

PROCEDURE

Data summaries for 343 weather observation stations were obtained from the National Climatic Center for this study. Data on the frequency of occurrence of dust, blowing dust and sand for annual, monthly and three-hourly periods were then extracted, plotted and analyzed. Frequency of dusty periods was based on summarized hourly weather observations from stations that recorded dust, etc., when

visibility was less than 7 miles (11 km) or when the dust concentration was generally over 3 mg m^{-3} .² Further details on the procedures are explained in the final report.

RESULTS

Regional Average Dust Frequency

The country was divided into seven different regions; South Central, Rocky Mountain, North Central, Pacific Coast, Southeastern, Great Lakes, and Northeastern. The divisions considered relative dust frequency, spatial dust frequency, patterns and geography. To quantify the dustiness in these seven regions a simple regional average dust frequency was defined as:

$$\text{RADF} = \sum_{i=1}^{i=N} \frac{f_i}{N} \quad (1)$$

where RADF is the annual or monthly regional average dust frequency; f_i

is the annual or monthly percent frequency of dust occurrence for the i th station in each region, and N is the total number of stations for each region.

A comparison of relative average dustiness of the seven regions was made by computing an annual regional average dust frequency from Eq. (1) for each region. These frequencies are shown in Table 1.

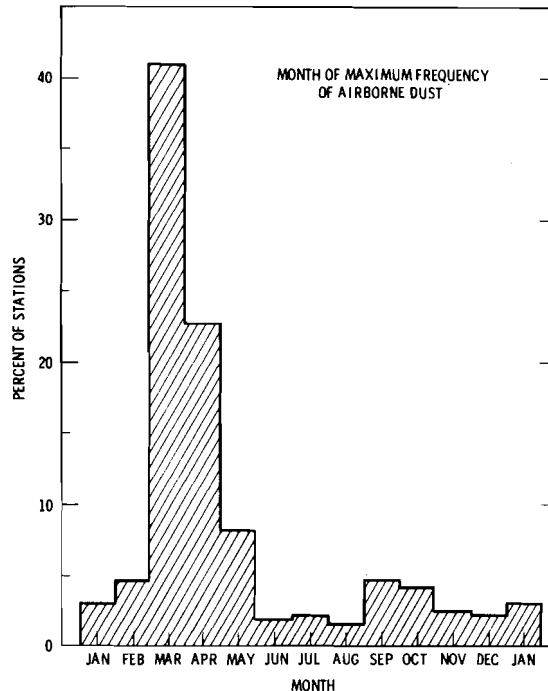
Comparison of the annual RADF for the seven regions shows that the South Central, Rocky Mountain (Western States) and North Central regions of the country have the highest relative dust frequency. The Northeast and Great Lakes regions have the lowest frequency. However, the relatively high frequency of dustiness for the Pacific Coast region is due entirely to the relatively high dust frequency of Central and Southern California. The coastal regions of Oregon, Washington, and Northern California have one of the lowest (if not the lowest) dust frequencies in the nation.

TABLE 1. Annual Regional Dust Frequency for Seven Regions of the United States

	Annual RADF (%)
South Central Region	0.61
Rocky Mountain Region	0.17
North Central Region	0.17
Pacific Coast Region	0.07
South Eastern Region	0.06
Great Lakes Region	0.04
North Eastern Region	0.01

Seasonal and Monthly Variations of Airborne Dust

The maximum frequency of suspended dust for the country occurs during the months of March, April and May. Figure 1 shows that during this period around 72% of the stations showed maximum monthly dust frequencies. In March, 41% of the stations indicated maximum monthly dust frequencies.

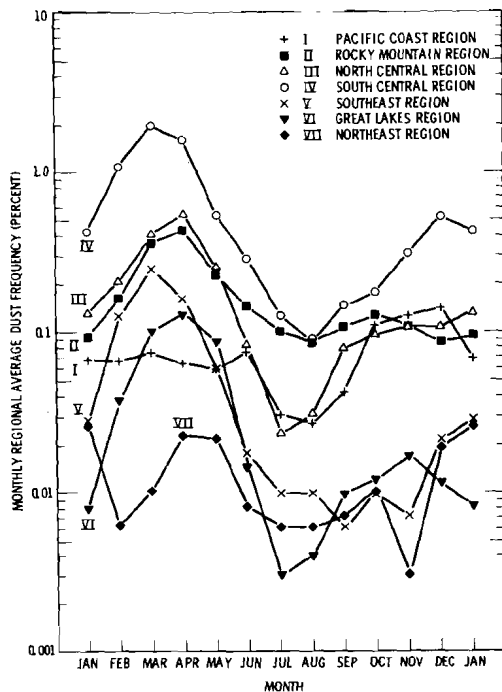


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FIGURE 1. The Percent of Observation Stations that have a Maximum Frequency of Airborne Dust During a Particular Month

A much smaller secondary maximum in dust frequency (4 to 5% of the stations) occurs during the months of September and October. This is primarily because several stations in the Pacific Coast and Rocky Mountain regions have their monthly maximum dust frequency during the fall months. This is the case in eastern Washington and the San Joaquin--Sacramento valleys of California.

A monthly RADF was computed for each of the seven regions of the country by Eq. (1). The results are presented in Figure 2. One of the most distinguishing characteristics of these results is the definite maximum in dust during the spring months for all areas except the Pacific Coast. In this latter case, the maximum occurs during the fall and early winter months. All the regions show a definite decrease during the summer months.



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FIGURE 2. Monthly Regional Average Dust Frequencies from Seven Defined Dust Regions in the Contiguous United States

Three, perhaps four, of the regions (South Central, Great Lakes, North Central, and Rocky Mountain), have a seasonal bimodal distribution in dust frequency with a primary maximum in the spring months and a secondary maximum in the fall or early winter months. Also, a number of individual stations in some of the other regions show bimodal dust frequency distributions.

The regional distribution of dust frequency for March, the month of greatest dustiness, is shown in Figure 3. The most significant aspect of the monthly dust frequency for March is the widespread increase of

dust in the east portion of the country. Apparently, this is due to "dust fronts" moving east from the South and North Central Regions and contributions from local dust sources. The regional dust frequency locations in the Western States are generally quasi-permanent and do not change much in position from month to month, but the frequency of dust is higher for the month of March.

DISCUSSION

The suspended dust potential of a region depends on several closely interrelated factors. Factors which may influence soil particle translocation are particle properties, surface properties, and meteorological factors.³ These factors and their components have been the subject of extensive research for several years.

It is important in assessing the dust potential of a region to consider the presence or absence of protective vegetative cover. When vegetative cover is disturbed by mechanical means, such as construction, plowing, etc., the regional dust potential is expected to increase. Since the early spring and fall months are usually when farmland is prepared for future crops, vast areas of disturbed soil become available for wind translocation. Apparently, agricultural activity may cause the high frequency of dustiness for the whole country during the spring months and possibly again during the fall months (as shown in Figures 1 and 2).

Generally, the study has shown, with some exceptions, that most regions of the continental United States are susceptible to suspended dust from wind translocation. Further details on the different regional characteristics of dust frequency would require additional studies on the principal factors which govern natural wind-caused dust suspension.



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FIGURE 3. Monthly Percentage Frequency of Dusty Hours from March Based on Hourly Observations from 343 Weather Observation Stations that Recorded Dust, Blowing Dust and Sand when Prevailing Visibility Was Less than 7 Miles (11 km). Shaded areas (N) represent no observations of dust.

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WIND PROFILES IN DUST STORMS

M. M. Orgill, G. A. Sehmel, and T. J. Bander

Vertical wind velocity profiles during dust storms in the Hanford Project were examined for possible multiphase flow effects of saltation and suspended sand and dust. The results to date are inconclusive.

INTRODUCTION

Vertical hourly-mean wind velocity profiles during severe dust storms in the Hanford project often exhibit typical nonlog-linear slopes. Our investigation was conducted to assess whether saltation and dust particles cause this type of velocity profile, and if multiphase flow effects may modify airflow and turbulent characteristics of the atmosphere surface boundary layer during severe dust storms.

The curvature of the wind profile during dust storms was anticipated to be due, in part, to saltation of sand and large dust particles. Saltation by alternate contact with the air and ground removes momentum from the air and represents a shear stress or drag on the wind. Hence, the lowest layers of air near the surface could be retarded as compared to higher levels. On the other hand, particles in true suspension, i.e., particles that very seldom make contact with the ground, must travel with the translocation speed of the wind and offer little resistance to the air.¹

PROCEDURE

Vertical wind velocity profiles were plotted using the hourly-averaged wind data from the 122 m meteorological tower for high wind episodes (>7 m/sec) with dust and without dust. Since the wind data were hourly averages, transient effects on the time scale of less than 1 hour were smoothed out or masked and only averaged features of the wind can be examined.

Vertical wind profiles for 64 high wind episodes in the Hanford project were plotted on semilog graph paper so the curvature of the velocity profile could be examined for linearity. These high wind episodes included dust storms from nearly all wind directions and high wind episodes with little apparent saltation or dust suspension (dust may have been observed but was recorded only in the remarks column of the observation). The dust storms had been determined from a previous study on the frequency of dust storms in the Hanford Project.²

RESULTS AND DISCUSSION

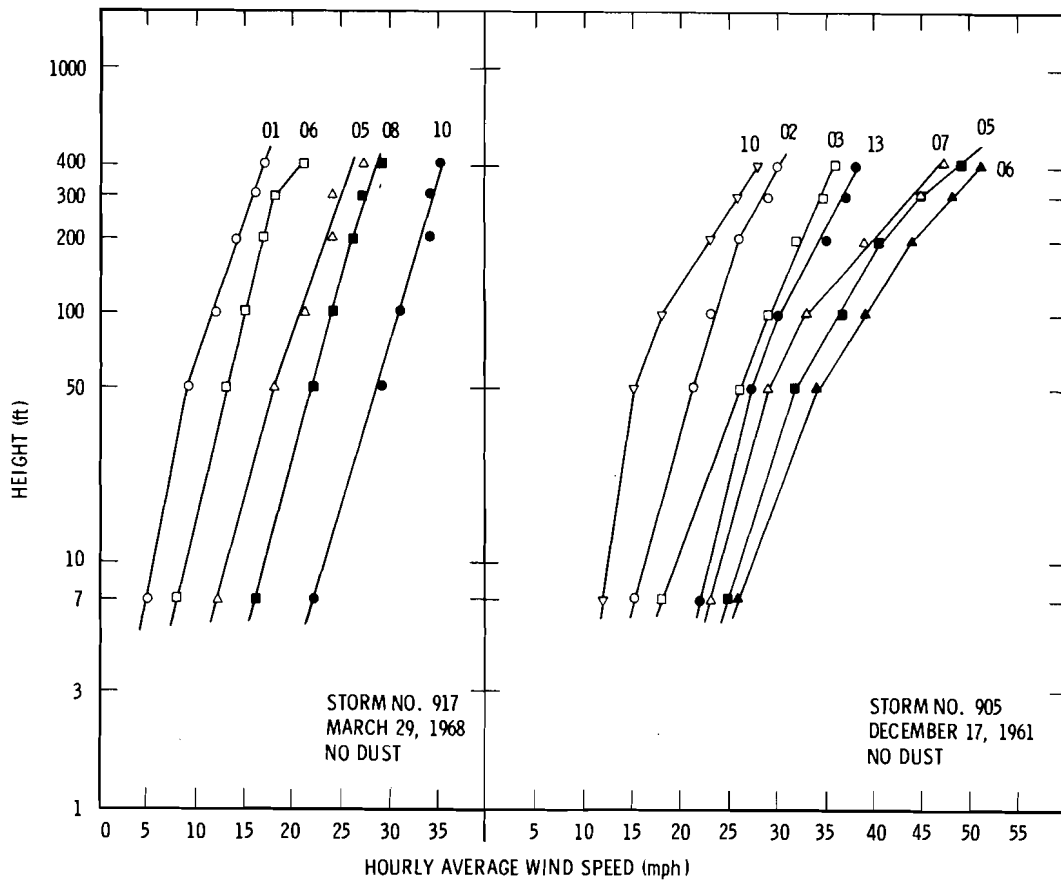
The vertical wind velocity profiles showed that log-linear profiles were present during dust storms and nondust storms. The nonlog-linear velocity profile was frequent during dust storms but not totally characteristic of all such storms. Figure 1 shows selected vertical wind profiles as a function of time during high wind episodes with little or no dust, that exhibited both types of wind profiles. Figure 2 shows examples of similar velocity profiles during dust storms. In this case, the prevailing visibility has been added as an index to the dustiness.

The vertical wind velocity profile for the May 9, 1966 dust storm shows a significant change in wind speed and shear near the 15 m (50 ft) level. This apparent change in velocity may indicate the height of the saltation level in the dust storm. It would be interesting to conduct experiments on dust-particle size distributions at 15 m (50 ft) and 30 m (100 ft) levels to determine the concentration of

significant-sized saltation particles at these heights in dust storms.

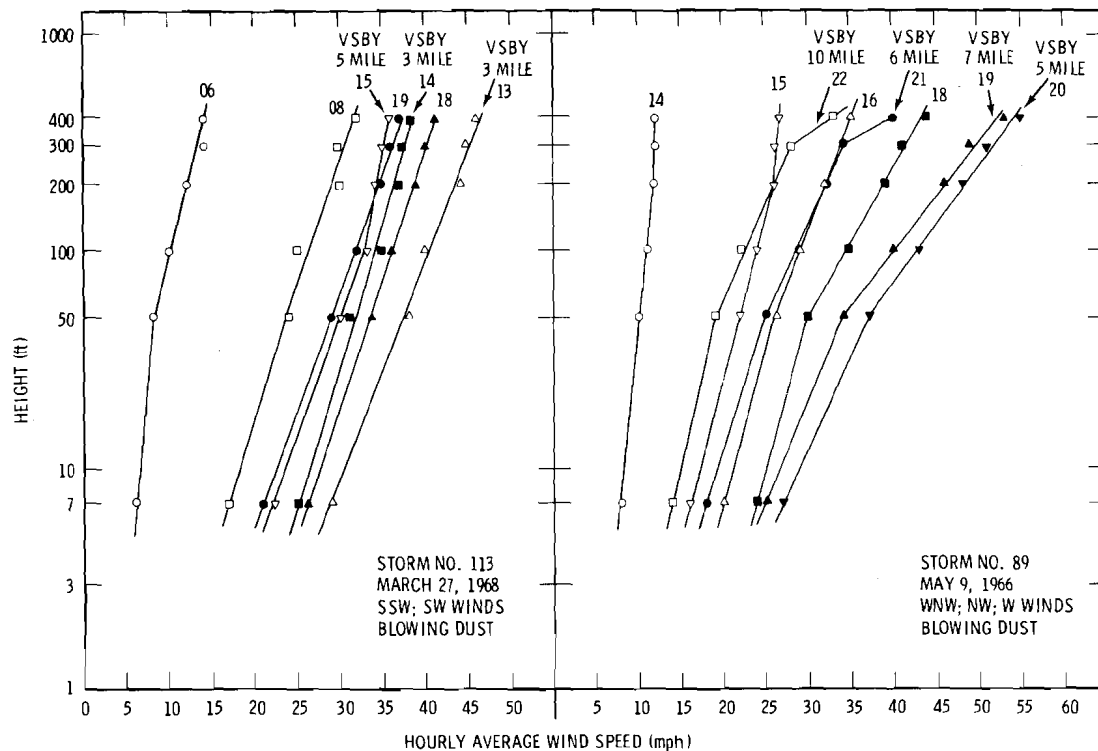
To date the results are inconclusive and will require further data

and analysis to show whether saltation is a possible causative factor in the production of non-log-linear vertical velocity profiles observed in dust storms.



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FIGURE 1. Selected Wind Profiles for Two High Wind Episodes with Little or no Dust. Profile for storm No. 917 is approximately logarithmic. Wind profile for storm No. 905 increases with height greater than logarithmic. Numbers are hours of day.



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FIGURE 2. Selected Wind Profiles for Two High Wind Episodes with Dust. Profile for storm No. 113 is approximately logarithmic. Wind profile for storm No. 89 increases with height greater than logarithmic. Numbers are hours of day. Prevailing visibility is an index of the dustiness.

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FREQUENCY OF DUST DEVILS IN THE HANFORD RESERVATION*

M. M. Orgill, J. M. Thorp and L. C. Schwendiman

Field observations in the Hanford Reservation have shown that about 20 dust devils per hour in a 1-mile² area may form under favorable wind, stability and surface conditions. Dust devil activity is more favorable during southwest-through-north wind directions during very unstable periods.

INTRODUCTION

A literature survey and limited field study¹ on dust devils sought to place in better perspective the frequency of occurrence and significance of dust devils and other convective vortices in the Hanford Reservation. We were particularly interested in the resuspension of contaminated soils and organic debris.

Dust devils are observed frequently in the reservation between April and October but little quantitative information is available on their frequency, location of occurrence, intensity, persistence and kinematics. However, theoretical, laboratory, and field investigations of convective vortices by other investigators have been undertaken in other locations and this information was reviewed and summarized during this interim study. Limited field observations of dust devils were made in the project area during the summer of 1974 to obtain better information on the frequency and persistence of these highly transient convective systems.

DUST DEVIL CHARACTERISTICS FROM LITERATURE SURVEY

The review of the literature on dust devils showed that in general five atmospheric elements necessary for their development and persistence are:

- A thick layer of unstable stratification and a strong superadiabatic layer ($< -0.10^\circ \text{ m}^{-1}$) in the surface boundary layer.
- A thermal disturbance or plume.
- Generation of large vorticity ($> 10^{-4} \text{ sec}^{-1}$) or rotary motion.
- Light to moderate wind speeds (0.5 to 5 ms^{-1}) or vertical wind shear.
- Vortex stability.

The existence and persistence of a dust devil appears to depend on whether a thermal plume or disturbance can acquire and maintain a critical amount of vorticity or rotation without friction, large-scale turbulence, or helical disturbances disrupting the concentrated vortex column. The fact that most dust devils seldom persist over 4 to 5 min suggests that vortex stability is very important to the genesis, persistence and dissipation of dust devils.²

Typical characteristics of dust devils are:

- Maximum frequency, 20 to 60 hr^{-1} ($\sim 1 \text{ mile}^2$ area).
- Duration, few seconds to 8 hours.
- Diameter, $< 3 \text{ m}$ to $> 30 \text{ m}$.
- Dust column height, $< 1.5 \text{ m}$ to 1200 m.
- Total height, up to 4800 m.

* Sponsored by the Atlantic Richfield Hanford Company.

- Tangential velocity, 1 ms^{-1} to 40 ms^{-1} .
- Vertical velocity, 1 ms^{-1} to 20 ms^{-1} .
- Dust loading, 11 - 60 particles cm^{-3} (density $\sim 5.3 \text{ gm m}^{-3}$).
- Dust particle sizes, $1 \text{ }\mu\text{m}$ to $350 \text{ }\mu\text{m}$.
- Potential gradient, up to 10^5 volts m^{-1} (negative).

FIELD RESULTS

Volunteer observers during the warmer months of 1974 observed over 36 dust devils in the Hanford Reservation. Dust devils were observed in several different locations in the project area, indicating that their development was generally ubiquitous. Out of the 36 dust devils, 3 had dust columns from 150 m to over 300 m in height, 4 had diameters 15 m to 30 m, and only one had an observed duration of over 10 min.

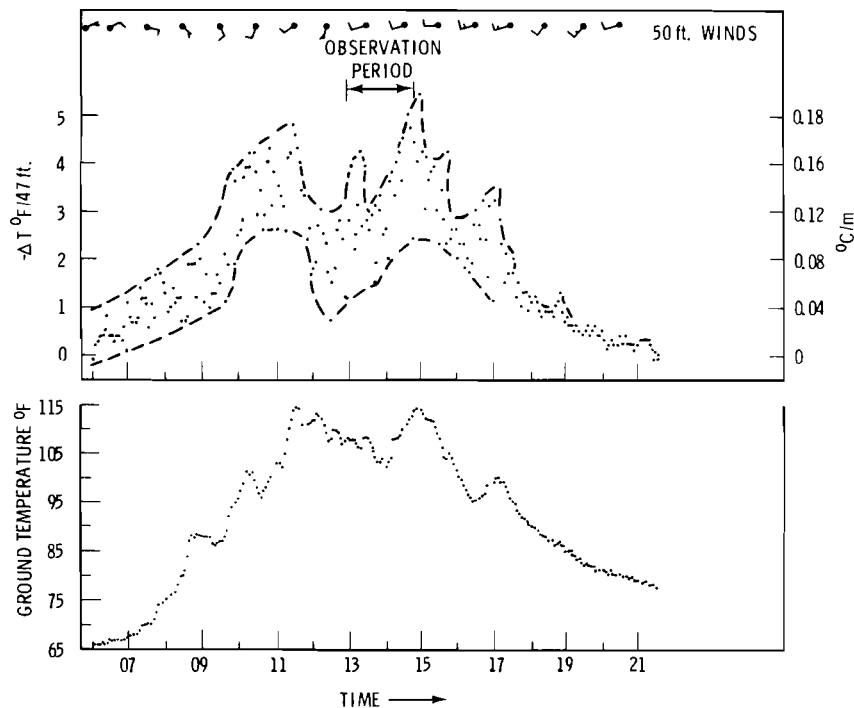
A special observation of the frequency of dust devils was conducted approximately 2.4 miles (3.8 km) northwest of the 300 Area in an area of about 1 mile². This area was involved in a grass-fire on July 15, 1974, and the surface and vegetation were partially blackened. The area consists of undulating rocky and sandy terrain. Observations were made on July 16, 17 and 18, and August 9, 1974. On one day, observations were taken in the 200-W area just north of the U-Pond.

A tabulation of observed dust devils according to approximate diameter of dust column is given in Table 1. The maximum observed dust devil frequency was about 20/hr. Duration of most dust devils was between 1 and 5 min with only one exceeding 15 min. On July 17, the total of 36 dust devils over a 2-hr period coincides generally with one of the unstable periods shown in Figure 1. The near-surface lapse rates at the 200-W tower varied between $-0.06^\circ\text{C m}^{-1}$ and $-0.21^\circ\text{C m}^{-1}$ during the period but due

TABLE 1. Dust Devil Observations 2.4 Miles Northwest of the 300 Area (Hanford) and 200-W According to Estimated Diameter

Date	Time Period (PST)	Location	Estimated Diameter ^(a) of Dust Column					Ambient Winds
			Very Small	Small	Medium	Large	Total	
16 July 1974	13:30-14:30	NW-300A	2	3	13	1	19	NE 5-10 mph
17 July 1974	13:00-15:00	NW-300A	3	13	17	3	36	SW-WSW 5-10 Gusts 15-20
18 July 1974	13:10-15:13	NW-300A	5	15	17	4	41	W-WNW 2-10 (Periods of east wind)
24 July 1974	12:30-14:30	200 W	1	3	1	0	5	Variable (Periods of SSE-4 mph)
9 Aug 1974	13:00-14:04	NW-300A	0	5	3	0	8	SE 3-10 mph
Totals			11	39	51	8	109	
%			10.1	35.8	46.8	7.3		

a. Very small <5 ft; Small 5- <10 ft; Medium 10-50 ft; Large 50-100 ft.



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FIGURE 1. Temporal Variation of the Magnitude of Superadiabatic Lapse Rates and Ground Temperatures at the 200-W Meteorological Tower, Hanford Project During July 17, 1974. Dust devils were observed near 300 Area during observation period.

to the sparse vegetation and dark surface at the burned area, near-surface lapse rates may have been greater (negatively) than at the tower located approximately 30 km northwest of the burned area.

The dust devil frequency occurred in bursts of activity, with 3 or more dust devils appearing near the same time. Then the activity would slack off with periods of little activity for several minutes. Periods of higher dust devil activity generally coincided with westerly to northerly winds (Table 1) which correlated well with the findings of a climatological study between local wind directions and highly unstable lapse rates ($<0.15^{\circ}\text{C m}^{-1}$).¹ The reason for these correlations is unknown at this point. However, westerly environmental flow may be more favorable for dust devil activity due to topographic production of mesoscale vorticity and the development of highly unstable lapse rates.

CONCLUSIONS

Limited field observations in the Hanford Reservation showed that 20 (visible) dust devils per hour ($\sim 1 \text{ mile}^2$) may form under favorable wind, stability and surface conditions. Light to moderate winds from the southwest through north sectors may be more favorable for dust devil activity than winds from the east sectors. Additional field work is needed to verify this tentative relationship.

Normal summertime dust devil activity may be a significant resuspension mechanism at times. This type of "convective" wind translocation mechanism, especially through "leap-frog" and cumulus transport modes may move tons of contaminated soil and debris downwind several miles from contaminated sites over a period of time. Additional research is needed to critically evaluate the role of dust devils as a "convective" wind translocation mechanism.

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RESUSPENSION OF RADIOACTIVE ISOTOPES INDUCED BY BURNING FOREST LOGGING SLASH

G. A. Sehmel and M. M. Orgill

Airborne particulates from burning forest logging slash were sampled with an aircraft-mounted air filter. Radioactive isotope concentrations in air samples collected from the smoke plume were compared to ambient airborne background concentrations. Smoke plume radioactive isotope concentrations for ^{155}Eu and ^{137}Cs were >9 and 22 times ambient background levels, respectively.

INTRODUCTION

In the fall of 1974 smoke plumes from cropland fires were sampled by an aircraft for airborne radioactive isotopes. The purpose was to determine whether low-level radioactive surface contamination from prior dry and wet deposition could be resuspended by fire.¹ The results suggested that smoke plumes from cropland fires did contain more radioactivity than background air concentrations.

The success of this initial experiment justified extending the effort to sampling smoke plumes from larger fires such as a forest fire. The experiment on a larger fire might provide additional confirmation that fires can resuspend low-level radioactive isotopes into the atmosphere at greater concentrations than ambient.

EXPERIMENTAL PROCEDURE

In the late summer and early fall active wild fires or prescribed for-

est logging slash burns are an occasional occurrence in the Pacific Northwest forest reserves. On September 6, 1975, the U.S. Forest Service planned to burn approximately 350 acres of logging slash about 15 miles (24 km) west-southwest of Mt. St. Helens, in the Gifford Pinchot National Forest. Ignition time was 1300, and by 1500 a dense smoke plume had formed and was slowly moving toward but slightly to the north of Mt. St. Helens (east-northeast). Figure 1 shows the extent of the smoke plume during the latter part of the sampling period. The fire front at this time was estimated to be about 1 km. The altitude of the smoke plume was estimated between 1500 and 3000 m (msl).

Prior to sampling the smoke plume by aircraft (Cessna 411), a background air filter sample was obtained approximately 10 miles (16 km) west of the fire site, between 0930 and 1200 at approximately 1400 m (msl). Sampling of the smoke plume commenced at 1506 and continued until 1736. Traverses were made through the smoke plume between 2000 and 2200 m (msl).



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FIGURE 1. Forest Logging Slash Burn about 15 m (24 km)
West-Southwest of Mt. St. Helens, State of Washington,
September 6, 1975

Samples were collected on IPC filter paper supported in an aircraft-mounted filter holder. Ram air produced an average air sampling flow rate of approximately $25 \text{ m}^3/\text{min}$. Air filter samples were taken over a 150-min period. The flow rate through the background and smoke filter was measured with an inline, negligible-pressure-drop vane flowmeter.* Flow rates were measured after the sampling period and showed that the smoke filter had decreased the flow rate by about $3 \text{ m}^3/\text{min}$ because of the accumulation of particulate matter. The flow rates were $26 \text{ m}^3/\text{min}$ for the background filter and $23 \text{ m}^3/\text{min}^{-1}$ for the smoke filter.

* PR-2 flow sensor: previously manufactured but no longer available from Sci-Med, Inc., Engineering Dept., Minneapolis, MN.

Since the smoke sample had taken a period of time to accumulate particulate matter it was assumed that the flow rate during the smoke filter sample was the average of the two flow rates or about $25 \text{ m}^3 \text{ min}$.

Filter samples were analyzed by nondestructive gamma ray spectrometry for 14 different radioactive isotopes. All 14 isotopes were identified on the filters in varying concentrations.

RESULTS

Concentrations of the radioactive isotopes were found to be greater in the smoke filter sample than in the ambient background filter sample. Eight of the radioactive isotopes had concentrations 2 or more times higher in the smoke plume than in the background air.

Table 1 shows the concentrations for each radioactive isotope and normalized airborne activity concentration ratios. These ratios were calculated from Eq. (1).

$$\text{Activity Concentration Ratio} = \frac{\left[S - BG \left(\frac{T_o}{T_{BG}} \right) \right]}{BG} \cdot \left[\frac{F_{BG} (T_{BG})}{F_s (T_s)} \right], \quad (1)$$

where S represents smoke concentration (dpm); BG is background concentrations (dpm); T_o is time out of smoke plume during sampling; T_{BG} is background sampling time; T_s is smoke plume sampling time; T_{BG} is average flow rate during background sampling; and F_s is average flow rate during smoke sampling.

In calculating the normalized concentration ratios, consideration was given to the fact that the aircraft was not in the smoke plume during the total sampling period (150 min) because of safety requirements. The aircraft was in the smoke plume about 120 min of the 150 min sampling period.

Concentrations of ^{137}Cs in the smoke plume were approximately 22 times the concentration in ambient background air. Airborne concentration for ^{137}Cs were $1.02 \times 10^{-15} \mu\text{Ci}/\text{cm}^3$ for background air and $2.29 \times 10^{-14} \mu\text{Ci}/\text{cm}^3$ for the smoke sample. Comparison of these concentrations with the 168-hr MPC for ^{137}Cs ($2 \times 10^{-8} \mu\text{Ci}/\text{cm}^3$) shows that smoke and background concentrations were a minute fraction of MPC values. This was also true for the remaining isotopes.

Measured isotope concentrations in the smoke were probably significantly

TABLE 1. Radionuclide Concentration and Ratios in Air Filter Samples

Isotope	Background (dpm/sample)	Smoke Sample (dpm/sample)	Normalized Activity Concentration Ratio
^7Be	1502 ± 30	1729 ± 32	1.2
^{54}Mn	0.52 ± 0.34	1.6 ± 0.5	3.8
^{60}Co	<0.92	<0.99	--
^{95}Zr	4.6 ± 1.1	7.5 ± 1.6	1.9
^{95}Nb	9.3 ± 0.98	14.4 ± 1.3	1.8
^{103}Ru	<0.67	<0.92	--
^{106}Ru	13.2 ± 6	31.5 ± 6.6	2.9
^{125}Sb	2.9 ± 1	10.6 ± 1.5	4.6
^{137}Cs	9 ± 0.8	153.6 ± 3.2	22.2
^{141}Ce	0.76 ± 0.5	0.91 ± 0.71	--
^{144}Ce	41 ± 2.5	99.8 ± 3.6	2.9
^{154}Eu	<3.2	<2.6	--
^{155}Eu	<0.88	6.4 ± 0.94	>9
^{241}Am	<1.8	<2.3	--

Error bar limits are at 1σ

Detection limit = 2σ of the interference

less than maximum possible concentrations. If the aircraft could have remained in the dense smoke plume for the full 150 min sampling period, higher concentrations might have been the result. Thus, the measured isotope concentrations were probably on the conservative side.

CONCLUSIONS

Surface radioactive contamination which was deposited earlier by wet

and dry deposition processes can be released by fires over croplands and forested areas at concentrations greater than ambient background. The possibility exists that the downwind transport and potential inhalation hazard could be significantly increased by fires in severely contaminated vegetation. Further field experiments may be advisable in order to confirm and evaluate the potential hazards of fire released radioactive isotopes from vegetation.

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

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

The Metropolitan Meteorological Experiment (METROMEX) is a study by several organizations of inadvertent weather modification, particularly precipitation changes, caused by urban-industrial effects in the St. Louis metropolitan area. Two main experiments are being conducted by the Earth and Planetary Chemistry Section during METROMEX. The study reported here measured trace elements, sulfates, SO_2 , trichlorofluoromethane, and CO upwind, in, and up to 130 miles downwind of St. Louis to determine the atmospheric concentrations of these contaminants produced by the St. Louis metropolitan area, and to determine the decreased concentrations downwind of the city that result from atmospheric mixing, dry deposition, and, in the case of SO_2 , chemical conversion processes. Results showed contaminant concentrations generally decreased rapidly in the first 10 miles downwind of the center of St. Louis as a result of mixing through the mixed layer and then decreased much more slowly further downwind. The trichlorofluoromethane and CO concentrations measured in 1975 indicate that atmospheric mixing decreased the concentrations by only about 35% from 40-120 miles downwind. The Pb concentrations indicated a 40% decrease due to dry deposition at this distance on August 4, 1975. The SO_2 concentrations reached a maximum 40 miles downwind of the city, presumably because SO_2 from stacks did not reach ground level immediately. Between 50 and 120 miles downwind the SO_2 concentration decreased by a factor of 10, probably because of its reaction with vegetation and the earth's surface.

INTRODUCTION

The St. Louis metropolitan area injects large quantities of pollutant materials into the atmosphere. These contaminants are carried large distances by the prevailing winds and may affect regions considerably downwind of St. Louis. The downwind decreases in the atmospheric concentra-

tions of these contaminants with time and distance depend upon atmospheric mixing processes, wet and dry deposition on the earth's surface, and in some cases, chemical conversion processes. The relative importance of mixing processes versus removal processes in decreasing the contaminant concentrations downwind of St. Louis may be determined by comparing the

concentrations of nonconservative contaminants with those of conservative contaminants. Trichlorofluoromethane, which is released into the atmosphere primarily by aerosol spray cans, is essentially conservative, having a residence time in the atmosphere of several years. CO may also be relatively conservative.

EXPERIMENTAL

The concentrations of trace elements, sulfates, SO₂, CO, and trichlorofluoromethane are being measured in air samples collected upwind, in, and downwind of St. Louis on August 3-9, 1975 to determine the atmospheric concentrations of these contaminants produced by St. Louis, and to determine the rates at which the concentrations decrease downwind of St. Louis. Aerosol samples were collected for trace element analysis using HiVol pumps to draw air through 11 cm diameter Whatman-41 filter paper at a flow rate of about 0.6 m³ min⁻¹. Samples were collected for SO₂ analysis by bubbling air through sodium tetrachloromercurate solutions at a rate of 1 l/min. Air samples for trichlorofluoromethane and CO analysis were obtained using a sampler which collects whole air by slowly drawing a piston up a cylinder. These samples were transferred to evacuated 460 cm³ gas cylinders for storage.

On each sampling day one set of air samples was collected upwind of St. Louis. Sequential samples were collected on the roof of the 11-story Federal Building in downtown St. Louis within a mile or so of the St. Louis Gateway Arch. Six people equipped with mobile air sampling equipment collected air samples from 10-130 miles downwind of the St. Louis Arch. Each person collected one set of 1-hr air samples either 10 or 20 miles downwind of the Arch, then moved several more miles downwind to collect a set of 2-hr air samples, and then moved downwind again to collect another 2-hr set of samples. The distance they moved downwind between samples was chosen so that their average rate of travel downwind was about the same as the rate at which the air traveled. In this manner, a grid of air sampling stations was set up which moved downwind with the air. Air sampling was

begun at around 1100 each day and continued throughout the afternoon.

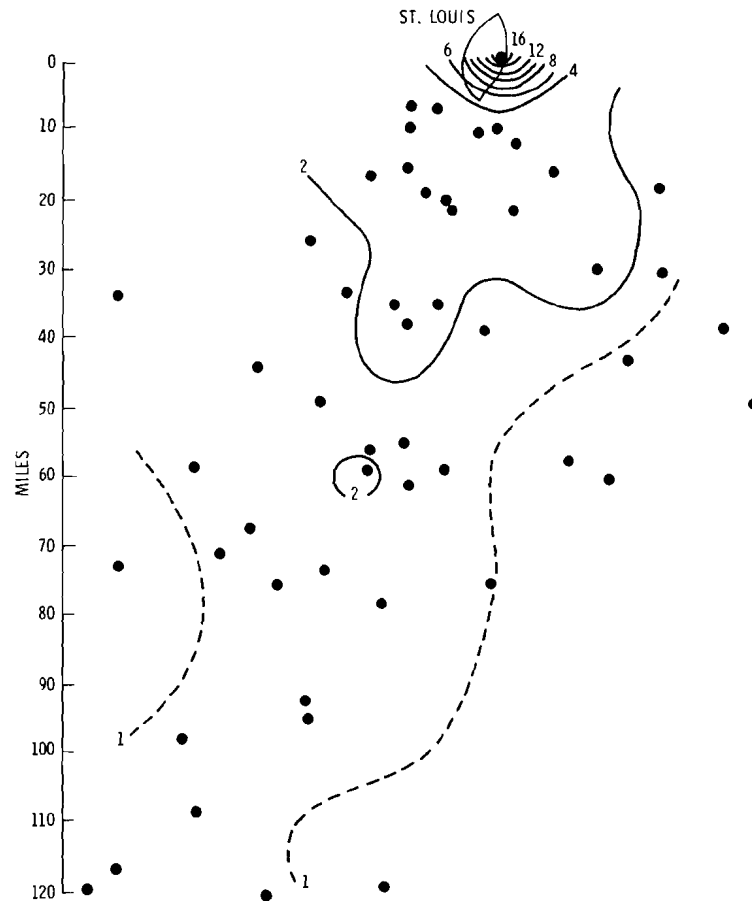
The Battelle Cessna-411 aircraft was used to collect air samples at three elevations in the mixed layer above the downwind air sampling stations on August 3, 4, 5, 7 and 8. Each sample was collected over a 45-min period while the Cessna was flying a racetrack pattern perpendicular to the wind. One vertical profile was taken on August 5, and profiles were taken at two distances downwind on the other days.

RESULTS

The analysis of the samples has not been completed, so results will be presented for only some of the sampling days in this report. On August 3 through 6 the wind was primarily from the north at about 8-10 mph and the contaminant concentrations for the 4 days were plotted together. Since the wind direction varied somewhat from day to day, the coordinate system for each day was rotated until the prevailing wind direction was equal to the average wind direction for the 4 days.

Trichlorofluoromethane

The concentrations of the conservative gas trichlorofluoromethane for August 3 through 6, 1975 are plotted in Figure 1. The concentrations decreased by a factor of around five between the Federal Building in downtown St. Louis and the samples collected 10 miles downwind of the Arch, as a result of vertical mixing through the mixed layer, and possibly horizontal mixing. Farther downwind the concentrations decreased more slowly. The concentrations 130 miles downwind averaged 65% of the concentrations 40 miles downwind. These results indicate that atmospheric mixing will decrease contaminant concentrations rapidly for the first 10 or 20 miles downwind of the source, as the contaminant becomes mixed through the mixing layer, but that beyond that distance mixing will only slowly decrease concentration. In general, the chemically nonreactive anthropogenic contaminants measured downwind of St. Louis have shown a rapid decrease in concentration followed by much slower decrease.



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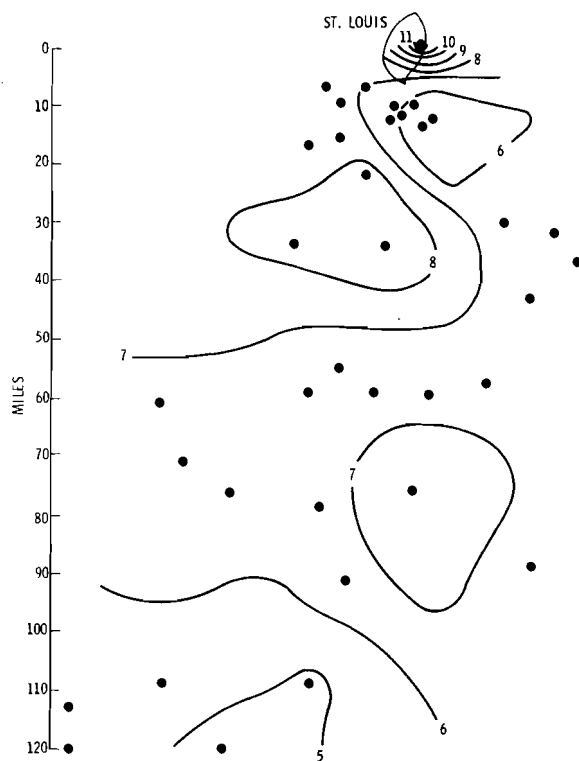
FIGURE 1. Trichlorofluoromethane Concentration in ppb on August 3-6, 1975

Carbon Monoxide

The CO concentrations of August 3 through 6 are plotted in Figure 2. The measured CO concentrations downwind of St. Louis perhaps should not be taken too seriously due to possible contamination from CO emitted by the gasoline generators used to power the HiVol samplers. Even though care was taken to collect the samples for CO analysis upwind of the generators, several of the samples were obviously contaminated and had to be disregarded. The CO concentrations also showed a very rapid decrease between downtown and 10 miles downwind followed by a much slower decrease. The average concentration decreased 35% between 40 miles downwind and 130 miles downwind.

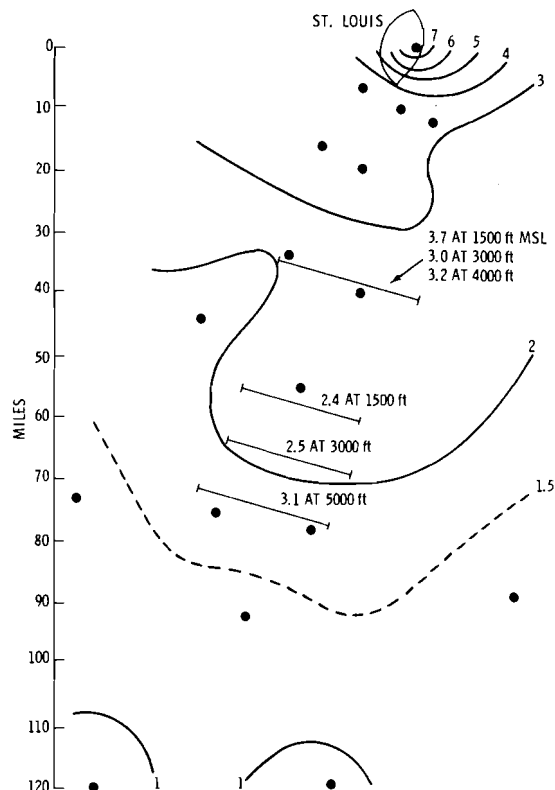
Lead

The Pb concentrations on August 4 are plotted in Figure 3. This is the only day on which the analysis for Pb is complete. Pb is introduced into the atmosphere primarily by automobile exhaust. The Pb concentrations measured by aircraft are also shown in the figure. The Pb concentrations decreased rapidly between the Federal Building and 10 miles downwind of the Arch, presumably as a result of atmospheric mixing. Beyond 10 miles the concentrations decreased more slowly. The Pb concentrations decreased 60% between 40 miles downwind and 130 miles downwind. The trichlorofluoromethane and CO analyses indicate that only about a 35% decrease can be accounted for by dilution due to atmospheric mixing, leaving a 40% decrease



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FIGURE 2. Carbon Monoxide Concentration in ppm on August 3-6, 1975



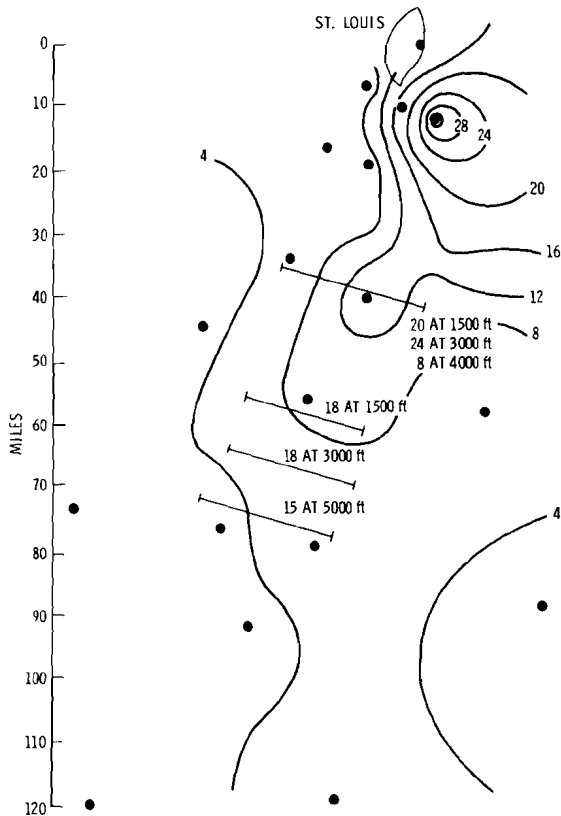
Neg 760035-1

FIGURE 3. Lead Concentration in 10^{-7} g/m³ on August 4, 1975

due to dry deposition. Pb measurements carried out in 1973 indicate that 70% of the Pb is present on particles smaller than 0.3μ in diameter and only 10% is present on particles larger than 3μ , so settling cannot be a major cause of the Pb concentration decreases. Therefore, most of the Pb concentration decrease between 30 and 120 miles downwind must have been due to dry deposition resulting from diffusion. Assuming that the Pb concentration was constant through a 4000-ft mixed layer, and that the wind speed was 8 mph, then a deposition velocity of 1.2 cm sec^{-1} would be required to decrease the Pb concentration by 40% in 90 miles. The Pb concentrations measured by aircraft tended to be somewhat higher than the ground level concentrations, indicating that deposition at the earth's surface lowered the surface level concentrations more than the higher elevation concentrations. Therefore the above calculated deposition velocity is probably greater than the true deposition velocity.

Zinc

The Zn concentrations measured at ground level and by aircraft on August 4 are plotted in Figure 4. The Zn in the atmosphere is primarily anthropogenic. The Zn concentrations in one of the samples collected 12 miles downwind of St. Louis was nearly three times the concentration measured in downtown St. Louis, suggesting a source of Zn either downwind of the downtown sampling site or east of the site. The downtown Zn concentrations were not significantly greater than the average of two other concentrations measured 11 miles downwind of the Arch, indicating that there were no major Zn sources within a few miles upwind of the downtown sampling site. The Zn concentration decreased by a factor of three from 40-120 miles downwind of the Arch, a rate of decrease about equal to that of Pb. For some reason the Zn concentrations measured by aircraft were considerably higher than those measured at ground level.

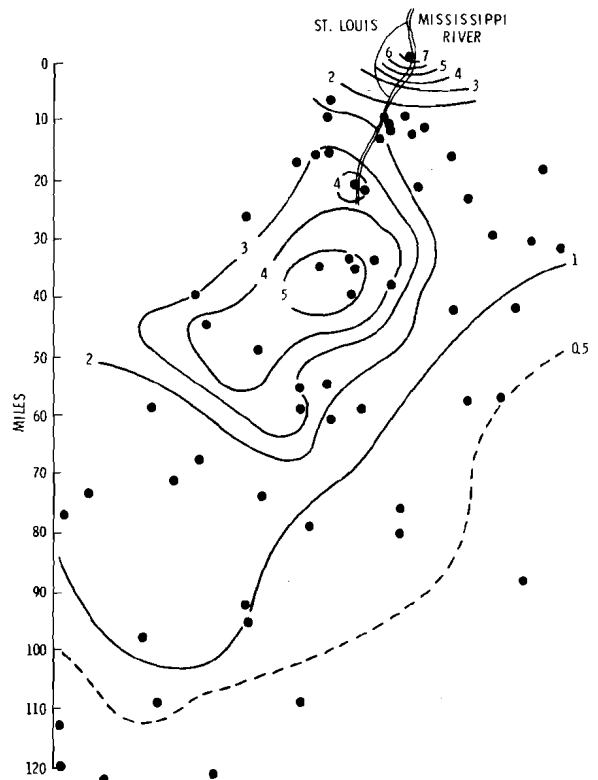


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FIGURE 4. Zinc Concentration in 10^{-8} g/m^3 on August 4, 1975 (upwind concentration = 1.8)

Sulfur Dioxide

The SO_2 concentrations for August 3-6 are plotted in Figure 5. The SO_2 concentrations decreased from an average of 79 ppb at the Federal Building to about 20 ppb 10 miles downwind of the Arch and then increased again to about 50 ppb 40 miles downwind of the Arch. Probably the low SO_2 concentrations 10 miles downwind of the Arch resulted from the fact that SO_2 released from stacks passed over these sampling locations and only reached ground level farther downwind. The sampling location on top of the Federal Building may have sampled SO_2 that had not yet reached ground level. The SO_2 concentrations measured in samples collected by aircraft 50 miles downwind of the Arch on August 5 were 101, 78, and 89 ppb at 1500, 3000, and 5000 ft above sea level, respectively; significantly higher than the ground level concentrations. Beyond 50 miles the SO_2



Neg 760035-8

FIGURE 5. Sulfur Dioxide Concentration in $\text{ppb} \times 10$ on August 3-6, 1975

concentrations at ground level decreased rapidly, to a value of 6 ppb 120 miles downwind of the Arch. This is a much faster decrease than shown by Pb and presumably results from the reaction of SO_2 with vegetation and the earth's surface. The sulfate measurements indicate that only about 10% of the SO_2 concentration decrease could be accounted for by the conversion of SO_2 to sulfates in the atmosphere.

Sulfates

The sulfate concentrations on August 3 and 4 are plotted in Figure 6 in units of $\mu\text{g S/m}^3$ air. The samples for the other sampling days have not been analyzed for sulfates as yet. The sulfate concentration, like the SO_2 concentration, increased to a maximum 40 miles downwind of the Arch and then decreased. However, the sulfate concentration at the Arch was about the same as that 10 miles

other compounds such as Pb, for comparison purposes). A low-volume flow system is provided for deposition tubes for collection of SO_2 . These systems represent some modification of a preliminary system developed for the Environmental Protection Agency. That system was tested in a 1975 experimental program downwind of the Centralia, Washington, fossil fuel plant. The present systems have been designed to minimize the possibility of sample contamination or modification due to the sampling method. All collectors are mounted on the meteorological tower. Additional systems were developed to more precisely measure total volume flow. Carefully measured flow, temperature and pressure with less than 1% error in combination with positive displacement flow meters allows flow volumes between levels to be assessed with a relative error estimated to be 1% or less. The meteorological data are recorded at high frequency on industrial-standard magnetic tape. The line pressures are manually recorded using manometers. Every effort has been made to improve the measurement accuracy.

Aerosol samples obtained will be analyzed by a specially-developed x-ray fluorescence procedure designed to minimize the relative error between samples; insuring as accurate gradients as are possible.

The deposition tubes will be used for SO_2 samples. Analysis of the meteorological profile structure will allow the determination of deposition velocities. We believe that the measurement precision is sufficient to provide the necessary accuracy to determine significant deposition rates for the pollutant species under consideration. Development of an SO_2 fluxmeter is being considered to serve as a totally independent and absolutely direct method of measuring deposition.

The application of the profile method of determining deposition rates for other pollutant species was found in the earlier study to require other specialized techniques. In particular the determination of the gradients of highly reactive gases (such as O_3 and NO) was found to be best performed using a vertically roving intake line with real time analysis. An effort has been initiated to develop this capability further.

Verification of results from the set of pilot experiments will allow the planning of experiments to assess the dry removal rates of the S effluents from use of western fossil fuels. Specification of the removal rates will permit assessment of the air quality terrestrial impacts associated with the development and utilization of these resources.

POLLUTANT SCAVENGING

The removal of pollutants by clouds and precipitation is a major factor in determining budgets of pollutants distributed on a regional scale. The quantity and patterns of pollutants delivered to sensitive ecosystems as well as the distributions remaining as inhalation hazards in scavenged air can only be defined by a thorough investigation of precipitation scavenging processes.

The scavenging program at the Pacific Northwest Laboratory has progressed according to the direction given in the previous annual report. Specifically, our research has emphasized the three following areas:

- In-cloud scavenging in frontal storms
- In-cloud scavenging in convective storms
- Below-cloud scavenging of submicron particulates

The following reports indicate progress made during the past year in these fields of investigation. Our reports also reflect two aspects of the developing program, which we expect to become more prominent during coming years. The first of these is the greater utilization of diagnostic models for interpretation of experimental data—especially with regard to in-cloud scavenging processes. The second feature is the evolving close level of cooperation with other research agencies. This year, the input from the Air Resources Laboratory/NOAA and the Wave Propagation Laboratory/NOAA has been particularly important in this regard, and we feel that the continuation of such interactions will be highly beneficial to the resolution of the complex problems associated with the precipitation scavenging process—especially as the program is directed more closely to the MAP3S objectives.

- **THEORETICAL STUDIES IN ATMOSPHERIC SCIENCES**
- **PRECIPITATION SCAVENGING**
- **PRECIPITATION SCAVENGING IN MAP3S**

COMMENTS ON DIFFUSIVE DEPOSITION AT SMALL FOURIER NUMBERS*

W. G. N. Slinn

Expanding the Laplace transformed equations for diffusive deposition yields asymptotic solutions useful for small Fourier numbers.

Further to the stimulating approach used by Pich¹ to calculate the diffusive deposition of particles on various surfaces at small values of $Fo = Dt/a^2$, it may be of some interest to demonstrate how solutions useful for small Fo can be found directly from the diffusion equation using the methods discussed, for example, in Carslaw and Jaeger.²

For the case internal to a sphere, a Laplace transform of Pich's Eq. (2) (hereafter his equations will be referenced by number, e.g., P-2) leads to

$$\hat{n}(r,s) = \frac{n_0}{s} \left[1 - \frac{a}{r} \frac{\sinh qr}{\sinh qa} \right] \quad (1)$$

where $q = (s/D)^{1/2}$, s is the Laplace transform parameter, and where other notation is, and will be, as in Pich although r will be used instead of ζ for the radial coordinate. Inverting (1) (e.g., see Erdélyi³) yields P-3 which as Pich notes, is not convenient for evaluations at small Fo since the series converges but slowly. To obtain a more convenient small Fo solution, (1) is expanded for large s and then inverted term by term. The result is

$$\frac{n(r,t)}{n_0} = 1 - \frac{a}{r} \sum_{k=0}^{\infty} \left[\operatorname{erfc} \left\{ \frac{(2k+1)a - r}{2(Dt)^{1/2}} \right\} - \operatorname{erfc} \left\{ \frac{(2k+1)a + r}{2(Dt)^{1/2}} \right\} \right] \quad (2)$$

which converges for all Fo and for small Fo requires the retention of only a few terms to obtain an accurate approximation.

To illustrate the last statement a comparison will be made with numerical values for $\hat{n}/n_0$ given by Pich. From P-4 and P-10 (or, more directly, by averaging P-2 over the sphere) one obtains

$$\frac{d\bar{n}}{dt} = \frac{3}{a} D \frac{\partial n}{\partial r} \Big|_{r=a} \quad (3)$$

A Laplace transform of (3) and the use of (1) leads to

$$\frac{\hat{n}(s)}{n_0} = \frac{1}{s} + \frac{3D}{as^2} \left[\frac{1}{a} - q \frac{\cosh qa}{\sinh qa} \right] \quad (4)$$

Expanding (4) for large s , inverting and then expanding the complementary error functions for small Fo yield the asymptotic expansion

* To be published in Atmospheric Environment, 1976.

$$\frac{\bar{n}}{n_0} \sim 1 - \frac{6}{\sqrt{\pi}} Fo^{1/2} + 3 Fo + \dots \quad (5)$$

For $Fo = (10 \pi^2)^{-1}$, Pich gives the exact solution as 68.98% while P-31 yields 69.65%. For comparison, (5) yields 68.97%.

Similarly, for the case of deposition inside a cylinder one obtains from P-16

$$\hat{n}(r,s) = \frac{n_0}{s} \left[1 - \frac{I_0(qr)}{I_0(qa)} \right] \quad (6)$$

where I_0 is a modified Bessel function.⁴ Then solving

$$\frac{d\bar{n}}{dt} = \frac{2}{a} D \frac{\partial n}{\partial r} \bigg|_{r=a} \quad (7)$$

in a manner similar to the case for a sphere leads to

$$1 - \frac{\bar{n}(t)}{n_0} \sim \frac{4}{\sqrt{\pi}} Fo^{1/2} - Fo - \frac{11}{24} \frac{Fo^{3/2}}{\sqrt{\pi}} + \dots \quad (8)$$

For the problem of deposition external to a sphere I_0 in (6) is replaced by the modified Bessel function K_0 and one finds that the only change in (8) is the replacement of the term $- Fo$ by $+ Fo$. The utility of this result is demonstrated in Table 1 (which is Pich's Table 1 with an additional column). However this comparison should not distract attention from the simplicity and presumed applicability of Pich's method for more complicated geometries.

TABLE 1. See Pich's Table 1

Fo	Exact	P-38	(8) (a)
0.001	0.072	0.073	0.072
0.005	0.164	0.166	0.165
0.01	0.235	0.238	0.235
0.02	0.337	0.345	0.338
0.05	0.550	0.568	0.552
0.1	0.805	0.841	0.806
0.2	1.190	1.264	1.186
0.4	1.632	1.937	1.762

a. With the change to external deposition, see text.

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PRECIPITATION SCAVENGING OF MONODISPERSE AEROSOL PARTICLES*

W. G. N. Slinn

Formulas are suggested for the approximate description of below- and in-cloud, rain and snow scavenging of monodisperse aerosol particles.

This note encourages caution against the immediate acceptance of the particle/snow scavenging rates recently published by Graedel and Franey¹ and presents alternatives. Their data, similar to unpublished data I have obtained, admit a number of plausible interpretations other than theirs of snow scavenging from an (implicitly assumed) stationary air mass. For example:

1. The data could reflect the relatively rapid passage of comparatively clean air which has been scavenged for an undertermined but presumably long time period;
2. With the onset of snow the wind might have shifted and aerosol particles from different sources were subsequently sampled;
3. The snow cover could cause a reduction in the flux of particles resuspended from various surfaces;
4. With the onset of snow, automobile traffic might have diminished;
5. If the humidity increased with the onset of snow, then condensational growth of particles larger than about 0.1 μm could contribute to their apparent removal.

With respect to the theoretical considerations in Graedel and Franey's letter, I have presented in detail elsewhere² my opinion that there is little evidence to support the assumption that diffusion and electrical processes are dominant for the

below-cloud scavenging of particles larger than 0.1 μm .

The alternatives I suggest, for both in- and below-cloud scavenging of monodisperse particles, are the following. For the rain scavenging rate

$$\Lambda = \frac{p}{2R_m} E(a, R_m) \quad (1)$$

where the collection efficiency is given by the semi-empirical expression

$$E = \frac{4}{Pe_m} \left(1 + 0.4 Re_m^{1/2} Sc^{1/3} \right) + 4\kappa \left[\kappa + \frac{1 + V\kappa}{1 + V Re_m^{-1/2}} \right] + \left(\frac{s-s^*}{s+c} \right)^{3/2} \quad (2)$$

For the corresponding snow-scavenging rate I suggest, with less confidence,

$$\Sigma = g \frac{\rho_w}{\rho_a} \frac{p}{\langle v_t \rangle^2} E(a, \lambda) \quad (3)$$

where the collection efficiency is

* To be published in Geophys. Res. Letters, 1976.

$$E(a, \ell) = \frac{4}{Pe_\ell} \left(1 + 0.4 Re_\ell^{1/2} Sc^{1/3} \right) + \left[1 - \exp \left\{ - \left(1 + Re_\ell^{1/2} \right) \frac{a^2}{\ell^2} \right\} \right] + \left(\frac{s-s_*}{s+c} \right)^{3/2} \quad (4)$$

In these formulae the symbols have the following definitions:

- Λ = the rain scavenging rate
- p = in- or below-cloud precipitation rate in rainwater equivalent
- R_m = the volume-mean drop radius
- a = particle radius
- Pe = $Re Sc$, the Péclet number
- Re_L = Lv/ν , the Reynolds number with the appropriate length scale $L = R_m$ or ℓ and appropriate terminal velocity $v = \bar{v}_t$ or $\langle v_t \rangle$
- Sc = ν/D , the Schmidt number
- ν = kinematic viscosity of air
- D = molecular diffusion coefficient for a particle of radius a
- $\bar{v}_t, \langle v_t \rangle$ = terminal velocity of volume mean drop size or average snowflake type and size, respectively
- ℓ = empirical constant characterizing the length scale of the collecting ice filaments; values tentatively

suggested are: $\ell = 10 \mu m$ for dendrites; $50 \mu m$ for powdered snow; $100 \mu m$ for rimed crystals; $1000 \mu m$ for sleet and graupel

$\kappa = a/R_m$, the impaction parameter

$\nu = \mu_w/\mu_a$, the ratio of dynamic viscosities for water and air

$s(>s_*) = v\tau/L$, the Stokes number with the appropriate speed and length scales

τ = particle stopping time

$s_* = \left\{ \frac{12}{10} + \frac{1}{12} \ln(1 + Re) \right\} / \left\{ 1 + \ln(1 + Re) \right\}$, the critical Stokes number, with

$Re = Re_m$ or Re_ℓ

$c = 2/3 - s_*$

g = acceleration of gravity

ρ_w, ρ_a = densities of water and air, respectively.

Comparisons of these semi-empirical formulas with essentially all appropriate pre-1975 data are given elsewhere.² The corresponding formulas for polydisperse aerosols and accounting for changes in particle sizes have been submitted for publication.³ The equations given here predict snow scavenging rates consistent with most previous data but one to three orders of magnitude less than Graedel and Franey's inferred values. However, there is the possibility that their data does indeed reflect snow scavenging but of particles much larger than the sampled dry size; condensation growth of particles appears to explain³ the otherwise unexpectedly rapid rain removal of plume particles recently reported by Radke et al.⁴

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A GENERAL FORMALISM FOR THE PRECIPITATION SCAVENGING RATES FOR AEROSOL PARTICLES

W. G. N. Slinn

From a continuity equation, general expressions are derived for the particle- and space-averaged precipitation scavenging rates for an aerosol pollutant. This formalism provided a framework for developing approximations for the removal rates.

INTRODUCTION

Precipitation scavenging or washout is a term used to describe removal of an air pollutant by various types of precipitation such as rain, snow, etc. Sometimes it is convenient to distinguish in-cloud from below-cloud scavenging if the elevation of the pollutant is clearly above or below the cloud base, respectively. This report demonstrates some general relationships for the scavenging rates which are appropriate both for in- and below-cloud scavenging. A more complete development of the results is given elsewhere.¹

THE CONTINUITY EQUATION

Let χda be the amount of contaminant per unit volume associated with particles of radii a to $a + da$ (viz., "a-particles"). Then the evolution of this contaminant is governed by a continuity equation:

$$\frac{D\chi}{Dt} = \nabla \cdot (K \cdot \nabla \chi) - \psi \chi + G(\chi) - L(\chi) \quad (1)$$

where $D/Dt = \partial/\partial t + \vec{v} \cdot \nabla$ is the usual total time derivative; $\vec{v}$ is the wind

field of the air, assumed incompressible; K is the turbulent diffusivity (or, more generally, $K \cdot \nabla \chi$ symbolizes the turbulent flux); ψ is the precipitation scavenging (or washout) rate coefficient; and G and L symbolize gain and loss of contaminant associated with a -particles because of condensation, evaporation, coagulation, etc. Dry deposition could also enter here but it is more convenient to (and elsewhere* we do) treat it as a boundary condition.

PARTICLE-AVERAGED REMOVAL RATES

One way to temporarily avoid the many complications in Eq. (1) is to average it over all particle sizes. This leads to

$$\frac{D\chi_t}{Dt} = \nabla \cdot (K \cdot \nabla \chi_t) - \bar{\psi}(\vec{r}, t) \chi_t \quad (2)$$

where the total contaminant density is

$$\chi_t(\vec{r}, t) = \int_0^\infty da \chi(\vec{r}, t; a) \quad (3)$$

and the particle-averaged removal rate is

$$\bar{\psi}(\vec{r}, t) = \int_0^\infty da \psi(\vec{r}, t; a) \chi / \chi_t \quad (4)$$

To obtain (2) from (1) it has been assumed that turbulence acts on all particles similarly. Use has been made of the observations that in all processes contributing to G and L , the contaminant is not lost from the space volume.

SPACE-AVERAGED REMOVAL RATES

To obtain an equation for the pollutant concentration in terms of a

space- and particle-averaged removal rate, Eq. (2) is integrated over a large enough space volume so that no contaminant is convected from the volume by the wind. Then use of the divergence theorem and $\nabla \cdot \vec{v} = 0$ and ignoring dry deposition leads to

$$\frac{\partial Q_t}{\partial t} = - \langle \bar{\psi}(t) \rangle Q_t \quad (5)$$

where the total contaminant present is

$$Q_t = \int d\vec{r} \chi_t(\vec{r}, t) \quad (6)$$

and the space- and particle-averaged removal rate is

$$\langle \bar{\psi}(t) \rangle = \int d\vec{r} \bar{\psi}(\vec{r}, t) \chi_t(\vec{r}, t) / Q_t \quad (7)$$

For the case that $\langle \bar{\psi} \rangle$ is time independent then the solution to (5) is

$$Q_t = Q_{t_0} \exp \left\{ - \langle \bar{\psi} \rangle t \right\} \quad (8)$$

which is a familiar expression used in precipitation scavenging studies.

CONCLUSIONS AND COMMENTS

The above analysis provides a general foundation for the theoretical description of the irreversible capture of pollutant particles by precipitation. Other considerations are needed for the reversible capture of gases.² However, it is clear that the formalism presented here only temporarily avoids the complexities of Eq. (1). Thus, when one attempts to evaluate $\bar{\psi}$ or $\langle \bar{\psi} \rangle$ in (4) and (7) he finds that it is first necessary to know $\psi(\vec{r}, a, t)$ and $\chi(\vec{r}, a, t)$. Now $\psi(\vec{r}, a, t)$ can presumably be specified*

* See the reports in this volume by author concerned with dry deposition.

*In this volume see the contribution by author entitled "Precipitation Scavenging of Monodisperse Aerosol Particles."

but to obtain χ apparently Eq. (1) must be solved. But if it could be solved, there would be no need for the development from (2) to (8). To extricate oneself from this circuitous development and yet to utilize the coarser descriptions of scavenging, a number of approximations for

χ can be introduced. Examples are given elsewhere in this volume* and in the original publication.¹

*In this volume see the contribution by author entitled "Approximations for the Precipitation Scavenging Rates for Polydisperse, Active, Aerosolized Pollutants."

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APPROXIMATIONS FOR THE PRECIPITATION SCAVENGING RATES FOR POLYDISPERSE, ACTIVE, AEROSOLIZED POLLUTANTS

W. G. N. Slinn

Approximations for the distribution among, and evolution of, different classes (free, growing and hosted) of aerosol pollutant are introduced into a general expression for the (below- or in-cloud) precipitation scavenging rate. The transparent result demonstrates the dependence of the average removal rate on time and the polydispersity of the aerosol.

INTRODUCTION

Elsewhere in this volume,* the author developed the expression for the particle-averaged precipitation scavenging rate

$$\bar{\psi}(\vec{r}, t) = \int_0^{\infty} da \, \psi(\vec{r}, t; a) \chi(\vec{r}, t; a) / \chi_t(\vec{r}, a) \quad (1)$$

*See "A General Formalism for the Precipitation Scavenging Rates for Aerosol Particles," this volume.

where χda is the amount of pollutant associated with particles of radii a to $a + da$, $\chi_t = \int da \chi$, and ψ is the precipitation scavenging rate for a -particles. To evaluate (1), clearly ψ and χ are needed. Now ψ can presumably be specified, even if only approximately,* but to obtain χ accurately, apparently one must solve a continuity equation for the particles. This is a formidable undertaking. However, in view of the present uncertainties about the removal rates, it would

*See the report by the author entitled, "Precipitation Scavenging of Monodisperse Aerosol Particles," this volume.

seem consistent to proceed with only approximations for the air concentration, χ . This report suggests approximations for χ and thereby, with the help of (1), develops approximations for the particle-averaged removal rate ψ .

ASSUMPTIONS

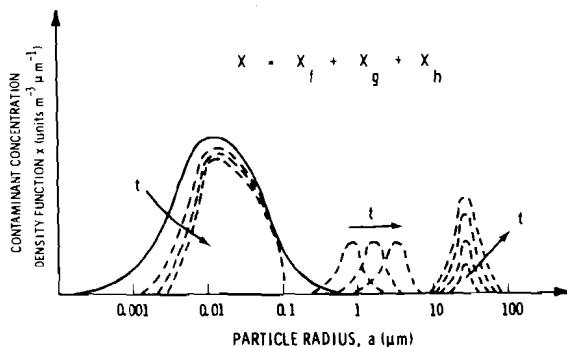
The following assumptions are involved:

- For each chemical species of aerosol present, three portions of the size spectrum are identified, qualitatively as shown in Figure 1 and labeled as χ_f (free), χ_g (growing), and χ_h (hosted). Thus the total air concentration of a-particles is taken to be

$$\chi = \chi_f + \chi_g + \chi_h \quad (2)$$

- The evolution of the free particles is approximately governed by

$$\frac{\partial \chi_f}{\partial t} = v \cdot (\mathbf{K} \cdot \nabla \chi_f) - \alpha \chi_f - \psi_f \chi_f \quad (3)$$



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FIGURE 1. A Schematic Illustration of the Contaminant's Assumed Distribution Among the Three Particle Classes (Free, Growing and Hosted) and Their Time Evolution

where $\mathbf{K} \cdot \nabla \chi_f$ symbolizes the turbulent flux of the particles, α is the rate at which the free particles attach to any hosts, and ψ_f is the (below- or in-cloud) precipitation scavenging rate.

- An adequate approximate solution to (3) is assumed to be

$$\chi_f = \dot{q}_f(a) P(\vec{r}, t; \mathbf{K}, \vec{v}) \exp \left\{ - \int_0^s (\alpha + \psi_f) ds \right\} \quad (4)$$

where $\dot{q}_f(a)$ is the number of free a-particles released per second, P is some plume model and s is either t or x/u .

- Similar approximate equations and solutions (with the same plume model) are assumed for χ_g and χ_h .
- The radius of the growing particles is assumed known and the host particles (e.g., plume or cloud droplets) are assumed to possess the same radius λ .
- The particle size distributions of χ_f , χ_g and χ_h are assumed to be log normal and integrals over particle sizes are approximated by evaluating the integrands at their mean values.
- The contaminant is distributed among the particles according to

$$\chi(a) = c a^j n(a) \quad (5)$$

where c is an obvious normalization constant, $n(a)$ is the particle number distribution function and, for example, $j = 3$ if the contaminant is just the mass of the particles.

RESULTS

If these approximations and assumptions are used in (1), the particle-averaged rain scavenging rate becomes, approximately,

$$\bar{\psi}_r = \frac{p}{2R_m} \left[F_{fo} \exp \left\{ -\int_0^s (\bar{\alpha} + \bar{\psi}_f - \bar{\psi}_r) ds \right\} \right.$$

$$E(a_{fj}, R_m) + F_{go} \exp \left\{ -\int_0^s (\bar{\psi}_g - \bar{\psi}_r) ds \right\}$$

$$E(a_{gj}, R_m) + \frac{\bar{\alpha}}{(\bar{\alpha} + \bar{\psi}_f - \bar{\psi}_h)} F_{fo} \cdot$$

$$\cdot \left\{ \exp \left\{ -\int_0^s (\bar{\psi}_h - \bar{\psi}_r) ds \right\} - \exp \left\{ -\int_0^s (\bar{\alpha} + \bar{\psi}_f - \bar{\psi}_r) ds \right\} \right\} E(\lambda, R_m) \Bigg] \quad (6)$$

where F_{fo} and $F_{go} = 1 - F_{fo}$ are the initial fractions of the contaminant associated with free and growing particles, respectively, and where

$$a_j = \bar{a} \exp \left\{ \left(j + \frac{1}{2} \right) \sigma^2 \right\} \quad (7)$$

in which $\bar{a}$ is the geometric mean (number) radius and $\sigma = \ln \bar{\sigma}$ where $\bar{\sigma}$ is the geometric standard deviation, each for the specific aerosol class. Similar expressions are obtained for snow scavenging.¹

For small time (i.e., for $\bar{\psi}_r t \ll 1$ or if the difference between any $\bar{\psi}_i$ and $\bar{\psi}_r$ is small and this $\Delta\psi$ satisfies $\Delta\psi t \ll 1$), for a time independent attachment rate, and for $j = 3$, then

(6) with $s = t$ simplifies to the transparent expression for the mass rain-scavenging rate (below or within a cloud):

$$\bar{\psi}_r = \frac{p}{2R_m} \left[F_{fo} \exp (-\bar{\alpha} t) E \right.$$

$$\left\{ \bar{a}_f \exp \left(\frac{7}{2} \sigma_f^2 \right), R_m \right\} + F_{go} E$$

$$\left\{ \bar{a}_{go} \exp \left(\frac{7}{2} \sigma_g^2 \right) + \left(\frac{t \mu m^2}{100 \text{ sec}} \right)^{1/2}, R_m \right\}$$

$$+ F_{fo} \left\{ 1 - \exp (-\bar{\alpha} t) \right\} E(\lambda, R_m) \Bigg] \quad (8)$$

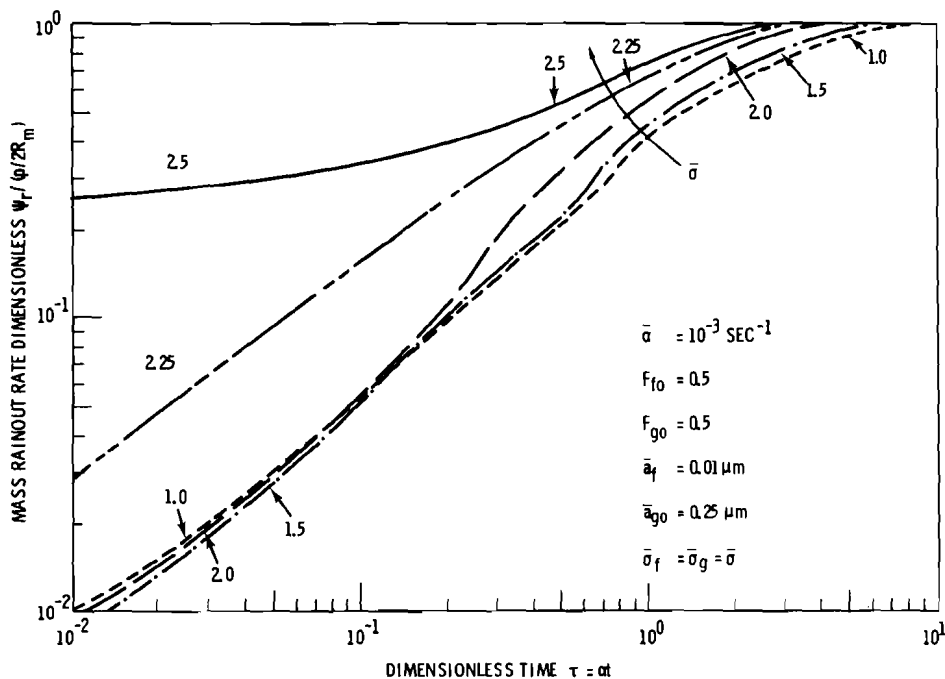
in which a specific, particle growth rate has been assumed. For snow scavenging, the quantities outside the square brackets, $[]$, are changed and the drop-particle collection efficiency E is replaced by the particle-ice crystal collection efficiency, E .¹

CONCLUSIONS

The results obtained here are demonstrated in Figure 2 for the specific case of the parameters shown. This figure illustrates the rapid increase in the mass rainout rate (8) both with time and with an increase in the polydispersity of the aerosol (viz. $\bar{\sigma}$). For further details on this latter point, see the original paper¹ and the recent results of Dana and Hales.²

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FIGURE 2. An Illustrative Example of Equation (8) for the Choice of Parameters Shown, Which Demonstrates the Strong Dependence of the Mass-Averaged Removal Rate on the Polydispersity of the Aerosol (Reflected in $\bar{\sigma}$) and on the Time for Attachment and Condensational Growth.

RAIN SCAVENGING OF ACTIVE AEROSOL PARTICLES FROM A PLUME

W. G. N. Slinn

Hypotheses are advanced in an attempt to explain the measured rates at which particles are scavenged by rain from a kraft-process paper-mill plume. Weaknesses of the assumptions are noted as well as the value of further research.

INTRODUCTION

Elsewhere,¹ the author has suggested that the in- or below-cloud rain scavenging rate for the removal of monodisperse aerosol particles of radius a could be approximated by

$$\psi_r(\vec{r}, t; a) = \frac{p(\vec{r}, t)}{2 R_m} E[a(t), R_m] \quad (1)$$

where ρ is the rainfall rate (which may depend on position, $\vec{r}$, and time, t); R_m is the volume-mean raindrop radius and E is the collection efficiency. For unit retention efficiency, a semiempirical expression

for the collection efficiency was given as

$$E(a, R_m) = \frac{4}{Pe} [1 + 0.4 Re^{1/2} Sc^{1/3}] + 4\kappa \left[\kappa + \frac{(1 + 2 V\kappa)}{(1 + VRe^{-1/2})} \right] + \left[\frac{s - s_*}{s + c} \right]^{3/2} \quad (2)$$

where

- $Pe = Re Sc$ is the Péclet number;
- $Re = R_m v_t / \nu$ is the Reynolds number in which v_t is the mean-volume drop terminal velocity and ν is the kinematic viscosity of air;
- $Sc = \nu/D$ is the Schmidt number where D is the molecular diffusivity of the particles;
- $\kappa = a/R_m$ is the interception parameter;
- $V = \mu_w/\mu_a$ is the ratio of dynamic viscosities of water to air;
- $s (>s_*) = \tau v_t/R$ is the Stokes number where τ is the particle relaxation time;
- $c = \frac{2}{3} - s_*$;
- $s_* = \left[\frac{12}{10} + \frac{1}{12} \ln(1 + Re) \right] / [1 + \ln(1 + Re)]$ is an empirical fit to the critical Stokes number.

This report compares these predictions with the experimental results recently obtained by Radke, Hindman and Hobbs.² A more complete description of the comparison is given elsewhere.³

DATA

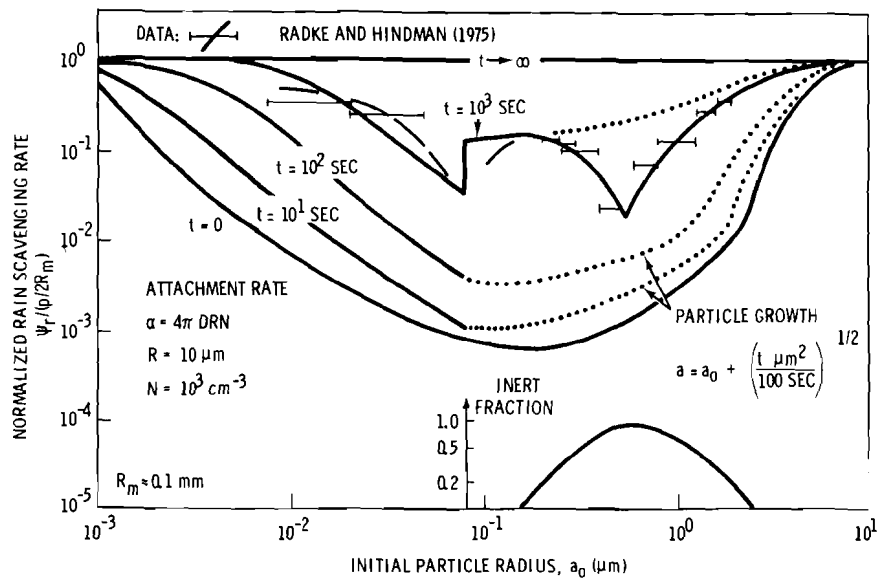
The important data set, the first of its kind, presented by Radke et al.²

was obtained using airborne aerosol size spectrometers that spanned the particle range from about 0.01 μm to 5 μm radius. They determined the particle size distribution in a plume about 10³ sec downwind from a Kraft-process paper mill, both before and after a rain shower had scavenged the plume. From the difference between these two size distributions and their airborne measurement of the raindrop size distribution they inferred the collection efficiency as a function of particle size shown in Figure 1, where the data points are the horizontal bars.

INTERPRETATION

One interpretation of the data is as follows. The $t = 0$ curve shown on Figure 1 is a plot of Equations (1) and (2). This is the expected collection efficiency for passive aerosol particles. If it is assumed that, instead of being passive, the particles were active and attached to the plume droplets at a rate $\alpha = 4\pi DRN$ where R and N are the mean radii and number density of the plume droplets,⁴ then the attached fraction of aerosol particles would have a collection efficiency appropriate for the plume droplets (viz., $E = 1$ for $R = 10 \mu m$). Consequently, especially for particles of radii $a < 0.1 \mu m$, the removal rate would apparently increase; the calculated results are shown as the solid curves in Figure 1 labeled $t = 10^1, 10^2$ and 10^3 sec for $a < 0.1 \mu m$. In the limit $t \rightarrow \infty$ (see Figure 1), $E = 1$ for all particle sizes.

A second effect is assumed to become important for some of the particles larger than about 0.08 μm radius; namely, condensational growth of the particles. This specific critical size of 0.08 μm was chosen to fit the data, but it is not inconsistent with theoretical considerations for the expected humidity in the plume.⁵ For the specific particle growth rate shown in the figure, the collection efficiency at $t = 10^1, 10^2$ and 10^3 sec is as shown by the dotted curves. Obtaining the correct growth rate expression (and thereby the correct collection efficiency $E[a(t), R_m]$) is a complex problem and the naivety of choosing a single growth rate for all particles is acknowledged.



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FIGURE 1. The Collection Efficiency as a Function of Particle Size as Measured by Radke et al,² and as Predicted by Theory (see text)

A third phenomenon is invoked to explain the apparent relative minimum in the collection efficiency at about 0.6 μm . It is assumed, based on comments by the authors, that a certain fraction of the sampled particles were inert to condensational growth (at least for $t \leq 10^3$ sec). The specific fraction assumed to be inert, chosen to fit the data, is shown in the inset to Figure 1. The source of these particles distinct from the sulfate particles from the Kraft process, is presumed to be the coal-fired power plant at the mill. The resulting collection efficiency is shown as the solid curve (only at $t = 10^3$ sec).

COMMENTS AND CONCLUSIONS

The analysis presented to interpret the data possesses a number of weaknesses. For example, to explain the data for $a \leq 0.1 \mu\text{m}$ particle, it was implicitly assumed that there was ignorable attachment of particles to plume droplets before the rain shower

occurred. There is no data to justify this assumption. Instead, the cause of the more rapid than expected removal rate for these small particles could be that the precursor gases were scavenged by the rain shower and therefore it was not that more particles were scavenged but that fewer particles were created. Another major difficulty is in the explanation presented for the relative minimum at about 0.6 μm . It was assumed that this corresponds to a maximum in the number of particles which are inert but Hindman (private communication) found no such maximum experimentally. Consequently we conclude that more theoretical and experimental work is needed before this particular type of plume scavenging is understood. That these studies are important is obvious when one realizes that without such data, the predictions for the rain scavenging of particles (with $0.01 \lesssim a \lesssim 1 \mu\text{m}$) would have been about 2 orders of magnitude smaller than the true values.

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WASHOUT RATIOS FOR PARTICULATE POLLUTION WITH AN APPLICATION TO CONVECTIVE STORMS

W. G. N. Slinn

General expressions for washout ratios are derived which demonstrate that these ratios represent averages over many variables. Comparison of the theory is made with recent field data.

INTRODUCTION

Washout ratios are defined to be the ratio of a pollutant's concentration measured in (ground level) precipitation, κ , to the pollutant's concentration in (usually ground level) air, χ . Analyses of a considerable quantity of κ/χ data (e.g., see Junge,¹ Gedeonov, et al.,² Engelmann,³) have revealed the relatively small variations of these washout rates. Elsewhere⁴ the author emphasized qualitatively that the reason for this relative invariance is that these ratios represent multiple averages over the pertinent variables (such as precipitation size and type, particle size and type, pollutant distribution in the atmosphere, storm type, etc.).

This report will demonstrate this concept quantitatively and compare the theoretical discussion with experimental data recently reported by Gatz⁵ of washout ratios for various particulate pollutants scavenged by convective storms.

AVERAGES

Here, rather than develop a single expression for the washout ratios which explicitly contains all appropriate averages, to simplify notation, averages will be evaluated at each stage of the analysis. Thus, for example, the rain scavenging rate (for both below- and in-cloud scavenging) is

$$\psi_r = \int dR E(a, R) N(R) \pi R^2 v_t(R) \quad (1)$$

where the symbols have their usual meaning. Now (1), which is an average of the collection efficiency, E , over all drop sizes is approximated by

$$\psi \approx \frac{p}{2R_m} E(a, R_m) \quad (2)$$

where p is the precipitation rate and R_m is the volume-mean raindrop radius. Similarly, for snow scavenging, one obtains⁴

$$\psi \approx 10^6 \frac{\text{cm}}{\text{sec}^2} \frac{p}{\langle v_t \rangle} E(a, \lambda) \quad (3)$$

where p is the precipitation rate in rainwater equivalent, $\langle v_t \rangle$ is an average fall speed for the ice crystals and λ is a characteristic capture length for the ice crystals, not necessarily related to crystal size.⁴

As shown elsewhere in this volume,* an approximation to the average of (2) and similarly for (3) over all particle sizes is

$$\begin{aligned} \bar{\psi}_r = \frac{p}{2R_m} \left[F_{fo} \exp\{-\bar{\alpha}t\} E \right. \\ \left. \left\{ \bar{a}_f \exp\left(\frac{7}{2} \sigma_f^2\right), R_m \right\} + F_{go} E \right. \\ \left. \left\{ \bar{a}_{go} \exp\left(\frac{7}{2} \sigma_g^2\right) + \left(\frac{t \mu m^2}{100 \text{ sec}}\right)^{1/2}, R_m \right\} \right. \\ \left. + F_{fo} \left\{ 1 - \exp(-\bar{\alpha}t) \right\} E(\lambda, R_m) \right] \quad (4) \end{aligned}$$

(the symbols are defined elsewhere*). For snow scavenging, (4) is modified

only by replacing $p/2R_m$ by the appropriate terms from (3) and E is replaced by E .

The space averaged removal for rain (and the obvious, similar expression for snow) is then

$$\langle \bar{\psi}_r \rangle = \frac{1}{Q_t} \int d\vec{r} \frac{p(\vec{r}, t)}{2R_m} [E] x_t(\vec{r}, t) \quad (5)$$

where, to save space $[E]$ has been written for the term in these brackets in (4) (and similarly for snow). In (5), Q_t is the total amount of pollutant in the volume over which the average is taken and x_t is the total contaminant concentration, i.e., summed over all particle size.

WASHOUT RATIOS

Now consider that washout ratio which is the ratio of the contaminant's concentration in surface level precipitation, κ_0 , to its concentration in surface level air χ_{to} . To obtain an expression for this ratio, it is noted that the rate of contaminant removal per unit volume is ψx_t . If the contaminant's distribution is fairly uniform horizontally, the flux of contaminant to the earth's surface is $\int \psi x_t dz$. The flux of precipitation to the surface is p_0 . Therefore, this washout ratio is

$$\frac{\kappa_0}{\chi_{to}} = \frac{1}{p \chi_{to}} \int_0^H \psi x_t dz \quad (6)$$

where H is the height to the top of the cloud or above which there is no contaminant, whichever is smaller. Clearly (6) is an impoverished version of the space averaged removal rate (5).

A special case of some interest is if the contaminant's concentration is distributed initially, fairly uniformly within the atmosphere mixed layer. If this layer is then scavenged by the updraft and subsequent rain shower from a convective storm, (6) with (5) (and similarly for solid precipitation) leads to

$$\frac{\kappa_0}{\chi_{to}} = \frac{[E]h}{2R_m} \quad (7)$$

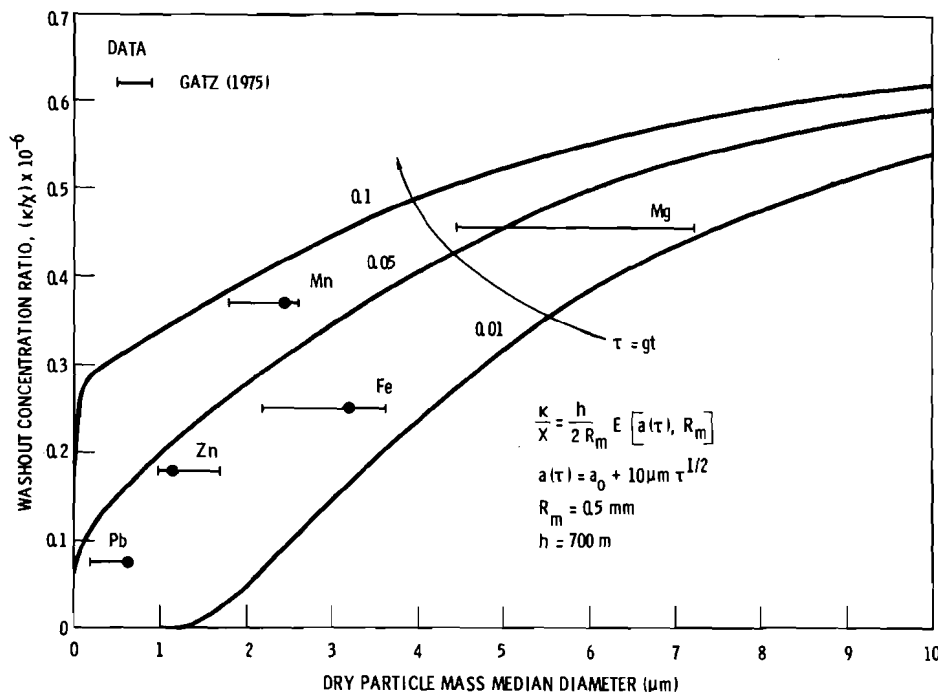
*See "Approximations for the Precipitation Scavenging Rates for Polydisperse, Active, Aerosolized Pollutants,"

where h is an unknown depth within the cloud from which the pollutant is effectively scavenged. An estimate for h can be found from the general behavior of the experimental data.

RESULTS AND CONCLUSIONS

The analysis presented above demonstrates that washout ratios are averages over a considerable number of variables and, since inherent in any averaging process is a smoothing of variability, it can be expected that washout ratios are relatively invariant for different aerosolized pollu-

tants and different storm types. Equation (7) predicts that the washout ratios for aerosol particles are independent of precipitation rate, p , except for the weak dependence of R_m (and, similarly, of $\langle v_t \rangle$) on p . This is a result well substantiated by field data (e.g., see Makhon'ko et al.,⁶ Engelmann,³ and Gatz⁵). Figure 1 shows that the magnitude and the particle size dependence of the washout ratios as found by Gatz⁵ are reasonably well described by (7), recognizing that neither the polydispersity nor the condensational growth rate of the particles is known.



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FIGURE 1. A Comparison of Predictions of (7) with the Average Washout Ratios for Convective Storms Measured by Gatz.⁵ The horizontal bars on the data reflect the different mass median diameters (mmd's) found in different cities; the dot on the bar is the mmd measured in the city (St. Louis) near which the washout ratios were measured. τ is nondimensionalized time; g is the unknown particle growth rate; a_0 is the dry particle mass median radius, i.e., 1/2 the mmd as given on the abscissa. The choice for h was made solely to improve the fit to the data.

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SYNOPTIC SCALE WET AND DRY DEPOSITION

W. G. N. Slinn

It is demonstrated that the atmospheric removal length scales for both wet and dry deposition of most industrial pollutants are typically in the range of 10^2 to 10^4 km.

INTRODUCTION

In an earlier report,¹ wet and dry deposition near a specific pollution source were compared. It was estimated that during rain, the ratio of wet to dry deposition was approximately

$$\frac{W}{D} = c \frac{p}{R_m} \frac{\bar{u}}{u_*^2} \frac{h}{\sigma_z} \left(\frac{\sigma_z}{h} \right) \exp \left(\frac{h^2}{2 \sigma_z^2} \right) \quad (1)$$

where p is the rainfall rate, R_m is the volume-mean drop size, u is the wind speed, h is the height of the release, u_* is the friction velocity, σ_z^2 is the variance characterizing the vertical diffusion and c is a numerical factor. The choice $c = 0.2$ provided a

fair fit to experimental data. This report extends the analysis to consider wet and dry deposition at larger space scales. For these scales it is assumed that the height of the pollution release is unimportant; for the estimate of dry deposition the pollution concentration is assumed uniform in the atmospheres mixed layer, of height H .

DRY DEPOSITION LENGTH SCALE

If the average concentration of the pollutant in the mixed layer (of height H and with mean wind u) is $\bar{\chi}$ and if horizontal diffusion is ignorable, the z -averaged steady-state continuity equation becomes

$$\bar{u} \frac{\partial}{\partial x} (H \bar{\chi}) = -v_d \bar{\chi} \quad (2)$$

Therefore, the e-fold dry-deposition length scale is

$$\lambda_d = \frac{\bar{u} H}{v_d} \quad (3)$$

This can predict substantially different numerical values. For example, over a forest in winter, if $\bar{u} = 5 \text{ m sec}^{-1}$, $H = 200 \text{ m}$ and $v_d = 10 \text{ cm sec}^{-1}$, then $\lambda_d \sim 10 \text{ km}$. At the other extreme, say for $0.1 \text{ }\mu\text{m}$ particles over water during a well-mixed summer day, with $\bar{u} = 5 \text{ m sec}^{-1}$, $H = 2 \text{ km}$ and $v_d = 0.1 \text{ cm sec}^{-1}$, then $\lambda_d \sim 10^4 \text{ km}$. Typically, though, $10^2 \leq \lambda_d \leq 10^3 \text{ km}$ and thus sources 100-1000 km upwind of a given area can be expected to make a major contribution to the dry deposition of pollution in that area.

WET DEPOSITION LENGTH SCALE

To estimate the length scale over which wet deposition significantly contributes to the total deposition, it is not profitable to pursue a wash-out model which leads to an expression such as

$$\bar{u} \frac{\partial \bar{\chi}}{\partial x} = - \langle \bar{\psi} \rangle \bar{\chi} \quad (4)$$

where $\langle \bar{\psi} \rangle$ is an average precipitation scavenging rate. This predicts an e-fold length of 10-100 km. Obviously (4) is not relevant to the problem of interest because, at best, (4) is applicable only during precipitation.

The space scale of interest can be deduced from the following considerations:

- For the large scale problem, most wet removal occurs by the incorporation of the pollutant within a storm system, i.e., by in-cloud scavenging;
- In-cloud scavenging typically removes 10-90% of the pollutant ingested by the storm, depending on the efficiency with which the storm removes water;
- The wet-removal space scale is dictated by the distance over which the material is transported before it encounters a storm which efficiently removes precipitation.

Consequently, from the large scale, precipitation scavenging can be viewed to a first approximation as a Poisson process in time. A scavenging event occurs (and as a first approximation we assume the event completely cleanses the atmosphere) simultaneously with the occurrence of an "efficient" storm system. The deposition pattern is then estimated to be that for a Poisson process; the probability that a deposition event occurs during the time period (0 - t) is

$$P\{1 \text{ in } 0 - t\} = \bar{v} t \exp\{-\bar{v} t\} \quad (5)$$

where $\bar{v}$ is the average number of events over a long time interval. If statistics for the storm frequency, $\bar{v}$, are approximately the same for any area within the length scale $\bar{u}/\bar{v}$, then t in (5) can be replaced by $x/\bar{u}$. In this way, the statistics of scavenging events is reduced to a study of the statistics of rain events and the wet deposition e-fold length scale is

$$\lambda_w = \bar{u}/(\bar{v}\bar{e}) \quad (6)$$

where $\bar{e}$ is the average efficiency with which the pollutant is removed by an average storm.

As an example of these considerations, consider wet deposition in the northeastern U.S. From statistics presented by Huff,² 67% of the storms in east central Illinois, during 1955-1964, lasted less than 1 day. For the 50-year period, 1906-1955, 63% of the total precipitation occurred at a rate of 0.1-1 in./day. Roughly if these two statistics are incorporated with a typical annual precipitation of 40 in./year, we obtain that on the average, an efficient (~ 0.5 in. precipitation) storm occurs once in every 4.5 days. If this result is typical for the region, then in (6) we have $\bar{v} = (4.5 \text{ d})^{-1}$ and, therefore, $10^2 \leq \lambda_w \leq 10^4 \text{ km}$, depending on the wind speed and $\bar{e}$.

WET VERSUS DRY DEPOSITION

The above considerations imply the wet and dry e-fold length scales are of comparable size (at least when the total rainfall is about 1 m/year). In particular $10^2 \leq \lambda_w, \lambda_d \leq 10^3 \text{ km}$. Consequently, it is suggested that wet and dry deposition make similar contributions to the total deposition. An independent check on this result

can be obtained by estimating the relative fluxes. The dry flux is approximately $v_d \bar{x}$. The wet flux by in-cloud scavenging during rain is roughly $(10^5 - 10^6) p \bar{x}$ where p is the precipitation rate. Then, accounting for the durations over which each flux operates and for a total precipitation of 100 cm/year^{-1} , we obtain

$$\frac{W}{D} = \frac{(10^5 - 10^6) (100 \text{ cm yr}^{-1})}{(0.3 - 3 \text{ cm sec}^{-1}) (3 \times 10^7 \text{ sec yr}^{-1})}$$

$$\sim 0(1) \quad (7)$$

That is, to within an order of magnitude and consistent with the other analysis in this report, it appears that in the northeastern U.S., wet and dry processes contribute comparable amounts to the total deposition.

COMMENTS

The space scale for removal from the atmosphere of industrial pollutants such as SO_2 and SO_4^{2-} is estimated to be typically $10^2 - 10^4 \text{ km}$. From this estimate arises a strong argument against the "tall-stack solution" to the pollution problem. Of course, tall stacks can reduce the local air pollution and diffusion will dilute the contribution from a specific source, but as the distribution of sources become more diffuse and ubiquitous, diffusion becomes of no significance to the receptor. Tall-stacks are a "solution" for one nation only if that nation is small or if, for example, the 200 new coal-fired plants to be constructed in the U.S. during the next decade are distributed: 100 near Cape Cod and the other 100 near Cape Hatteras--and the load is shunted between the two centers, depending on which site has westerly winds!

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THE BATTELLE-NORTHWEST IN-CLOUD SCAVENGING
PROGRAM--AN OVERVIEW

J. M. Hales

This contribution summarizes the rationale for direction of the in-cloud frontal storm scavenging program during the past year, and is an introduction to specific reports that follow. Expected direction of this program during future years is also discussed.

The in-cloud precipitation scavenging program at Battelle-Northwest has been performing in-cloud tracer studies to determine mechanisms by which trace gases and/or aerosol particles are taken into cloud water droplets. Typical experiments have involved in-cloud injection and subsequent collection of rain-borne material on a surface network. These experiments have been valuable in providing gross estimates of scavenging rates; however, they have been of comparatively little use in establishing the specific mechanistic information needed to formulate reliable predictive models. We decided that further studies of this type would have little value unless provision were made to improve the resolution of the specific mechanisms involved. Because of this decision and the increase in funding to this DBER program via the Defense Nuclear Agency, the in-cloud scavenging program was modified to overcome the deficiencies of earlier experimental studies.

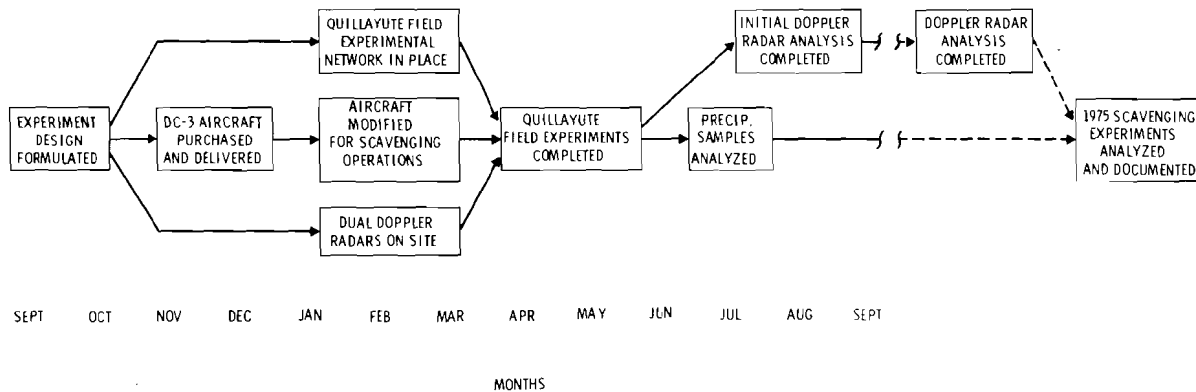
Past experiments provided only limited mechanistic information because they measured the end result of a complex integral process, involving several simultaneous and consecutive steps. Without extensive support measurements assessing the relative importance of these steps is difficult, as is determining appropriate ways to incorporate their influence in modeling endeavors. During past experiments at Quillayute the lack of complete wind-field information has been particularly important in this regard, due to both the complex structures of the precipitating systems and to the

orographic influence of the Olympic peninsula. An additional important lack of support information--that pertaining to cloud-physics parameters--has been caused primarily by limited carrying capability and severe-weather performance of the Battelle Cessna 411 aircraft, which was used in most of the previous experiments.

Program modifications to fulfill the desired objectives included implementing a dual-Doppler radar system at the Quillayute experiment site, acquisition and modification of a larger aircraft for tracer releases and cloud physics measurement, and the acceleration of diagnostic modeling. The dual-Doppler system described in detail wind fields in the experiment area. It was obtained under contract from the NOAA Wave Propagation Laboratory, which installed and operated the radars in cooperation with Battelle-Northwest scientists during the field experiments.

Owing to the short schedule between initiation of the revised program and expected termination of the prime storm period in April, the scavenging program underwent highly accelerated experiment design, aircraft modification and field deployment, as shown in Figure 1. As seen from this figure, final analysis of the field data will be performed and documented upon completion of Doppler-radar data processing, expected in the near future.

Because difficulties with the new aircraft system permitted only one



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FIGURE 1. Experiment Design Flow Chart

tracer release to be conducted concurrently with radar operation during the March 1975 tests, the field results provided rather marginal information on tracer recovery. They were highly rewarding, however, in demonstrating the utility of Doppler radar in support of scavenging experiments, and have led us to conclude that such support is essential in all frontal-storm scavenging tests of this type. The radars described in detail the flows over the Olympic peninsula during passage of storm systems, and are expected to contribute substantial insight to interpretation of not only this year's experiments, but those of previous years as well.

Owing to an unexpected reorientation of Defense Nuclear Agency fund-

ing priorities, plans to conduct similar experiments during the coming year have been abandoned; it is expected, however, that similar in-cloud experiments will be performed in the future--probably in the northeastern United States under the MAP3S program. The in-cloud component of the scavenging program is now entering a phase of detailed analysis of existing results and is performing a critical review of the design bases for future field experiments. Results of this program, including field measurements, radar results, aircraft application, and modeling are described by the contributions immediately following.

PRECIPITATION SCAVENGING FIELD EXPERIMENTS
IN MARCH 1975 ON THE OLYMPIC PENINSULA

C. E. Hane and J. A. Young

Field operations during March 1975 which were a part of the precipitation scavenging research program are described, giving tentative conclusions regarding scavenging within weaker frontal storms and the use of chaff rockets in such experiments. Preliminary dual Doppler velocity calculations from one storm are presented and the possible uses of this data discussed.

INTRODUCTION

The experimental design described in an accompanying article¹ was taken in March 1975 to the northwestern Olympic Peninsula with the intent of performing as many of the experiments as possible within the time allowed. Dual Doppler radars from the Wave Propagation Laboratory in Boulder, Colorado, were deployed at Quillayute Airfield and at Makah Air Force Base near Neah Bay, Washington.¹ Figure 1 A newly-acquired DC-3 aircraft² equipped with instruments for monitoring cloud physics properties and for releasing tracers was stationed in Everett, Washington. The aircraft stood by for call-up by the experiment leaders, who monitored the weather conditions closely with the surveillance radar at Makah, special radiosonde releases, and telecopier transmissions from the National Weather Service Office in Seattle.

SAMPLING AND ANALYSIS

On March 14, 1975, a cold front approached the experimental area from the northwest. The satellite photographs indicated an extensive cloud shield accompanying the front, and radar indicated precipitation moving into the area during the early morning hours. The aircraft was called to the area and tantalum was released (est. 230 g) within a small cell at an altitude of 5000 ft (-3°C). The cell was located approximately 5 nm southwest of the Quillayute Airfield and was moving toward the northeast at the time of release (1024 PST). The release, made over a 2-min period,

was done mechanically (without burning) due to electrical failure aboard the aircraft which prevented conventional burning in the acetone generators. The Makah RHI radar and the Doppler radars indicated cloud tops to be at only 7000 ft during this time, and precipitation elements did not attain sufficient size to be observed by dual Doppler techniques because of the shallow cloud layer. Using the winds obtained by special radiosonde releases during the experiment at Quillayute Airfield, along with assumptions regarding incorporation of tracer into falling rain, an estimate was made of the deposition pattern, and a number of collected samples were analyzed for the tracer.

No tracer was detected above background in the analyzed samples. The reason for the lack of scavenging in this case is very likely the lack of storm intensity. A possible conclusion, therefore, is that scavenging by frontal storms less than 5000 ft thick is inefficient, compared to scavenging by storms ~10,000 ft or more deep which are known to deposit on the ground a significant fraction of released tracer.³ This conclusion, however, needs substantiation through additional field experiments with weaker storms, or through calculations from a microphysical model which has been verified by field experiments in stronger storms. Since a substantial fraction of the storms approaching the northwest U.S. coast occurs with northwest flow aloft and low moisture supply (as was the case here) perhaps it is important to attempt to understand scavenging by some means in

these storms as well as in the more intense storms occurring under more southwesterly flow aloft.

On March 15, 1975, Experiment III¹ was run with some last-minute modifications. The aircraft, equipped with instruments for vertical soundings and a chaff cutter, was not available due to mechanical difficulty, so that the experiment was performed using chaff rockets and radiosondes only. Winds during the experiment were southerly to southwesterly at 20-30 m/sec with only slow veering between the surface and ~7000 ft. Rockets were launched near the 4 locations noted in Figure 4a,¹ including 6 rockets releasing a total of 10 chaff packets at various altitudes between 2000 and 5000 ft. The rocket firings at locations closer to the ridge were delayed so that chaff from all sites might cross the ridge at more nearly the same time. Radiosondes were released from Quillayute Airfield a few minutes before the first rocket firing and a few minutes after the last firing (time interval between radiosondes was 75 min). The Doppler radar at Quillayute Airfield was able to see only one chaff packet and the Doppler radar at Makah saw none of the chaff packets. The reason for this surprising result is not clear, although it is possible that the terrain between the radars and packets prohibited return from some of the lower altitude packets. Also, the weather on this day was rather showery; this may have obscured some of the packets, which yield a weaker return than chaff released by other means. During a test flight on another day, chaff was released by means of a chaff cutter on board the DC-3 aircraft. That strip of chaff provided an excellent return to the Doppler radar which could be tracked for a considerable length of time. Orographic airflow experiments of this type, therefore, might be performed more satisfactorially using aircraft chaff releases instead of rocket releases, even though this would sacrifice the capability for near-simultaneous releases at different locations.

Some of the best data obtained by the dual Dopplers resulted from some moderately intense convective storms which occurred during the morning and afternoon of March 16, 1975. This convection resulted from the destabi-

lizing influences of very cold air aloft in an upper level trough which moved from west to east over the area. The activity could be seen clearly, far to the west over the Pacific Ocean, on satellite photos from the previous day, and was well behind a cold front which had passed through the area on the previous morning. It was definitely not a locally produced phenomenon. During the day on the 16th, the local weather fluctuated remarkably with sunshine one hour and heavy precipitation the next, including several instances of graupel covering the ground.

Data from this day are being analyzed at the present time by the Meteorological Radar Unit, Wave Propagation Laboratory of the National Oceanic and Atmospheric Administration (NOAA) in Boulder, Colorado. A sample of this day's output is shown in Figure 1. The arrows represent velocity perturbations within the rain area of one of these convective storms. The velocities result from computing the mean total velocity at all the points and then subtracting the mean of the field from the total velocity at each point. These velocities are not quite the same as the relative velocities since the relative velocities are obtained by subtracting the motion of the system from the total velocity at each point. The motion of the system may differ from the mean motion of the air within the system.

CONCLUSIONS

Several comments concerning precipitation scavenging may be made with reference to the data in Figure 1. Relative inflow occurs along some of the periphery of this storm, and relative outflow occurs in other locations. Therefore a material from an outside source which might be scavenged may or may not be incorporated into the rain area, depending on where it approaches the storm periphery. In this case, the velocities are at low levels (1-1.5 km msl) where the storm motion is probably faster than the component of air motion in the direction in which the storm is moving. In other words the storm is overtaking the air at this altitude. Outside material would therefore be located on the east-northeast edge (the direction toward which the storm is moving) and would be incorporated into the rain area



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FIGURE 1. Dual Doppler Velocities Obtained from a Convective Storm on 16 March 1975 over the Olympic Peninsula

readily because of the confluent nature of the flow. Whether the material would be scavenged or not depends upon several more factors: 1) Will it be incorporated directly into the rain? or 2) Are the motions in the rain area where it enters the storm upward (in which case it might be scavenged in the condensation process)?

These questions can better be answered when more data are available at other levels in the storm. In conjunction with future field experiments it may be possible with such data to compute the horizontal convergence at

different levels, assume some vertical velocity at different points just below the lowest horizontal plane, and integrate the mass continuity equation to obtain vertical air motions within the rain area. Given the three-dimensional velocity field and the spatial distribution of liquid water, one might calculate trajectories of materials injected into the storm, assume various possibilities for scavenging mechanisms, and compute the deposition pattern on the ground. To complete the picture, scavenged amounts obtained from field experiments should be compared with the calculated result. The

calculation might be done first, however, to estimate where the material might be deposited in relation to where it is released. The field experiments in March 1975 were not designed for this type of storm which very likely produces only a small fraction of the annual rainfall on the Olympic Peninsula. Whether it might accomplish only a small fraction of the total scavenging is yet another question.

One final observation relating to Figure 1 is the possible effect of orographic influences in determining the velocity pattern shown. There appears to be some correlation between the high velocity vectors through the center of the pattern pointing toward the southwest and the higher terrain features (also coarsely depicted in the figure). When radar data at other times and locations become available, the relation between terrain features and possible stronger areas of upward motion will be carefully studied. We hope that a better knowledge of this relation will aid in interpreting some of the deposition patterns of tracer released in past

frontal storm field experiments in exactly this same section of the Olympic Peninsula.

ACKNOWLEDGEMENTS

The March 1975 field operation was a largely developmental field series characterized by innovations in observing systems. Storms highly suitable for experiment were lacking and some unavoidable instrument failures did occur in spite of the dedicated efforts of the field personnel who, besides the two authors, included: W. E. Davis, D. R. Drewes, D. R. Edwards, R. F. Edwards, O. P. Gifford, F. O. Gladfelder, D. W. Glover, J. M. Hales, M. C. Miller, B. C. Scott, T. M. Tanner, C. W. Thomas, and J. M. Thorp. In addition, others too numerous to mention were closely involved in preparations for going into the field. We are also grateful for the kind cooperation of personnel of the National Weather Service at Quillayute, of the 758th U.S. Air Force Radar Squadron at the Makah Base, and of the National Weather Service in Seattle. In addition, special thanks are due to A. S. Frisch, R. G. Strauch, and W. C. Campbell of the Wave Propagation Laboratory (NOAA-Boulder) for their efforts in siting and manning the Doppler radars.

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DESIGN FOR IN-CLOUD SCAVENGING EXPERIMENTS
ON THE OLYMPIC PENINSULA

C. E. Hane and J. A. Young

The design of three separate experiments is described for application in the in-cloud scavenging program field experiments on the Olympic Peninsula. The experiments deal with the effect of particle nature and size, and of cloud dynamics upon scavenging. The experiments involve tracer releases and cloud physics monitoring by an instrumented aircraft, and the use of dual Doppler radar to determine air motions within and in the immediate environment of separate elements of these storms.

Experiments have been designed to study precipitation scavenging in frontal storms utilizing dual Doppler radar and an instrumented aircraft with tracer release capability. The three experiments described here were designed specifically for execution on the Olympic Peninsula of western Washington, although with certain modifications they could be performed at other locations. The three experiments described are:

- (I) Determination of the effect of altitude, temperature, and particle nature on scavenging.
- (II) Comparison of scavenging resulting from tracer release at the leading and trailing edges of a cloud band or cell.
- (III) Determination of vertical air motions induced by orographic features on the northwest Olympic Peninsula.

The purpose of (I) is to determine the effect of height within the cloud upon scavenging, the effect of release in a freezing versus a nonfreezing zone upon scavenging, and the effect of particle size and hygroscopicity upon scavenging. The experiment is designed to determine air motions in and around the clouds as much as possible, to infer the relation between these air motions and microphysical processes within the cloud, including scavenging.

The method used in (I) is to first find a discrete cell or area of relatively heavy rain by means of radar. This cell should, of course, be located with respect to the precipitation collection network such that if a tracer were released, rain containing the tracer would fall over the network of collectors. The precipitation sampling network, locations of the various radars, and release areas in an idealized case are shown in Figure 1. If time permits, chaff can be released from the aircraft and tracked by the radar to determine whether the winds derived from the special radiosonde releases are representative of conditions at release time and location. The tracer releases (6 in all) in this experiment are done in-cloud at approximately 9000, 7500, 6000, and 4500 ft. At one of the altitudes, three tracers are released (hygroscopic, nonhygroscopic, and flare particles). Releases are made approximately at the locations shown in Figure 2, which represents a vertical cross section along a line running north-south approximately through Queets (see Figure 1). Chaff rockets are launched ideally just before the passage of the cell and just after the passage of the cell. A single Doppler is used to track the chaff packets at the altitudes shown in Figure 2 to determine air motions relative to the cell. The aircraft is used following the tracer release to monitor the cloud physics within the



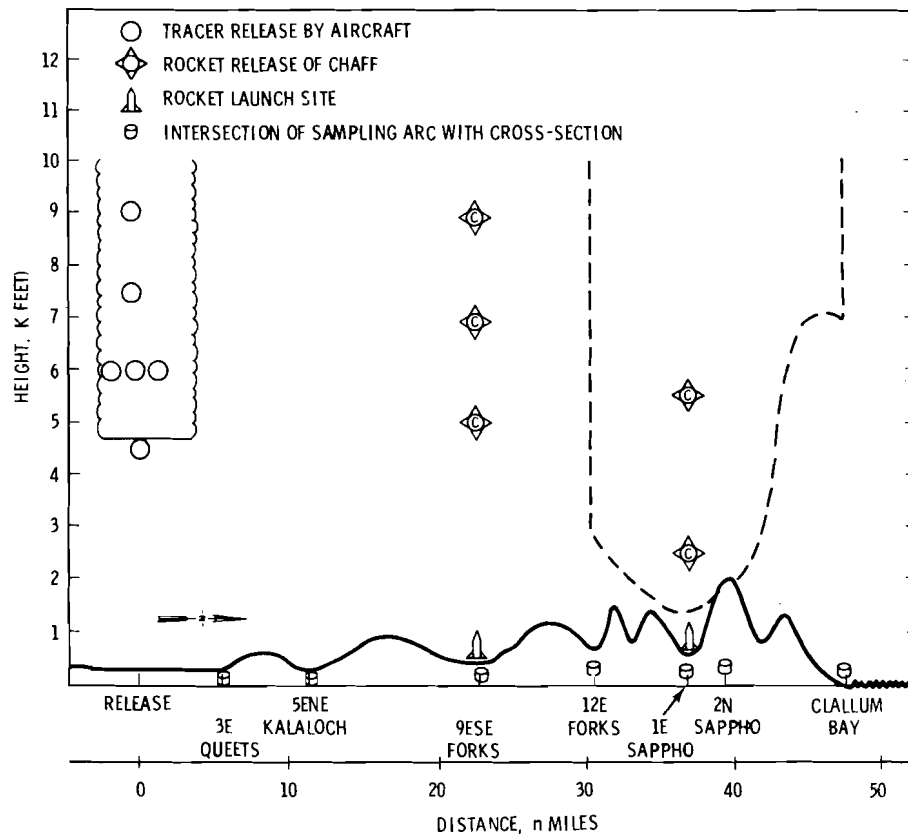
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FIGURE 1. Locations of Instrument Sites on the Olympic Peninsula. Large shaded area indicates locations where dual Doppler coverage extends down to 5000 ft MSL or lower.

cell and to release chaff outside the cell within the dual Doppler coverage area (shaded in Figure 1). Sequential sampling is used along the arcs in the sampling network to determine the time variation in the tracer deposition. Precipitation samples are analyzed for each tracer and the deposition pattern analyzed to determine differences resulting from release location, particle nature, and particle size. The dual Doppler radar coverage is located over and upwind from the heaviest expected deposition, in order to determine horizontal air motions and liquid water contents. Attempts would be made with the aid of all

these analyses to deduce the trajectory of each of the tracers from release to deposition.

Experiment (II) is similar to (I) in many ways. However, in this case the main purpose is to determine the environmental air motion relative to the cell or band just above cloud base. These relative motions are important because they determine whether a plume of material will enter the cell from outside the rain area. The second purpose of (II) is to determine whether the maximum deposition which has occurred in past experiments along the



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FIGURE 2. Schematic Vertical Cross Section for Experiment (I)

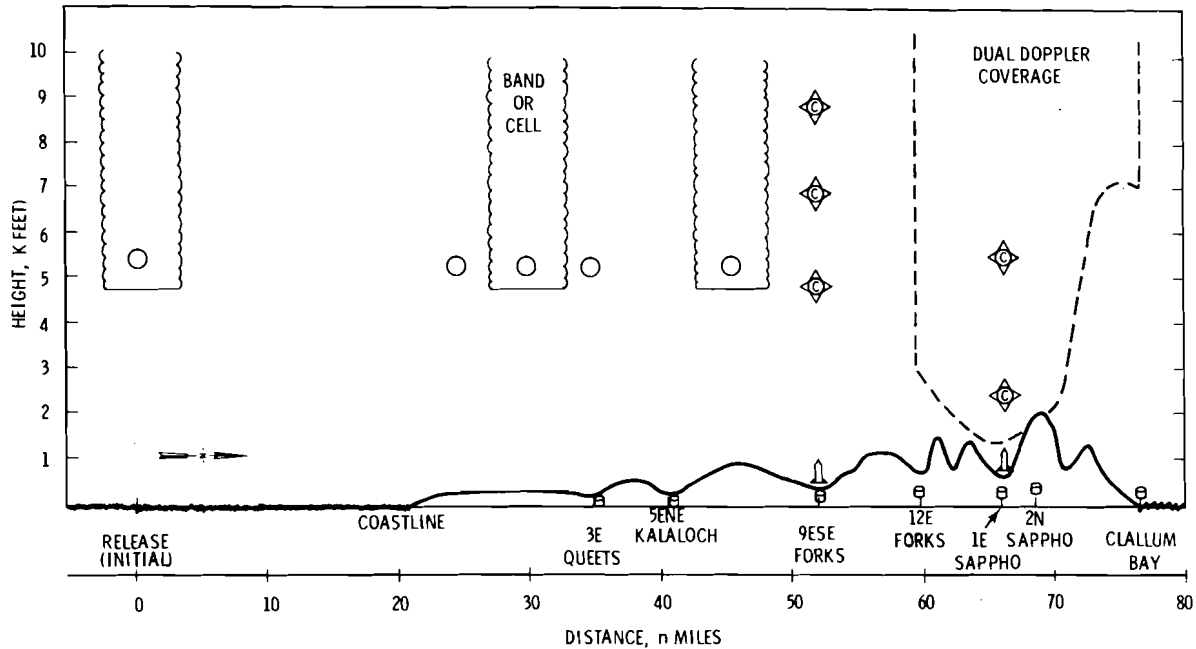
Juan de Fuca coast is due to the orographic effects just upwind or to the fact that significant deposition is occurring to the north.

Some variations in procedure are necessary for the different purposes of (II). One of the release locations is located farther from the sampling network than in (I) (see Figures 1 and 3). A series of three releases is made near the same location as in (I). One of these is made at the leading edge, one at the trailing edge, and one within the precipitation band or cell. A final tracer release is made closer to the arc system, to further test the dependence upon orographic lifting of air over the ridge.

Experiment (III) does not involve tracer releases, but is designed to

measure orographically-induced vertical motion over the ridge near the north coast of the peninsula. The results would help determine preferred rainfall regions in this area and provide an additional basis to approximate lower boundary vertical motions. These would be used in conjunction with dual Doppler radar measurements to determine three-dimensional motions in Olympic Peninsular storms.

It is intended to release chaff packets upwind of the ridge and track the packets with Doppler radar to determine air trajectories as the packets travel over the ridge. Figure 4a shows possible launch locations and aircraft vertical sounding locations. The rockets would release chaff at two or three altitudes in order to determine trajectories at various levels. Altitudes of release would depend on wind speed and fall

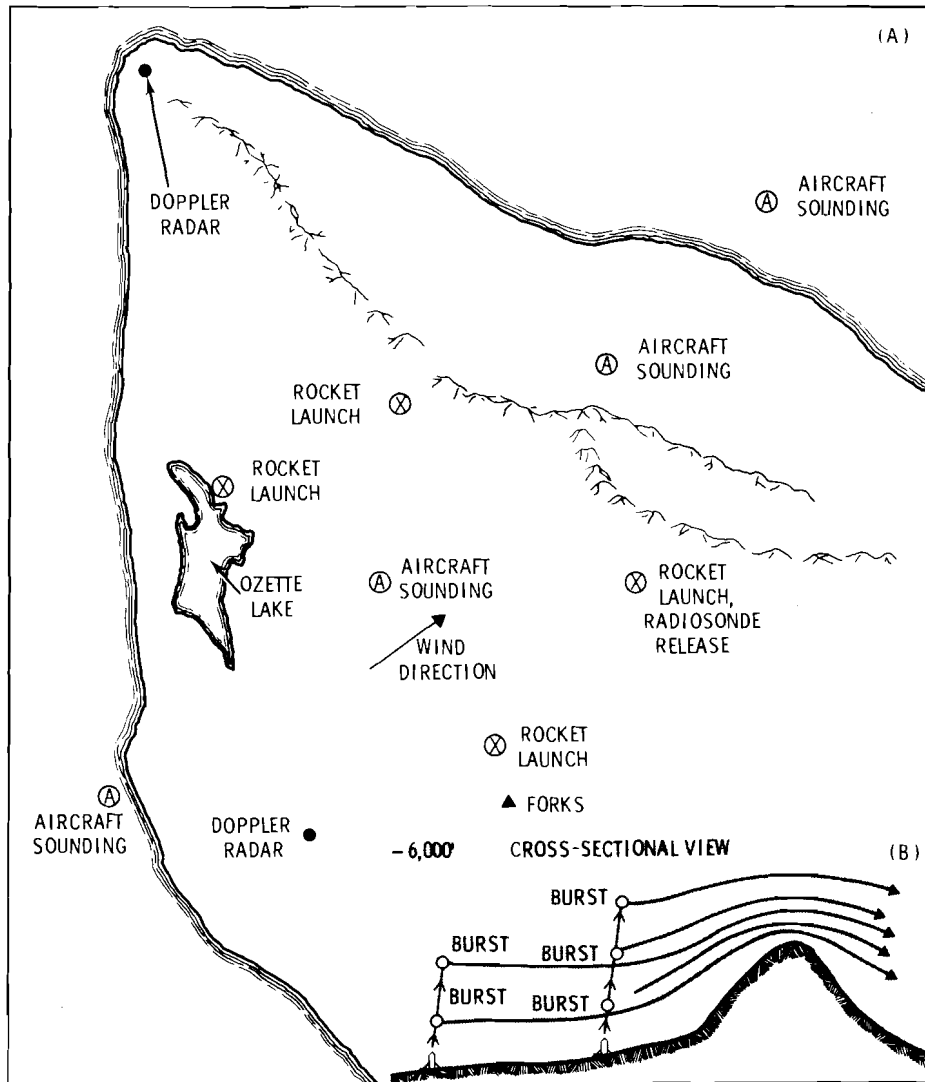


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FIGURE 3. Schematic Vertical Cross Section for Experiment (II)

speed of the chaff; chaff packets must be released high enough to clear the ridge. A schematic vertical cross section of the release altitudes and trajectories is shown in Figure 4b. This experiment would be run on days when the wind direction is roughly perpendicular to the ridge line. Recognizing that these will be directional vertical shear in the wind profile, releases would be made at two cross-

wind locations in order that chaff would be passing over the ridge at the same location at different altitudes at least once. The aircraft soundings would be used to provide stability measurements along a line perpendicular to the ridge, and recording rain gauges would measure the rainfall distribution also along a ridge-perpendicular line.



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FIGURE 4. a. Schematic Plan View of Site Locations for Experiment (III). b. Schematic Vertical Cross Section Showing Idealized Chaff Packet Trajectories for Experiment (III).

A REVIEW OF MOIST MESOSCALE MODELS WITH POSSIBLE
APPLICABILITY TO PRECIPITATION SCAVENGING

C. E. Hane

A current review of moist mesoscale models which might be applied to precipitation scavenging research is described. The need for modeling of this kind in precipitation scavenging is discussed. Nine models which are currently operative or under development are compared based on 16 criteria. The review is incomplete, but preliminary recommendations are given regarding the more promising models for the scavenging program.

A new area in the precipitation scavenging research program which has been initiated in the last few months is a review of presently existing moist mesoscale numerical models. This review has been undertaken to develop within the program a mesoscale model into which precipitation scavenging concepts in frontal storms may be integrated. The model formulation will not replace the field experiments, but act with the field experiments in a combined model-observational approach, each component dependent upon the other. The mesoscale is not the only scale involved in precipitation scavenging; the cloud and microphysical scales are also important and must be involved in the model formulation. With this in mind, some general objectives of the modeling program may be stated:

1. To isolate or identify those mechanisms most important in determining how much material will be placed on the surface. Controlled experiments with systematic variation of one parameter at a time allow the role of a particular parameter to be isolated. Bits and pieces of the scavenging process are understood, but individual components are so interrelated and complex that a model is needed for the integrated picture;
2. To indicate the type and location of critical field experiments, resulting in better-designed experi-

ments. Field experiments are used, of course, for model verification. Following verification, a great variety of natural conditions may be examined at much lower cost than with many field experiments.

Benefits which might be realized from a mesoscale model, in particular, include:

1. The model could provide environmental conditions for a more detailed microphysical model incorporating precipitation scavenging calculations. These environmental parameters include horizontal wind speed, temperature, humidity, and mesoscale vertical motion at various levels in the atmosphere;
2. Such a model could provide information on changes in the large scale flow resulting from terrain-induced or land-sea induced horizontal and vertical air motion. Such information could be used to interpret the results of past field experiments;
3. The model could provide information to aid in planning future field experiments, such as supplying air motions in the frontal rain areas. Determinations might be made, for example, of whether tracer releases upwind of frontal rain bands might be incorporated into the cloud at various altitudes.

With regard to the latter usage, past tracer releases have been made within the rain-producing clouds of frontal storms. This is clearly unrealistic if one is concerned with the scavenging of pollutants by frontal rain, since the pollutant would have its source near the ground below cloud base in most cases and would be delivered to the cloud layer through cloud base by large-scale, forced lifting over large horizontal distances. In-cloud tracer releases are perhaps more realistic in experiments designed to simulate the scavenging of materials incorporated into the cloud above cloud base by horizontal motions relative to the cloud. However, even in this case, the three-dimensional distribution of tracer concentrations within the cloud resulting from an aircraft release may bear little resemblance to that distribution resulting from a plume of material converging upon a cloud due to the relative motions of the cloud and plume. This is not to say that experiments involving in-cloud tracer releases are without value; these experiments may yield relative suggestions (as opposed to absolute estimates) of what would occur as a result of variation of experimental parameters (e.g., hygroscopic versus nonhygroscopic tracer, variations in release height within the cloud, variation in tracer particle sizes) to simulate natural variations of material plumes. It would seem that a worthwhile goal of future field experiments, however, should be to strive for tracer releases outside the condensed cloud.

There is a potentially important role for the mesoscale model to play with regard to the above discussion. The model might be used, for instance, to simulate aircraft tracer releases and (with the incorporation of a microphysical sub-model) deposition fluxes, and to determine whether, in a release outside the rain cloud, sufficient tracer may be concentrated in the fallen rain to permit a meaningful analysis. There is also the possibility of using urban pollutant directly as the material to be scavenged by the frontal storms in field experiments and in the model, as has been done in a case study of scavenging by convective storms in the St. Louis urban area.² The model might be utilized in this frontal storm case to determine whether significant concentrations of pollutant exist in fallen

precipitation (i.e., to aid in the planning of field experiments).

With these needs in mind, the following summary of mesoscale moist numerical models is presented. One model, or a modification thereof, will eventually be selected and utilized to satisfy as many as possible of the above needs existing within the scavenging program. In all, nine models were reviewed, including most (but probably not all) moist mesoscale models which have been formulated. The ordering of the models is done according to the temporal sequence in which they were located in the literature.

The table includes 16 bases for comparison for each of the 9 models. The superscript following the year in which the model was described in the literature (column 1) refers to the numbering in the list of references.

The models may be categorized in a number of ways. An obvious difference, for instance, is in the horizontal scale or grid point separation, according to which, Models 1-5 may be separated from 6-9. The vertical structure of most of the models is similar, with 6-12 constant height or constant pressure levels, with the exception of Model 3, which contains a single mixed layer with upper and lower fluxes supplied by boundary layers not governed directly by time-dependent equations. Also, Model 5 includes increased vertical resolution compared to the other models. The variables predicted in all of the models are for the most part the same: two horizontal wind components, pressure on constant height surfaces or height of constant pressure surfaces, temperature, water vapor, and vertical air motion (determined diagnostically). Another important criterion is the cost involved in running the model. These costs are very difficult to categorize since they vary over a wide spectrum; Model 3 is least expensive to run since it is not completely three-dimensional and Model 5 will be the most expensive when it reaches the final planned stage of development.

Judgments on the best model for the scavenging program are extremely preliminary; priorities have not been established at this time regarding the modeling needs of the scavenging program, nor have the resources available for the development and running

TABLE 1. Summary of Moist Mesoscale Models

AUTHORS	APPLICATIONS	DIMENSIONS	HORIZONTAL SCALE	NUMBER OF GRID PTS (x, y, z)	VERTICAL STRUCTURE	GRID SCALE VARIABLES† (* INDOTES DERIVED QUANT)	HYDROSTATIC?	HOW IS PRECIPITATION HANDLED? CONVECTION INCLUDED?	OROGRAPHY?	BOUNDARY LAYER FORMULATION	RADIATION?	BOUNDARY CONDITIONS	INITIALIZATION PROCEDURE	RUNNING?	COMPARED WITH OBS?	COMMENTS ON INCLUSION OF SCAVENGING - COMPUTATION COSTS
1. KREITZBERG, PERKEY (1974)2 AFGL RPT.	COLD FRONTAL RAIN WITH IMBEDDED CONVECTION	3	100 km AND 35 km	40x20x12	12 CONSTANT HEIGHT LAYERS TO 10.55 km	$u, v, p, T, q, \rho, \theta, w$ *	YES	STABLE PRECIP. AUTOCONVERSION COLLECTION, EVAPORATION, CONVECTIVE PRECIP. - ALSO INCLUDED BY 1-D SUBMODEL	NO	50 m SURFACE CONTACT LAYER ALLOWS FOR FLUXES OF HEAT, MOMENTUM AND MOISTURE	NO	BOUNDARY CONDITIONS (LATERAL) SUPPLIED BY SYNOPTIC SCALE FORECAST.	THE 35 km MESOSCALE GRID IS INITIALIZED FROM THE 1.25° (400 km) GRID FORECAST.	YES	YES. REASONABLE AGREEMENT ~ 24 HRS.	HISTORY TAPES WITH THE RESULTS OF HYDRODYNAMIC MODEL CALCULATION ARE AVAILABLE FOR TRAJECTORY CALCULATIONS. RUNNING MODEL FOR A NEW CASE PROHIBITIVE BECAUSE OF COMPUTER TIME. AUTHORS HAVE ALREADY INCLUDED SOME CRUDE SCAVENGING CALCULATIONS. 24 HOUR FORECAST TAKES 40 HOURS ON CDC 6600.
2. BUSHBY TIMPSON (1967)3 QJRM5	COLD FRONTAL RAIN	3	40 km	95x63x10	10 CONSTANT PRESSURE LAYERS TO 100 mb	$u, v, h, \theta, q, \rho, w$ *	YES	STABLE PRECIP ONLY. EXCESS LIQUID WATER IN LAYER PASSED TO LAYER BELOW. EVAPORATION.	NO	FRICTIONLESS, INVISCID	NO	BOUNDARY CONDITION ON WIND EXTRAPOLATED FROM INTERIOR.	INTERPOLATION FROM RADIOSONDE DATA OF GEOPOTENTIAL HEIGHT AND MOISTURE.	YES	YES. FRONTAL POSITIONS FORECAST REASONABLY AFTER 24 HOURS.	SCAVENGING CALCULATIONS COULD BE INCLUDED SINCE AIR MOTIONS AND LIQUID WATER ARE INCLUDED. HOWEVER, RUNNING MODEL (800 AT • 24 HOUR FORECAST), MODERATELY EXPENSIVE.
3. LAVOIE (1972)4 JAS	LAKE EFFECT STORMS	2+	6 km (dx) 12 km (dy)	50x60x3	MIXED LAYER WITH BOUND. COMDS. SUPPLIED FROM LAYERS ABOVE AND BELOW	$u, v, h, \theta, q, w, \theta^*$	YES	PRECIP. IS ENTIRELY CONVECTIVE AND PARAMETERIZED IN TERMS OF CLOUD DEPTH AND VERTICAL VELOCITY AT CLOUD BASE.	YES	B. L. IS 50 m THICK AND BULK AERODYNAMIC METHOD USED TO COMPUTE FLUX OF MOM., HT., MOIS.	NO	LATERAL B. C.'S EXTRAPOLATED FROM INTERIOR	INTERPOLATION FROM SYNOPTIC SCALE FORECAST MADE ON 127 km GRID.	YES	YES. AGREEMENT BETWEEN PREDICTED AND OBSERVED PRECIP. PATTERN IS EXCELLENT.	MODEL RELATIVELY ECONOMICAL TO RUN, BUT ASSUMPTION OF MIXED LAYER MAY HAVE TO BE ALTERED. SCAVENGING CALCULATIONS BE INCLUDED IN FINAL 3-D MODEL (OR POSSIBLY 2-D) USING ENVIRONMENTAL QUANTITIES SUPPLIED BY THIS MODEL.
4. PAINE KAPLAN (1972)5 JAS	WATER INDUCED HEAT ISLAND. MESOSCALE FORECASTS INVOLVING SEVERE STORM OUTBREAKS	3	25-32 km	36x32x9 44x34x10 49x49x10	9 CONSTANT PRESSURE SURFACES TO 100 mb	$u, v, h, T, q, \rho, \theta, w$ *	YES	STABLE PRECIP. ONLY. EXCESS WATER IN LAYER PASSED TO LAYER BELOW. STORAGE TERM. EVAPORATION.	YES	B. L. FROM SURFACE TO 950 mb OR 875 mb. BULK AERODYNAMICS USED TO COMPUTE FLUXES	NO	LATERAL B. C.'S EXTRAPOLATED FROM INTERIOR	INTERPOLATION FROM SYNOPTIC SCALE FORECAST MADE ON 127 km GRID.	YES	YES. REASONABLE AGREEMENT IN 9. SNOWFALL PATTERN AND RADAR PATTERN.)	SCAVENGING CALCULATIONS COULD BE INCLUDED AS A PART OF TIME DEPENDENT CALCULATIONS. RUNNING MODEL, HOWEVER, WOULD PROBABLY BE COSTLY. 3-D AIR MOTIONS AND LIQUID WATER ARE MODEL OUTPUT.
5. GEUL (1974)6 SESAME OPENING VOLUME	URBAN HEAT ISLAND AND SQUALL LINE EVOLUTION	3	~ 1-3 km (est.)	100x100x30	30 CONSTANT HEIGHT LEVELS TO TROPOPAUSE. VARIABLE SPACING	$u, v, w, \theta, T, q, \rho, \theta, \theta^*$	NO	MAY PARAMETERIZE MOIST CONVECTION EVENTUALLY.	NOT SPECIFIED	UNSPECIFIED	YES	OPEN BOUNDARIES. PERMITTING OUTSIDE CHANGES TO BE TAKEN INTO ACCOUNT AND FOR OUTFLOW OF PERTURBATIONS	UNSPECIFIED, BUT PRESUMABLY OBTAINED FROM LARGER SCALE MODEL OR OBSERVATIONS.	NO. IN PROCESS OF DEVELOPMENT. 2-D IS RUNNING.	NO	3-D AIR MOTIONS AND LIQUID WATER WILL BE COMPUTED IN MODEL. THIS MODEL WILL BE EXTREMELY COSTLY TO RUN WITH ITS LARGE CORE REQUIREMENTS.
6. JONES (BMJ) (1974)7 SUBSYNOPTIC EXTRATROPICAL WEATHER SYSTEMS	COLD FRONTAL RAIN (THIS IS AN EXTENSION OF 12) ABOVE	3	100 km	64x64x10	10 CONSTANT PRESSURE LEVELS TO 100 mb	$u, v, h, \theta, q, \rho, w$ *	YES	SAME AS 12) ABOVE	YES	FRICTION, HEAT AND MOISTURE TRANSFER FROM SURFACE INCLUDED	NO	BOUNDARY CONDITIONS SUPPLIED BY LARGER SCALE FORECAST	HEIGHT FIELD ANALYZED AND WIND FROM BALANCE EQUATION.	YES	YES. QUITE ACCURATE IN PREDICTING FRONTAL RAINBANDS.	MODERATELY EXPENSIVE. SCAVENGING CALCULATIONS POSSIBLE. COMPUTER TIME CUT BY SBNL IMPLICIT SCHEME. 36 HOUR FORECAST TAKES 50 MINUTES COMPUTER TIME.
7. OKAMURA (JMA) (1974)8 SUBSYNOPTIC EXTRATROPICAL WEATHER SYSTEMS	DISTURBANCES OF TIME INTERVAL ~ 1 DAY AND 100 km SPATIAL WAVE-LENGTH	3	15x4 km	37x33x6	6 PRESSURE LEVELS (LAYERS) CENTERED AT 900, 850, 750, 650, 500, 300 mb	u, v, h, T, q, ρ, w *	YES	PRECIPITATION INCLUDED ONLY CRUDELY.	YES	BULK AERODYNAMIC MOMENTUM, HEAT, AND MOISTURE FLUX BETWEEN EARTH AND LOWEST LAYER	NO	BOUNDARY CONDITIONS SUPPLIED BY LARGER SCALE FORECAST.	STREAM FUNCTION AND VELOCITY POTENTIAL DERIVED FROM BALANCE EQUATION AND CONTINUITY EQUATION.	YES	YES. LOCATION OF FEATURES CORRECT AFTER 24 HOURS. LOWS NOT DEEP ENOUGH.	SCAVENGING INCLUSION POSSIBLE. WITH 3-D MOTION FIELD AND WATER VAPOR, 24 HOUR FORECAST TAKES 27 MINUTES ON THE HITAC 8700/8800 (JAPANESE) COMPUTER.
8. BODIN (1974)9 SUBSYNOPTIC EXTRATROPICAL WEATHER SYSTEMS	SUBSYNOPTIC WEATHER SYSTEMS OF LARGER SCALE THAN GRAVITY WAVES (QUASI - GEOSTROPHIC MODEL)	3	150 km	~ 1280 x 10 IRREGULAR SHAPE	10 CONSTANT PRESSURE LEVELS TO 100 mb	u, v, z, T, q, w *	YES	CALCULATION OF PRECIPITABLE INTEGRATED AMOUNT OF PRECIPITATION.	YES	RYAN SUCION AND BULK AERODYNAMIC SENSIBLE HEAT TRANSPORT. NO SURFACE EVAPORATION.	NO	SUPPLIED FROM COARSE MESH AT EVERY TIME STEP.	OBSERVED DATA INTERPOLATED TO GRID.	YES	NOT INDICATED	SCAVENGING INCORPORATION WOULD BE EXTREMELY CRUDE SINCE INTEGRATED PRECIPITATION ONLY IS AVAILABLE. QUASI-GEOSTROPHIC MODEL WOULD INVOLVE LESS COMPUTER TIME THAN PM MODELS.
9. NEAR LAM (1975)10 SUBSYNOPTIC EXTRATROPICAL WEATHER SYSTEMS	SYNOPTIC AND SUBSYNOPTIC EXTRATROPICAL WEATHER SYSTEMS	3	66.4 km 138.8 km 277.5 km (CAN BE VARIED)	144x86x12 OR 24 (MAXIMUM)	6, 12, OR 24 CONSTANT HEIGHT LEVELS	u, v, ρ, T, q, w *	YES	ALL CONDENSED WATER FALLS IMMEDIATELY. NO RE-EVAPORATION. CONVECTIVE ADJUSTMENT SCHEME OR KDD - KRISHNAMURTI CONVECTIVE PARAMETERIZATION.	YES	SURFACE BOUNDARY LAYER INCLUDED. BULK AERO-DYNAMICS	YES	INFLOW-OUTFLOW CONDITION. INFLOW, GCM SUPPLIES B. C.'S. OUTFLOW, EXTRAPOLATED FROM INTERIOR.	OBSERVED GEOPOTENTIAL INTERPOLATED TO GRID PITS. BY HAND.	YES	YES. REASONABLE AGREEMENT. DETAIL MISSING FROM LARGE DX FORECASTS.	SCAVENGING INCLUSION POSSIBLE. HOWEVER, COMPUTER COSTS ARE VERY LARGE.

+ EXPLANATION OF SYMBOLS:

- u , COMPONENT OF AIR MOTION IN X-DIRECTION

v , COMPONENT OF AIR MOTION IN Y-DIRECTION

ρ , PRESSURE

q , MIXING RATIO OF CONDENSED WATER OR VAPOR

T , TEMPERATURE
- q_w , WATER VAPOR MIXING RATIO

w , COMPONENT OF AIR MOTION IN Z-DIRECTION

θ , POTENTIAL TEMPERATURE

w , TIME RATE OF CHANGE OF PRESSURE
- h , HEIGHT OF CONSTANT PRESSURE SURFACE

n , THICKNESS BETWEEN CONSTANT PRESSURE SURFACES

θ_n , VERTICAL MOTION AT TOP OF MIXED LAYER

q_w , CONDENSED WATER MIXING RATIO

of such a model been established. Of the presently running models, Model 1 seems most valuable to the scavenging program (especially if orographic effects are added) since the effects of convection imbedded in the mesoscale environment are included in the model. However, the expenses involved in running Model 1 are quite large. The possibility exists for using the history tapes from Model 1 to provide a mesoscale framework within which to do scavenging calculations for planning purposes, etc.; however, to apply the model to past cases where scavenging field experiments have been done may not be financially feasible. Model 1 has been used by Kreitzberg, Lutz, and Perkey to compute what they call "precipitation cleansing" or the time-integrated stable and convective precipitation passing through parcels whose three-dimensional trajectories are computed by the model.¹¹ Such a computation might be extended to include the details of the precipitation scavenging process in both convective and stable rainfall, as suggested by Perkey.¹²

Model 3, in contrast to Model 1, appears quite promising from the standpoint of computer resource requirements. Its applicability is somewhat limited, however; 1) it is restricted to problems in the low levels, and 2) it is in its present form restricted to dealing with a deep quasi-adiabatic layer. It does take orography into account and works quite well in cases of "lake effect storms." It may be well suited to dealing with storms coming off the ocean over rugged terrain, but would have to be modified to deal with the generally nonadiabatic conditions existing over land areas within rain areas associated with extratropical cyclones. Model 3 could probably best be used for atmospheric transport and to calculate environmental and boundary parameters for a microphysical submodel dealing with the precipitation scavenging processes.

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AN ESTIMATE OF IN-CLOUD SCAVENGING OF A TRACER
INJECTED INTO TWO FRONTAL STORMS

W. E. Davis

Results are presented from the application of a model developed to compute surface deposition rates. These rates were compared with calculated rates of the deposition from In tracer released in prefrontal clouds on two occasions. The first case, March 11, 1971, resulted in an estimated in-cloud scavenging coefficient of 10^{-3} - 10^{-2} sec^{-1} , and the second case, December 5, 1973, resulted in an estimated in-cloud scavenging coefficient of 10^{-4} - 10^{-3} sec^{-1} for the first four of five arcs. The deposition on the fifth arc for December 5, 1973 was greater than computed by the model.

INTRODUCTION

Air pollution removal by precipitation is important as a cleansing agent for our air.¹ One of the ways in which air pollution can be deposited in precipitation is the in-cloud scavenging of the pollution through cloud water and precipitation of picking up and removing pollution. These removal processes can occur in both convective storms and frontal storms. (For studies of the processes in convective storms, see Burtsev, et al., 1968,² Dingle, et al., 1968,³ Summers, 1972,⁴ Warburton, 1973,⁵ and Young, et al., 1974.⁶) The study presented here was initiated to determine the effectiveness of these removal processes in frontal storms by the injection of a tracer, In, into frontal storms.

In-cloud scavenging has been studied through the building of a model that incorporates the in-cloud scavenging process, the removal and the deposition of air pollution in precipitation.^{7,8,9,10} The model results may be compared with observed deposition from the tracer injected into a rain-producing cloud during a frontal storm. This comparison permits an estimate to be made of the

effectiveness of in-cloud scavenging in the removal process.

In the past 4 years, injections of a tracer, In, have been made into frontal storms near Quillayute, Washington, on the Olympic Peninsula.¹¹ Here we attempt to compare a model's computed deposition rates with the rates calculated from observed deposition patterns. The aim is to determine the effectiveness of in-cloud scavenging.

MODEL

The model previously reported by Davis^{8,9} was chosen because neither in-cloud measurements nor information concerning the tracer were detailed enough to permit use of the Dingle and Lee model.¹⁰ The Davis model is based on a three-phase process. The first phase is the interaction of the tracer with the cloud water. In this case the transfer of material, In, from air to cloud water, will be referred to as in-cloud scavenging, λ . The second phase, removal of In from the cloud water by rain or snow, will be referred to as dilution, ψ . The third phase is the transfer of In by rain or snow to the ground.

For a detailed account of the model and assumptions, see References 8 and 9.

CASE I. MARCH 11, 1971

Results of an analysis of surface deposition to compute a surface deposition have been reported by Davis.¹² Only the comparison of the model curves with the calculated deposition rate will be reported.

In comparing the model's curves with derived deposition rate curves (Figure 1), the curves that best approximate the derived curve are those with the in-cloud scavenging coefficient, λ between 10^{-2} sec^{-1} - 10^{-3} sec^{-1} . Finer resolution is difficult since varying λ and ψ can produce the same deposition rates.^{12,13} Some limits to ψ and λ can be calculated. As shown in the figure, the curve with $\psi = 2 \times 10^{-4} \text{ sec}^{-1}$ and $\lambda < 10^0 \text{ sec}^{-1}$ yields deposition rates lower than those needed to explain the calculated deposition rates. Therefore, one could conclude that ψ is greater than $2 \times 10^{-4} \text{ sec}^{-1}$. An independent estimate of ψ was made¹⁴ based on assuming as constant a derived liquid water content of 0.2 g/m^3 and a mean rate of rise as determined from aircraft and radiosonde data.¹² The dilution coefficient ψ was computed to be $4 \times 10^{-4} \text{ sec}^{-1}$ - $2 \times 10^{-3} \text{ sec}^{-1}$. These values are greater than the minimum of $2 \times 10^{-4} \text{ sec}^{-1}$ indicated in the modeling study, and are in the same range of values for ψ which with the λ computed above would explain the calculated deposition rate.

CASE II. DECEMBER 5, 1973

Results of the analysis of surface deposition have been reported by Davis and Young;¹¹ only the comparison of the model's curves and the calculated deposition rate will be reported.

The model curves which best approximate the derived curve out to the fourth arc are those with an in-cloud scavenging coefficient, λ between 10^{-4} sec^{-1} - 10^{-3} sec^{-1} . The increase in deposition rate on the fifth arc could not be explained by the model. Also, as shown in the figure, the curve with $\psi = 10^{-5} \text{ sec}^{-1}$ and $\lambda < 10^0 \text{ sec}^{-1}$ yields deposition rates too low to explain the calculated deposition rates. Hence one could conclude that ψ , the removal of In in the cloud water by rain or snow, is greater than 10^{-5} sec^{-1} .

An independent estimate of ψ was difficult, because no measurement of liquid water or temperature structure downwind of the release point was made during the test. An estimate of liquid water was made using the temperature at release point and the lapse rate together with observations of liquid water in frontal storms¹⁵ of $\sim 0.30 \text{ g/m}^3$. Two methods were used to estimate ψ . The first took the change in height along the trajectory path as equal to the change in mean elevation as the parcel moved across the peninsula. The second was based on the mean rise of the air parcel necessary to produce the observed precipitation. In both cases the liquid water content was assumed constant and the amount of water removed from the air parcel was assumed to be the excess above saturation. The resulting dilution coefficient was estimated at $\sim 7 \times 10^{-4} \text{ sec}^{-1}$ for the second method. These values were greater than the minimum values estimated by the model of $1 \times 10^{-5} \text{ sec}^{-1}$ and in the same range as those computed by the model. One should note that the observations of liquid water content in frontal storms, as reported by Ponomarenko¹⁵ did have a minimum of 0.08 g/m^3 and a maximum of 0.78 g/m^3 . This would extend the range of estimated ψ dilution coefficient, from $\sim 1 \times 10^{-4} \text{ sec}^{-1}$ - $\sim 3 \times 10^{-3} \text{ sec}^{-1}$. Again these values are within the acceptable range of ψ to explain the calculated deposition pattern.

COMPARISON OF CASE I AND CASE II.

There are some definite differences in the two cases. First, the wind speeds varied from 30 m/sec in the March 11 case to 22 m/sec in the December 5 case. Also, the point of release although at the same altitude was made further to the south for Case II. The percent deposited was about the same, $\sim 30\%$ for March 11, 1971, versus $22\text{-}24\%$ December 5, 1973. The wind speed and increase of distance produced a time of deposition of $\sim 55 \text{ min}$ in the December 5 case, versus $\sim 15 \text{ min}$ in the March 11 case. Thus, as one would expect, the rate of deposition for December 5 was less than that for March 11.

Both cases show increasing deposition rate with time. In the March 11 case, the last two of the three arcs

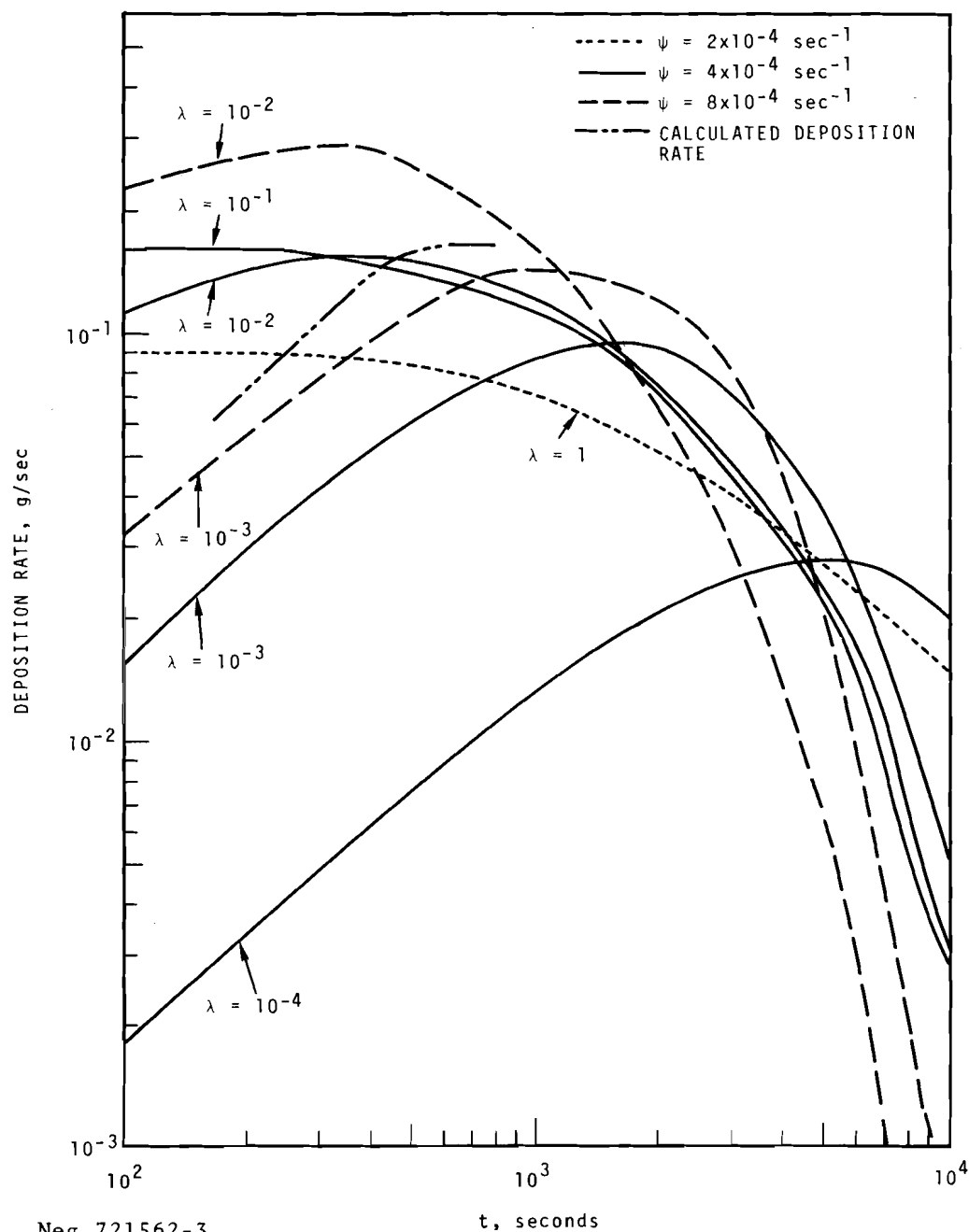


FIGURE 1. Comparison of Model's Deposition Rates with Calculated Deposition Rates for March 11, 1971 Storm

crossed showed essentially the same rate deposition. In the case of December 5 the trend was toward increasing deposition with time except on Arc 4 which showed a decrease. This trend toward increasing deposition is shown by curves in the model with λ ranging from 10^{-3} - 10^{-4} sec^{-1} .

The most interesting feature of either deposition is the increase on Arc 5 of the December 5, 1973 case. The model could not account for this increase. When the estimated mean air mass rise was imposed, the tracer injected air parcel would have risen

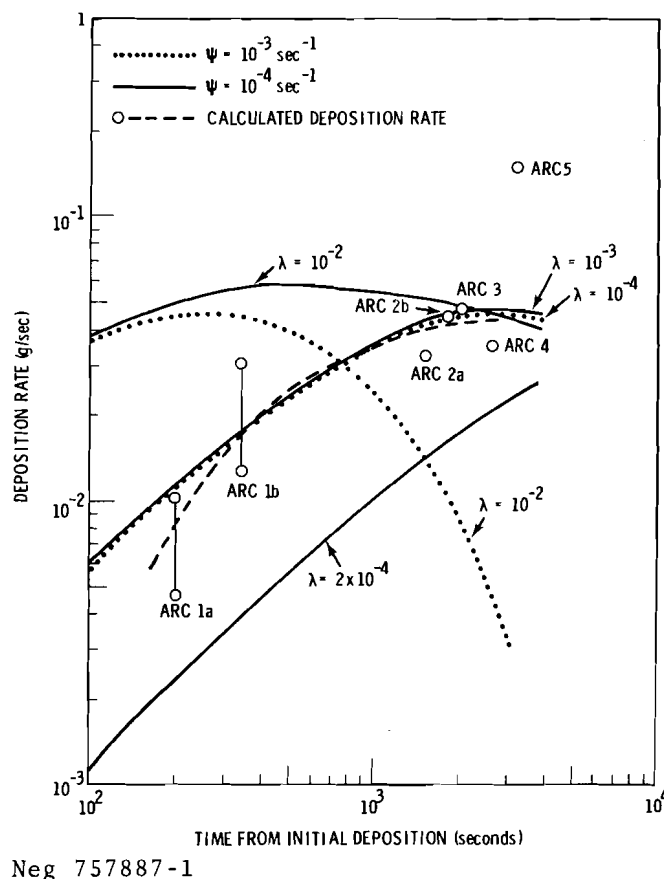


FIGURE 2. Comparison of Model's Deposition Rates with Calculated Deposition Rates for December 5, 1973 Storm

to a height where the temperature was $\leq 0^\circ\text{C}$ before the time when removal would have deposited material on the fifth arc. It may be that the melting of falling snow and freezing of cloud droplets as well as the observed increase in the precipitation rate could have enhanced the removal rate. One should note that In released on March 11 was injected at $\sim 0^\circ\text{C}$ - -1.5°C level and at a higher mean precipitation rate,¹⁴ result in a higher calculated deposition rate.

DISCUSSION OF RESULTS AND CONCLUSIONS

Two cases have been compared where In has been injected by flares into frontal storms. A range of values for in-cloud scavenging coefficients was determined for each storm with different results. For the March 11, 1971 storm a range of the in-cloud

scavenging coefficient λ , of 10^{-2} sec^{-1} - 10^{-3} sec^{-1} was calculated. For the December 5, 1973 storm a λ was calculated between 10^{-3} sec^{-1} and 10^{-4} sec^{-1} for four of five arcs. The range of estimated ψ 's for the March 11, 1971 and December 5, 1973 cases were $4 \times 10^{-4}\text{ sec}^{-1}$ - $2 \times 10^{-3}\text{ sec}^{-1}$ for March 11 and $3 \times 10^{-4}\text{ sec}^{-1}$ - $7 \times 10^{-4}\text{ sec}^{-1}$ for December 5.

Two measurements that can be compared are 1) the injection temperature of 2°C for the December 5 case versus -1.8°C - 0°C for the March 11 case and 2) the mean precipitation rate of 4. mm/hr - 5. mm/hr for the March 11 case versus 0.7 mm/hr - 2. mm/hr for the December 5 case. The possibility exists that the freezing and melting process as well as the increased precipitation rate could have enhanced In removal. It is suggested

that more work with additional in-cloud measurements be done in order to test this suggestion.

Dingle and Lee,¹⁰ as a result of their model studies found agreement with Makhon'ko⁷ and Davis⁹ estimates of the in-cloud scavenging coefficient (designated the diffusive attachment rate between particles and droplets by Dingle and Lee) and the dilution coefficient (designated the rate of accretion of droplets by raindrops by Dingle and Lee). The values they arrive at by a model¹⁰ were for λ , 10^{-5} - 10^{-4} sec⁻¹, and for ψ , 10^{-4} - 10^{-3} sec⁻¹. Davis¹² indicated values of $\lambda > 10^{-4}$ sec⁻¹. Makhon'ko⁷ indicated a value of ψ ranging from 0.4×10^{-4} sec⁻¹- 22×10^{-4} sec⁻¹. Based on values reported by Makhon'ko an in-cloud scavenging coefficient was calculated from 3×10^{-5} sec⁻¹ for Ra- 2.2×10^{-3} sec⁻¹ for dissolved inorganic contaminant. The results of the In analysis indicate that the value for λ in the case of In may extend

higher than the values arrived at by Dingle and Lee¹⁰ and the computed values from Makhon'ko, with λ as great as 10^{-2} sec⁻¹ for the March 11 case.

Considerable caution should be taken in view of the assumptions that were made. Also, these values should be considered preliminary because of the lack of sufficient in-cloud measurements. Other tests should be made which will include not only sequential sampling at the surface but in-cloud measurements of liquid water, cloud drops sizes, ice crystal types, temperature and wind speed. Additional efforts should be made to size the tracer particles from the In flares. With these additional measurements it may be possible to arrive at a conclusive explanation for the difference in the in-cloud scavenging coefficient λ , 10^{-3} - 10^{-2} sec⁻¹ for the December 5, 1973 case versus the λ of 10^{-4} - 10^{-3} sec⁻¹ for the March 11, 1971 case.

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DEVELOPMENT OF A DC-3 AIRCRAFT
FOR ATMOSPHERIC POLLUTION RESEARCH

J. M. Hales

This report summarizes progress to date in modifying the Battelle-Northwest DC-3 aircraft for research application in the scavenging and pollutant analysis programs. The aircraft, purchased in December 1974, has operated in two major field programs thus far, and is expected to be an important component of the forthcoming MAP3S project.

INTRODUCTION

During recent years a strong need for a heavier research aircraft has arisen within the Battelle-Northwest Atmospheric Sciences program. Although felt generally throughout the total program, this need is especially pronounced in the in-cloud scavenging group, who require higher load-carrying capability, longer range, and greater reliability under icing conditions for tracer release and cloud physics measurements. The need is also felt strongly in the aerosol and trace-gas component, where it is necessary to carry a large quantity of pollutant monitoring equipment simultaneously for extended periods of time. These needs have been filled by the purchase and conversion of a DC-3 aircraft, which arrived on-site in December 1974. Acquisition and development of the aircraft up to now are summarized in this report.

CHOICE OF AIRCRAFT

The DC-3 aircraft was chosen over other contenders after systematic evaluation according to a number of criteria. These included capital cost, availability, operating schedule and costs, load-carrying ability, performance under icing conditions, range, cruising speed, ceiling altitude, and capability of the airframe to accept modifications. The initial capital investment for the aircraft was \$37,000. Basic properties of the aircraft are shown in Table 1.

TABLE 1. Basic Aircraft Characteristics--Battelle-Northwest DC-3

Cruising speed	90-150 kn
Payload (with full fuel load, and two crew members)	1500 lb
Range (no reserve)	7 hr
Ceiling altitude	20,000 ft
Icing rating	moderate

CLOUD PHYSICS AND TRACER INSTRUMENTATION

Immediately upon its arrival in December 1974 the aircraft was substantially modified from an executive style to have a scientific application. Since its first scheduled mission was to fly in the in-cloud scavenging program at Quillayute, cloud-physics and tracer release equipment were installed first.

The tracer system used consists of flare racks and acetone generators, both activated remotely from a firing panel in the interior of the aircraft. Flare racks are the standard type used for cloud-seeding, while the acetone generators are the Brad-Patton units from the Battelle Cessna 411 aircraft. The container system for the acetone-tracer solutions was fabricated specially for the DC-3, and consists of six polyethylene-lined,

cylindrical, aluminum containers. These are oriented vertically and mounted adjacent to one wall of the aircraft inside a ventilated Plexiglas cabinet. The volume of each container is approximately 20 l. The containers are connected by manifolds to exit lines running to the acetone burners; the flows from individual tanks are controlled by remotely-activated solenoid valves which permit sequential or simultaneous discharge from the individual vessels. Pressure for the system is provided by a regulated nitrogen cylinder in the rear of the aircraft.

Cloud physics instruments installed during initial aircraft development consisted of a G. E. Aitken nucleus monitor, an ice nucleus counter, and a laser cloud-droplet spectrometer. In addition, a Rosemount temperature sensor and a Cambridge dew-point hygrometer were placed on the aircraft during this period, and provision for chaff releases in support of the Doppler radar system was incorporated by installing a chaff cutting mechanism.

Air monitored by the Aitken and ice nucleus-measuring equipment was sampled using a special isokinetic probe, located on top of the aircraft and protruding through the fuselage to the monitoring units. This probe was fabricated from 2-in. stainless tubing, the leading end being gradually tapered to a 0.5-in. diameter to facilitate isokinetic operation of the probe.

POLLUTION AND SOLAR RADIATION MEASUREMENT FACILITIES

After the Quillayute scavenging program, conducted during March 1975, the research aircraft was further modified. This was in preparation for the St. Louis aerosol and trace-gas transformation study, and included provision for extensive monitoring of pollution levels as well as solar radiation. Equipment installed for pollution monitoring is summarized in Table 2, and the aircraft instrumentation configurations used during the St. Louis experiment are shown in Figure 1.

Solar observations were facilitated by installing an observation dome in the aircraft roof just aft of the cockpit. Measurements obtained

TABLE 2. Summary of Pollution Monitoring Equipment Operated in the BNW DC-3 During 1975 St. Louis Program

<u>Constituent</u>	<u>Monitor</u>
Aerosol	High-volume filter
Aerosol	Light scattering
Aerosol	Electrostatic mobility
Aerosol	Aitken nucleus
SO ₂	Flame-photometric
NO	Chemiluminescence
NO _x	Chemiluminescence
O ₃	Chemiluminescence
Hydrocarbons	Flame-ionization G. C.

to date using the DC-3 have consisted of observations from multiwavelength hand-held instrumentation. More sophisticated tracking devices are contemplated for future application.

DATA ACQUISITION AND ADDITIONAL FEATURES

During this second period of development additional features have been incorporated which involve state measurement and data acquisition. A data acquisition system has been installed consisting of a NOVA 1220 minicomputer linked to a 7-track magnetic tape unit. Analog data signals from the entire array of monitors are fed to this system, where they are multiplexed, digitized, and written onto tape for subsequent batch processing. Temperature, airspeed, pressure, and position signals are conditioned before reaching the computer by a standard Metrodata M-8 interface system. Aircraft position data for this purpose are furnished by VOR/DME radio equipment located in the aft of the aircraft for the scientists' convenience.

FUTURE DEVELOPMENTS

Because of rack mounting and modular configuration of the research equipment, additional units can be mounted on the aircraft as required. At present it is anticipated that additional monitoring equipment for CO, sulfate, and halocarbons will be incorporated within the near future.



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FIGURE 1. Aircraft Interior Showing Configurations

Also a low-frequency navigation system is scheduled to be installed within the coming year. In addition to providing more reliable position data, this unit will be capable of computing wind speeds and thus will add substantially to the existing capability for pollutant material-balance measurements.

The primary intended function of the DC-3 aircraft at present is its future operation in conjunction with the forthcoming MPA3S program. It is expected also to contribute to less extensive field programs prior to im-

plementation of this major undertaking, which is anticipated to begin sometime during FY-1977.

ACKNOWLEDGEMENTS

A great many individuals contributed significantly to the development of this facility during the past year. These include (but are not confined to) A. J. Alkezweeny, K. G. Busness, C. E. Elderkin, O. N. Dodson, R. F. Edwards, F. O. Gladfelder, N. S. Laulainen, R. N. Lee, P. M. Potter, R. G. Rieck, and C. W. Thomas.

A COMPUTER ALGORITHM FOR REDUCTION AND PRESENTATION OF INFORMATION FROM THE BNW DC-3 DATA ACQUISITION SYSTEM

D. R. Drewes

The data acquisition system aboard the BNW DC-3 research aircraft records data, as voltage signals, in a compact format on 7-track magnetic tape. Computer software has been written which converts these data to appropriate units and makes them available for analysis. The user has direct control over the important aspects of the analysis, and the modular nature of the code allows great flexibility in the types of analyses to be performed. In addition to printed output, graphic output may be generated using the highly versatile DISSPLA graphics package.

INTRODUCTION

An efficient data acquisition system must necessarily store a large amount of data as compactly as possible. Data density can be increased by recording fewer parameters, increasing the time between successive recordings of each parameter, decreasing the resolution of each parameter, and by eliminating gaps or formatting in the recording process. The first three options involve decisions by the experimenter to ensure that enough types of data, with sufficient

resolution in both time and data space, are recorded to validate the experiment. The fourth option is more a matter of taste and convenience. Data recorded in this compact form, however, do not lend themselves readily to analysis. Some process must be applied to convert the recorded data to a useful format, while simultaneously reducing the volume of data, by an appropriate averaging or culling process, and possibly also performing various operations on the data.

The data acquisition system aboard the BNW DC-3 research aircraft records binary data with 12-bit resolution for 17 parameters (see Table 1) on 7-track magnetic tape. All parameters except the time and aerosol data are sampled five times per sec and averaged over the 1-sec interval, and the average value is recorded. Computer software has been written to recover the recorded data for further calculations, analysis, and presentation.

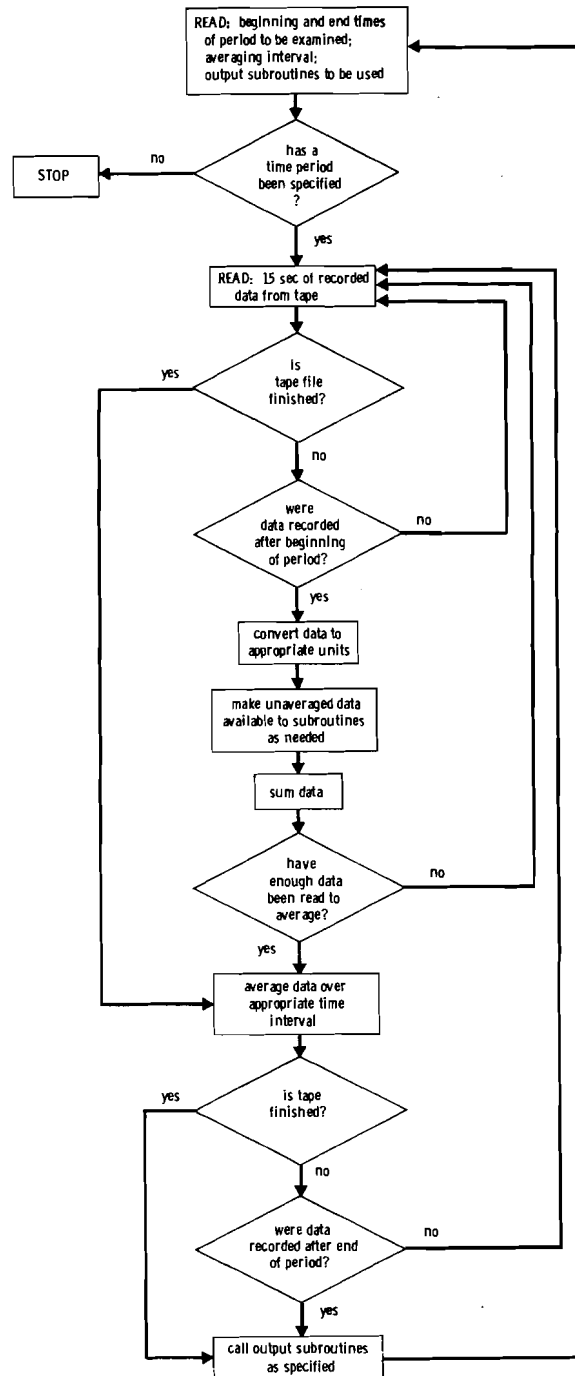
TABLE 1. Data Recorded by DC-3 Data Acquisition System

Channel	Parameter
1	Time (min)
2	Time (sec)
3	Altitude
4	Airspeed
5	VOR 1
6	VOR 2
7	DME
8	Temperature
9	Dewpoint
10	Ozone
11	NO
12	NO ₂
13	Condensation nuclei (data)
14	Condensation nuclei (range)
15	Aerosol (data)
16	Aerosol (range)
17	Aerosol (event mark)

DATA RECOVERY

A schematic flowchart of the FORTRAN software developed for the analysis of the data tapes is shown in Figure 1. The program has been run on the Cyber 73 system, but should be compatible with the FORTRAN system on any large computer.

The program has been written to give the user a great deal of versa-



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FIGURE 1. Schematic Flowchart of the FORTRAN Software

tility in examining the data. Essentially, the program reads data from the 7-track tape, converts it to the appropriate units, averages over a

time interval specified by the user, stores the averaged values and any unaveraged data required, and finally, writes an output file. Each of these functions is performed by a separate subroutine, allowing modification of the analysis to suit the needs of the user. For instance, a minor change in the subroutine which converts the data to the appropriate units would allow the user to see temperatures in °K rather than °C. Likewise, one could replace the simple averaging subroutine with one which calculates an integral average.

Flexibility is more welcome, however, in the routines which calculate the data and print results. There are five subroutines in this category, called OPT1, OPT2, ..., OPT5. OPT4 and OPT5 are reserved primarily for manipulation of the data before averaging. For example, the aerosol data cannot be averaged like the other parameters, so any desired calculations on them must be done in either OPT4 or OPT5. OPT1-OPT3 are primarily reserved for manipulation and output of the time-averaged data, and thus handle, among other things, the graphics.

The user can specify any combination of the five subroutines on any section of the tape.

As presently used, OPT1 lists the time-averaged data. OPT2, using the DISSPLA graphics package, plots many of the parameters versus time. OPT3 performs a regression analysis on the averaged O_3 and NO/NO_2 data, and prints and/or plots the results. OPT4 extracts and prints the aerosol data, and OPT5 prints the SO_2 data as read from data cards. By substituting subroutines which perform other functions for those which are not deemed necessary, however, the user can examine the data in the way which is most beneficial.

Essentially, then, the package may be rather easily modified to accommodate the changing needs of the user, and also any future change in the data recording format. The user is given the flexibility of choosing sections of the tape to be examined, the averaging interval, the number of calculations to be performed, and the output format.

SCAVENGING OF URBAN POLLUTANTS BY THUNDERSTORM
RAINFALL: NUMERICAL EXPERIMENTATION

C. E. Hane

A two-dimensional time-dependent convective cloud model is applied to the squall line case of August 21, 1972, in the St. Louis, Missouri, area. A pollutant plume with dimensions and concentrations characteristic of the St. Louis plume is allowed to interact with the thunderstorm circulation. The scavenging of the pollutant by precipitation is calculated along with the pollutant surface deposition by rainfall. The deposited amounts are compared with the amounts observed in the precipitation from the same storm, resulting in reasonable agreement between model and observations.

INTRODUCTION

It is well known that high concentrations of pollutants are at times found in the lower atmosphere in the immediate vicinity of large urban areas. It is less well known that rainfall can quite efficiently remove pollutants from the urban atmosphere. It is reasonable to suppose that the highest removal efficiencies are achieved by well-organized thunderstorms, whose structure is such that low-level pollutant-bearing air is quickly delivered to the region of the storm where water vapor is condensing, and subsequently travels to the rain formation region. These storms may be contrasted in their scavenging efficiency with rain from clouds produced by large-scale forced lifting (e.g., midlatitude extratropical cyclones), where the pollutant is either removed by falling rain below the clouds (a much less efficient process) or by rising very slowly over large horizontal distances to reach regions of condensation. Each of the latter processes would result in much smaller fluxes of pollutant in rain to the ground than in the case of squall lines, thunderstorm clusters, or large isolated thunderstorms. The research described here seeks to understand how much and where pollutant is deposited on the

ground as a result of thunderstorm passage through an urban pollutant plume.

MODEL APPLICATION

A two-dimensional time-dependent thunderstorm numerical model has been used to produce a realistic thunderstorm circulation using the environmental conditions present in the St. Louis area on August 21, 1972. A steady state is assumed for the storm for the purpose of including precipitation scavenging calculations, utilizing the fields of motion and rain-water mixing ratio at one of the more intense stages during the storm's evolution. For a description of 1) environmental conditions on this day, 2) the storm evolution, and 3) storm structure during the assumed steady-state period, the reader is referred to an earlier paper.¹ The model itself is described in yet another paper.² Previously reported results¹ indicate that 1) in the absence of precipitation scavenging a significant portion of the pollutant is transported to the upper troposphere, and 2) the amount of pollutant reaching the region of the storm containing heavy rain has a significant dependence upon the vertical profile of pollutant concentration in the low-level air, which is the

source of energy for the updraft. In particular, a pollutant with maximum concentration near the ground is more efficiently transported to the heavy rain region than one whose maximum concentration is aloft.

Here, the pollutant is allowed to enter the rainwater via a single process. The following assumptions are made:

1. All pollutant entering the cloud base is immediately incorporated into cloud droplets.
2. The pollutant enters the rain (rain is defined as liquid water moving with a velocity different than the air velocity) when the cloud droplets are accreted by rain.
3. Diffusion of the pollutant is neglected.
4. Evaporation of rain containing pollutant is accounted for only on the boundary of the rain area.
5. Chemical transformation of the pollutant is neglected.
6. Pollutant located where rainwater exists but cloudwater does not (a rare situation) is accreted at the same rate as if it were in cloudwater.

The incorporation of pollutant in cloudwater into the rainwater is accomplished through accretion, represented by the following term:

$$\left(\frac{dq}{dt}\right)_s = E c_p Q / \lambda^{3.5},$$

where $c = 3.08 \times 10^2$ is a dimensioned constant and $\lambda = (2.51 \times 10^2 / q_r)^{1/4}$. In the above expression q is the pollutant-in-rainwater mixing ratio, E is the collection efficiency, ρ is air density, Q is the pollutant-in-cloudwater mixing ratio and q_r is the rainwater mixing ratio. The above term is calculated at the midpoint of the trajectories of pollutant-bearing cloudwater and pollutant-bearing rainwater in the Lagrangian type scheme which is used for the time integrations. The term adds pollutant to the rain and reduces the cloud bearing pollutant at a given point, but by differing amounts, since in gen-

eral the rain and cloud at a given point arrive along different trajectories.

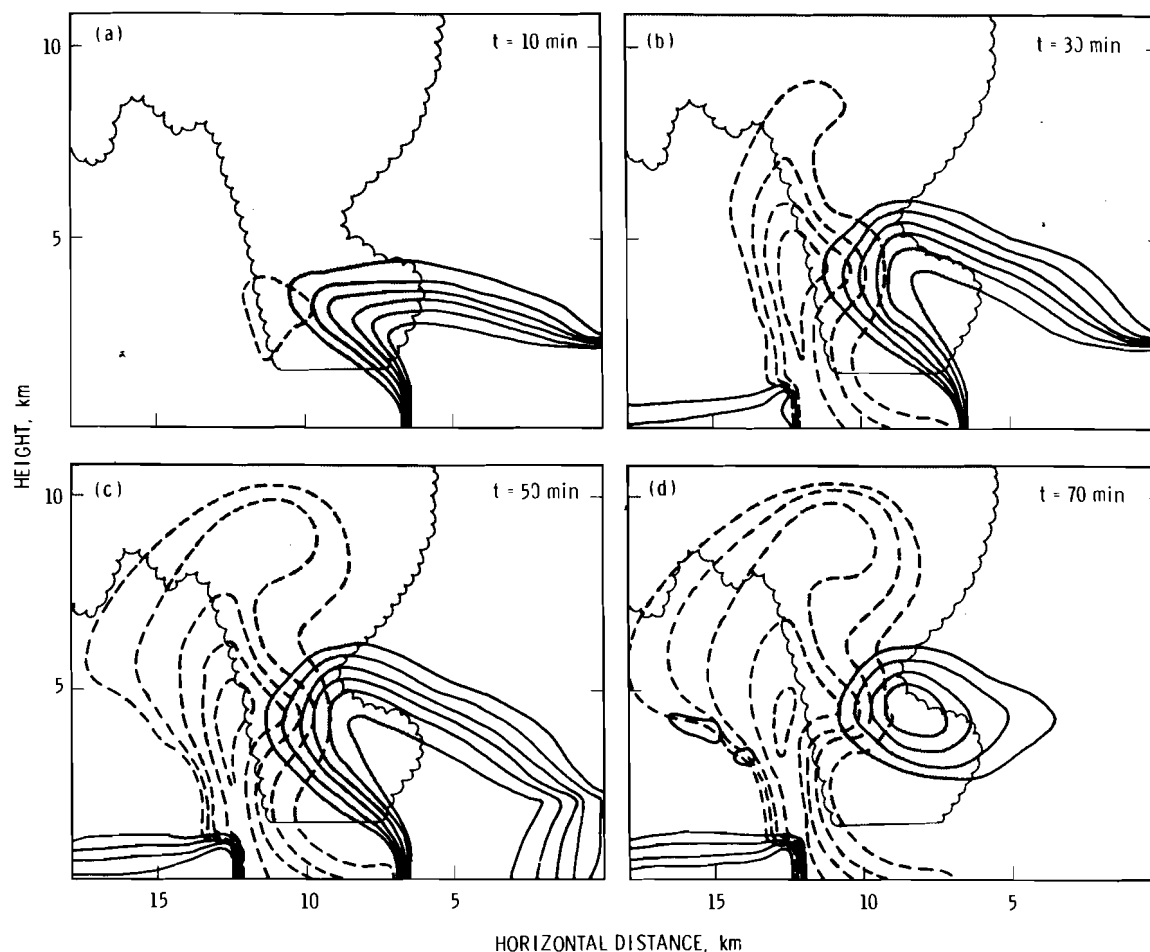
MODEL RESULTS

A number of cases were run, including variations in the initial vertical distribution of pollutant. These runs are summarized in Table 1. In each case the pollutant is initially distributed in the lowest 2.4 km of the atmosphere in a location such that it would begin to enter the cloud updraft region a few minutes later. The pollutant continues to enter through the boundary for a length of time corresponding to an urban plume with a horizontal extent of approximating 22 km. Each case was run for 2 hr model time with pollutant entering through the boundary for the first 45 min. This is shown in Figure 1 where the pollutant in air or cloud distribution, pollutant in rain distribution, and the cloud outline are contoured at four different times (10, 30, 50, and 70 min) in the vertical cross section. The cloud outline is scalloped, the pollutant in air or cloud mixing ratio isolines (contour interval = 2.5×10^{-8} g/g) are solid, and the pollutant in rain mixing ratio isolines are dashed.

At 10 min (Figure 1a) the pollutant is transported into the cloud by the updraft, and the pollutant within the cloud (assumed to be in cloud droplets) is accreted by falling rain, as can be seen along the left edge of the cloud. At 30 min (Figure 1b) the pollutant can be seen to be reaching the ground in the rainfall. The pollutant in rain is also spreading upward for one of two reasons: 1) the pollutant is contained within drops

TABLE 1. Summary of Cases Run Noting Varied Parameters

Case	Collection Efficiency	Type of Pollutant Profile
A	1.0	Low level maximum (surface)
B	0.1	Low level maximum
C	1.0	High level maximum (2.0 km)
D	1.0	Constant with height



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FIGURE 1. Model Cloud Outline, Pollutant in Air or Cloud Distribution, and Pollutant in Rain Distribution at Times (a) 10 min, (b) 30 min, (c) 50 min, and (d) 70 min for Case D.

whose terminal fallspeed is less than the upward air speed or 2) the pollutant is entering the rain at higher levels (the zero contour for pollutant quantities is not drawn). At 50 and 70 min the upward spread is continuing and the distributions are very similar. A quasi-steady state distribution would be achieved except that the urban pollutant plume has been traversed by the storm and no more pollutant enters the cloud after 80-90 min. After the times shown, the pollutant ceases to be carried to the ground; it remains in the rain in the upper portion of the cloud.

The pollutant-in-air maximum found in low levels to the left of the rain

area results from evaporation of rain containing pollutant. This maximum is thought to be exaggerated in these experiments because a time was chosen for the steady-state motion and assumed rainwater fields when very little rain was reaching the ground, although a large amount of rain was present higher in the cloud. This means that the pollutant in rain, as it approaches the ground, decelerates and travels horizontally (to the left) more than it would when the rainwater mixing ratio near the ground was larger. This factor, then, leads to a slight underestimation of the deposition of pollutant on the ground.

In each case a pollutant budget was computed as a function of time

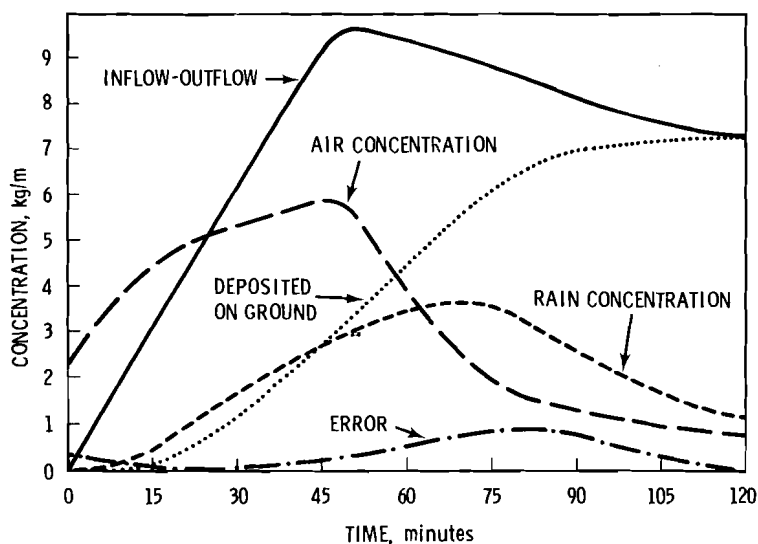
accounting for the following: total pollutant in rain (P1) in the two-dimensional slab per meter slab thickness; total pollutant in air or cloud (P2); cumulative influx minus efflux of pollutant (P3); and cumulative deposition of pollutant in rain on the ground (P4). If P5 represents the amount of pollutant present initially then the following balance is required

$$P5 + P3 = P1 + P2 + P4. \quad (1)$$

An error was also computed as a function of time, defined by the right hand side of (1) minus the left hand side. The result of budget calculations for Case C are shown in Figure 2. The cut-off of inflow at 45 min is evident in the curve representing inflow through the boundaries minus outflow. This cut-off has a direct effect upon the pollutant-in-air concentration, which decreases rapidly from 45 to 75 min when the peak pollutant in rain concentration occurs. The deposition on the ground (cumulative) increases steadily from 15 min until 75 min. After this the accumulation is slower, due to a lesser amount of pollutant in the rain. The error (numerical due to the finite differencing) is small at most times compared to other budget quantities, and certainly does not obscure the important processes. The

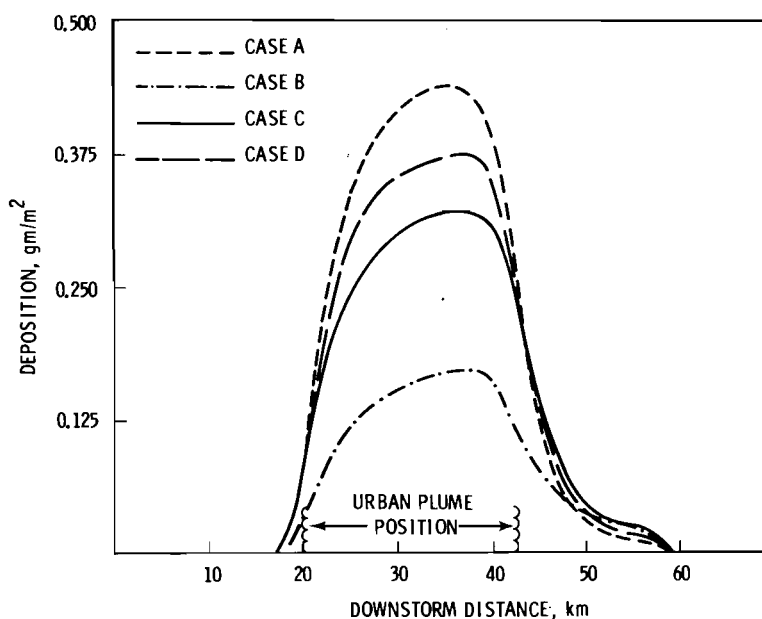
budget results for other cases (not shown) are quite similar. The error in other cases is slightly larger, indicating that the error very likely results from finite difference inaccuracies in computing the flux through the lateral boundary at the surface level (since surface level lateral fluxes are larger in other cases) rather than inaccuracies in the internal computations.

The deposition of pollutant in rain as a function of downstorm distance was also calculated and is shown in Figure 3 for all four cases. It is clear that the largest deposition occurs when the pollutant is concentrated near the ground, rather than concentrated at higher levels in the environmental air. This is because the air near the ground enters the heavy rain area of the cloud more directly than does the air at higher levels in the inflow region. The location of the urban plume relative to the deposition pattern is also shown. The deposition pattern is skewed with maximum deposition 5-10 km downstorm from the middle of the urban plume. This skewness is due to the pollutant being carried by the storm for some time before being deposited on the ground. In addition, the pollutant-bearing air has a component of motion opposite to the direction of storm motion. This air conserves part of



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FIGURE 2. Budget Parameters as a Function of Time for Case C



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FIGURE 3. Pollutant Deposition in Rainfall as a Function of Downstorm Distance for Cases A, B, C, and D

that momentum as it travels through the storm so that the same pollutant can be deposited upstorm from the point where it entered the storm. This combination of factors results in the pollutant being deposited over a distance which is approximately twice that across the urban plume. The curve representing Case B shows the result for a run where the collection efficiency was reduced by a factor of ten compared to Case A. The cumulative deposition in Case A is only slightly more than twice that in Case B despite this change. This lower value of the collection efficiency of rain was used to crudely indicate what sort of changes would occur if some of the pollutant entering the cloud were not contained within cloud droplets. As can be seen a significant amount of deposition occurs in this case.

COMPARISON WITH OBSERVATIONS

An attempt was made to compare the model deposition with the deposition of several pollutants measured for this storm.³ The comparison is made more difficult because 1) during the 1972 field program³ in St. Louis,

precipitation was collected and analyzed at only two downstorm distances, and 2) no data are available for air concentrations of pollutants for model input. Therefore only two observed points will be compared and air concentrations used for model input will be based upon average values for East St. Louis, Illinois.⁴

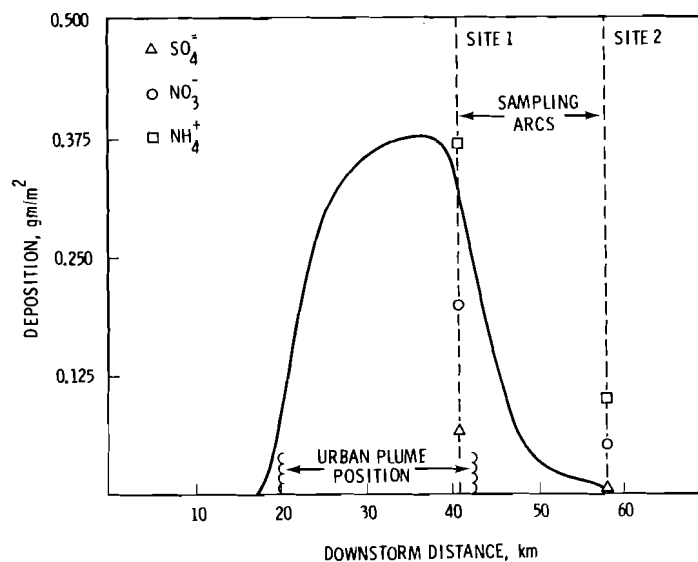
The speed of the storm in question was calculated from radar observations over a 90-min period to be approximately 6 m/sec. The storm speed resulting from model computations also was computed to be approximately 6 m/sec. The intensity of the model storm was, of course, time invariant, since a steady state was assumed. The actual storm, however, fluctuated in intensity as can be ascertained from the rainfall pattern (see Reference 3, Figure 20). Deposition from the actual storm at two downstorm points is listed in Table 2. These "points" are actually the average of values at 7 and 6 cross-storm collection sites along arcs approximately 27 and 45 km, respectively, from the NWS radar at Lambert Field (Reference 3, Figure 20).

TABLE 2. Deposition from August 21, 1972, Storm

Site	Distance From Lambert Field (km)	Flux for Storm (gm/m ²)			Adjusted Due to Observed Air Conc. (gm/m ²)		
		SO ₄ ⁻	NO ₃ ⁻	NH ₄ ⁺	SO ₄ ⁻	NO ₃ ⁻	NH ₄ ⁺
1	27	.43	.16	.023	.068	.20	.37
2	45	.044	.040	.0064	.0070	.050	.10

The model results were obtained for a particular initial pollutant-in-air concentration which averaged 0.17×10^{-6} g/g for the 2.2 km layer near the surface. The cumulative deposition, as it turns out, is directly proportional to this initial air concentration in the model, for a given initial vertical distribution of pollutant. Accordingly, the numbers in Table 2 may be adjusted for comparison with the calculated curve by multiplying the values by the ratio of model initial air concentration to observed air concentration. These adjusted values are also shown in Table 2. The other factor which must be estimated before a comparison can be made is the downstorm location of the two sampling sites in relation

to the urban plume on the day in question. No observations of this plume position are available on this day. So that a comparison might be made, the plume center was assumed to be along the Mississippi River or approximately 17.5 km toward the southeast from Lambert Field. The sampling sites as a result of this assumption were located 9.5 and 27.5 km from the plume center, as shown in Figure 4. This figure compares the deposited amounts at these two distances with the calculated amounts. The agreement is quite encouraging at Site 1 except for the SO₄⁻ which is calculated to be higher than observed by a factor of 4; at Site 2 all observed values are slightly higher



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FIGURE 4. Comparison of Observed Deposition Amounts with Model (Case D) Calculations

than calculated. It is quite possible that these observations at Site 2 represent deposition of a significant amount of "background" pollution rather than urban pollution. Inclusion of "background" pollution in the model input would have produced closer agreement at Site 2. In the author's opinion it is not worthwhile to speculate further upon the reasons for differences between calculated and observed, due to the overwhelming and obvious need for pollutant air concentration measurements prior to thunderstorm passage over the city in connection with such field programs.

CONCLUSIONS AND RECOMMENDATIONS

1. Organized convective storm structure is such that urban pollutant enters such storms with very high efficiency. The model calculations indicate that a significant fraction of this pollutant is scavenged by these storms, given that the pollutant becomes incorporated in cloudwater upon entering the storm. In Case D, approximately 80% of the pollutant entering the storm is deposited.
2. Comparison of observed and calculated pollutant depositions indicates that the model is predicting the amounts and downstorm locations of deposition with very reasonable accuracy, but additional observations are needed.
3. Pollutant deposition in rain is secondarily influenced by the vertical profile of pollutants in the pre-storm environment. The variation in cumulative deposition is on the order of 10-20% of the mean as a result of variation in vertical distribution.
4. There exists an overwhelming need to measure pollutant concentrations in the pre-storm environment in connection with future precipitation scavenging field experiments, such as those discussed in Reference 3. Each individual pollutant measured in rainfall should be measured in the air also. There is also a need for more downstorm precipitation sampling locations, including sites within the city, if possible.
5. After running improved field experiments and model verification and improvement, a much simpler model could and should be formulated to provide such information as 1) the aerial distribution of long term deposition of pollutants surrounding an urban area and 2) maximum short-term doses of pollutant in rainfall. This information would be of great importance to problems in areas of agriculture, water pollution, city planning, and regional and global air pollution.

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THE ENTRAINMENT OF TRACERS NEAR THE SIDES
OF CONVECTIVE CLOUDS

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

During the Metropolitan Meteorological Experiment (METROMEX) inert tracers were released sequentially near the sides of convective clouds at 8,500-16,000-ft altitudes to determine 1) whether air is being entrained into the sides of convective clouds, 2) whether the particulate material in the entrained air is being scavenged, and 3) to provide information on the trajectories taken by the entrained particulates. In 1975 tracers were released near the sides of convective clouds on July 18, 19, 23 and 30. On July 19 Ta, Ir, Re, Au, and Eu were released sequentially into a convective cloud moving southeastward across the precipitation network. The scavenging efficiency for the five tracers varied from 5-95%. A region of heavy tracer deposition was measured 10-20 miles east of each tracer release. The deposition for each successive tracer moved eastward as the location of the tracer release moved eastward. A second region of high tracer deposition was measured 25-35 miles downwind of the tracer releases in a region of increasing precipitation.

PROCEDURE

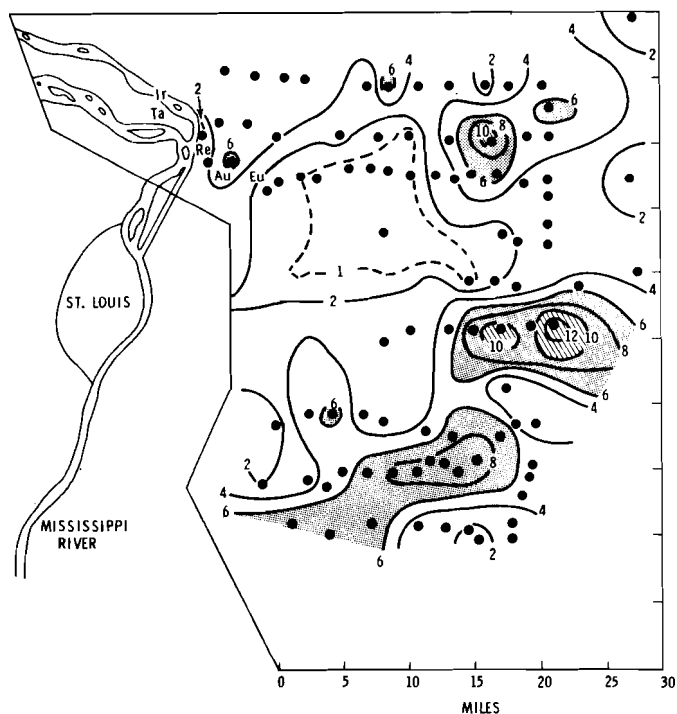
Inert tracers were released sequentially by means of acetone generators at 8,500-16,000 ft near the sides of convective storms in the St. Louis area on July 18, 19, 23, and 30 in 1975. The times and locations of the tracer releases are reported in Table 1. The release of each individual tracer required 2.5 min. Following the tracer releases samples of rain were collected using a 35 x 50-mile network of 219 rain collectors. These samples were frozen using dry ice and returned to the Richland laboratory where they are being analyzed for the tracers and other elements by instrumental neutron activation.

Many of the rain samples from the July 19 storm have been partially analyzed, but analysis has not yet begun on the rain samples collected on July 18, 23, or 30. On July 19 a convective cloud crossed the north-

western boundary of the precipitation network and moved southeastward through the network. Ta was released first at 10,500 ft on the northeastern side of the cell; the Battelle Cessna-411 aircraft then moved to the western side of the cell where it released Ir, Re, Au, and Eu in that order. Atmospherics Inc. released Li into the updraft of this cell at cloud base for the Illinois State Water Survey. The precipitation pattern from this cell, as calculated from the volumes of water in the rain samples that have been boiled down for analysis, is shown in Figure 1. The locations of the samples are indicated by points in the figure. The locations of the tracer releases are also indicated by the chemical symbols of the tracers in the figure. Less than 1 cm of rain fell at most locations. At least some of the water in the samplers near the southern end of the network may have come from a different convective cell.

TABLE 1. Tracer Releases in 1975

Time Start (LDT)	Date	Altitude in K ft	Position (Troy Vortac)		Tracer	Weight Released in g	Direction of Aircraft from Storm
			Start	Finish			
1425	7/18/75	10,5-11	240°/22	240°/20	Tantalum	210	N
1554		11	330°/21	339°/23	Iridium	6.1	NNW
1533	7/19/75	10.5	295°/13	300°/13	Tantalum	420	NE
1543		10.5	300°/14	300°/14	Iridium	5.1	W
1553		8.5	290°/10	290°/9	Rhenium	90	W
1556		8.5	285°/8	285°/7	Gold	142	W
1004		9.5	290°/6	290°/6	Europium	75	W
1410	7/23/75	15-15.5	320°/18	306°/19.3	Tantalum	490	NE
1428		15	300°/33.8	300°/35.9	Iridium	5.5	WNW
1438		16	295°/35	296°/33.8	Rhenium	102	W
1513	7/30/75	11	105°/28.8	107°/22	Tantalum	470	ESE
1526		11	080°/24	080°/22	Gold	135	ESE
1540		11	070°/16	070°/17	Europium	71	ESE
1548		11.5	060°/14	060°/12	Iridium	6.8	E
1559		11.5	060°/14	060°/15	Indium	140	E



Neg 760035-7

FIGURE 1. Precipitation in Millimeters on July 19, 1975

RESULTS OF THE JULY 19 TRACER RELEASE

Tantalum

Only a few of the rain samples contained measurable quantities of Ta. The Ta deposition pattern is shown in Figure 2. Ta concentration was relatively high in one rain sample collected 10 miles downwind of the Ta release and in other samples collected 30 miles downwind of the release. A few other samples contained lesser amounts of Ta. The total Ta deposition in the area from which samples have been analyzed (calculated by dividing the total amount of Ta in the samples analyzed by the fraction of the total surface area covered by these samplers) was only about 5% of the Ta released. Ta was released on the northeastern side of the southeastward-moving cell, while the other tracers were released on the back (western) side of the cell. This may explain why the Ta deposition was less than that of the other tracers.

Iridium

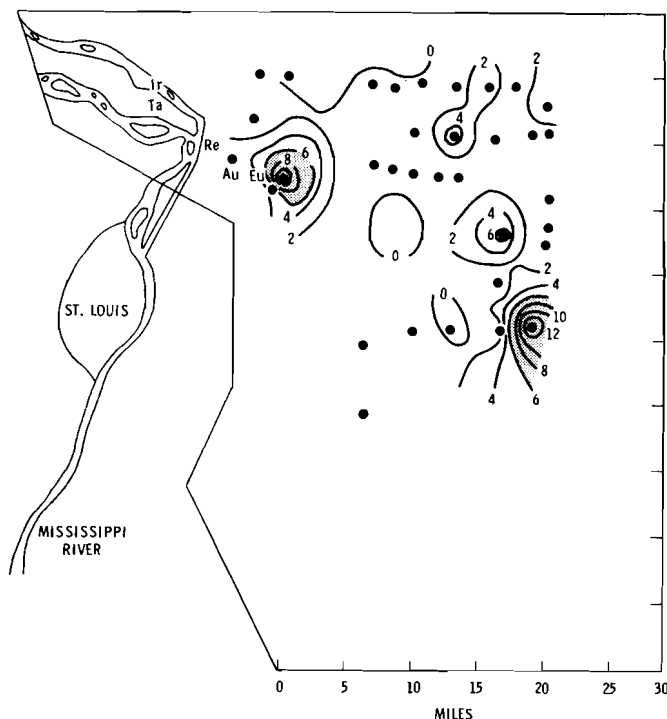
None of the rain samples have yet been analyzed for Ir.

Rhenium

The Re deposition pattern is shown in Figure 3. The highest Re deposition occurred 10 miles east of the Re release, and 5 miles east of the location of the sample containing high Ta. The highest Re deposition occurred in a region of very light rainfall. A region of low Re deposition and light rainfall extended several miles farther downwind; the rainfall rate and the Re deposition increased considerably 30-35 miles southeast of the Re release. The two rain samples containing the most Ta also contained fairly large amounts of Re. The calculated Re deposition in the region of sample analysis was 95% of the amount of rhenium released.

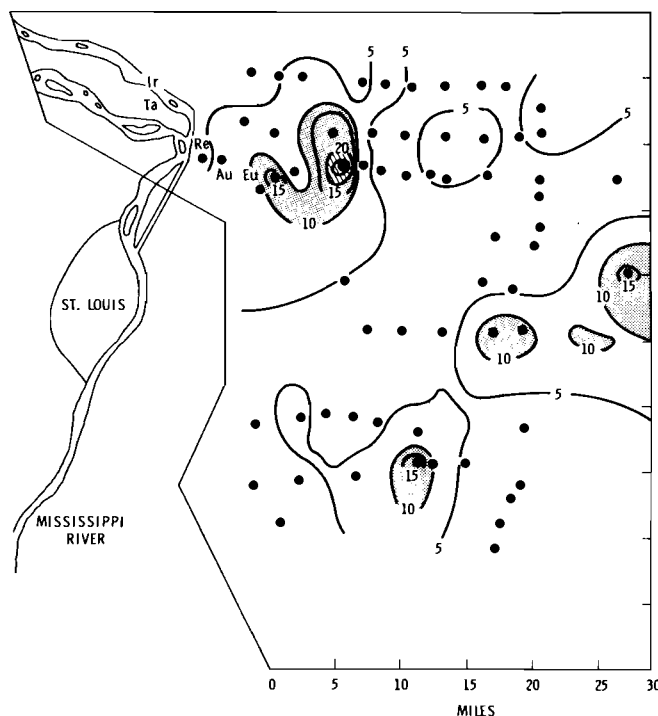
Gold

Au was released immediately after the Rh. CaCl_2 had been added to the Au solution for the purpose of producing mixed hygroscopic particles, so that the scavenging of the hygroscopic particles could be compared with that of the hydrophilic particles of the other tracers. The Au



Neg 760035-4

FIGURE 2. Tantalum Deposition in Picograms Per cm^2 on July 19, 1975



Neg 760035-5

FIGURE 3. Rhenium Deposition in Picograms Per cm^2 on July 19, 1975

deposition pattern is shown in Figure 4. The highest Au deposition occurred 10 miles to the northeast of the highest Re deposition, also in a region of light rainfall. The Au deposition was rather light in the region of heaviest Re deposition. As in the case of the other tracers, the Au deposition increased somewhat 25 miles downwind of the Au release when the precipitation increased. The calculated amount of Au deposited was 15% of Au released. No obvious difference between the Au deposition and the deposition of the other tracers could be attributed to the CaCl_2 added to the Au solution.

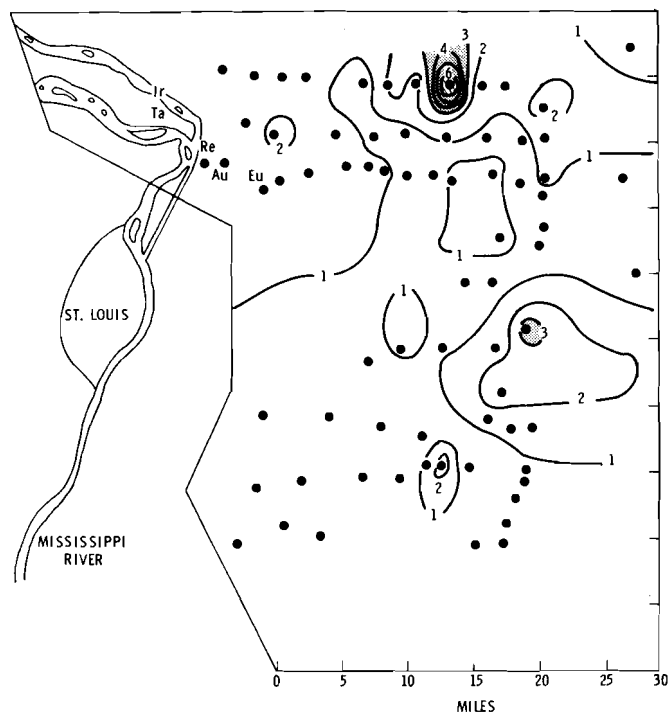
Europium

The Eu deposition pattern is shown in Figure 5. High Eu deposition occurred 19 miles east of the Eu release and 5 miles southeast of the maximum Au deposition, in a region of relatively heavy precipitation. The heaviest Eu deposition occurred 25 miles southeast of the Eu release,

in the same region that the other tracers showed relatively high deposition. The calculated Eu deposition was 30% of the amount released.

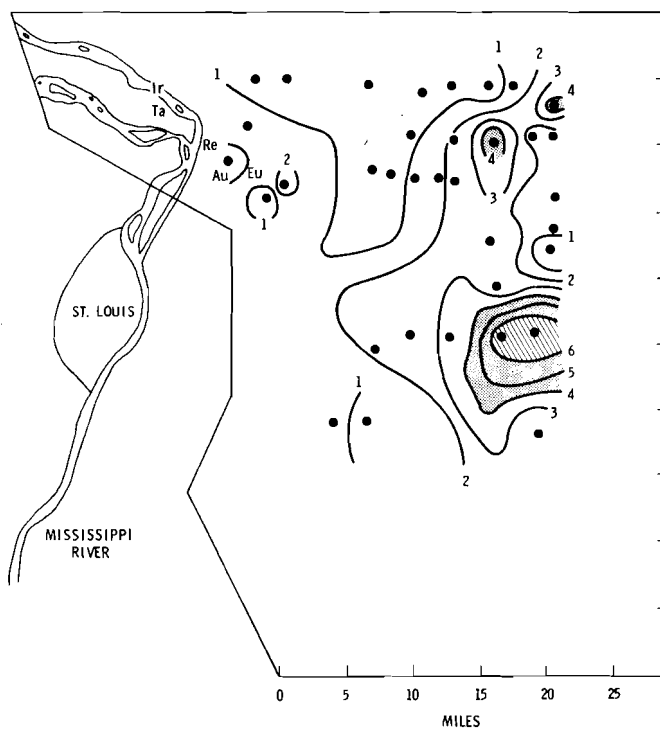
Arsenic

As was not released as a tracer, but was present in measurable quantities in the precipitation. The As in the precipitation probably had been released to the atmosphere primarily by the St. Louis urban-industrial complex. The As deposition pattern is shown in Figure 6. Regions of high As deposition generally coincided with regions of heavier precipitation. The calculated As deposition in the region of sample analysis was 1000 g. An estimate will be made of the fraction of As scavenged by the precipitation when air samples collected in and downwind of St. Louis in August 1975 are analyzed for an estimate of the rate of injection of As into the atmosphere by St. Louis.



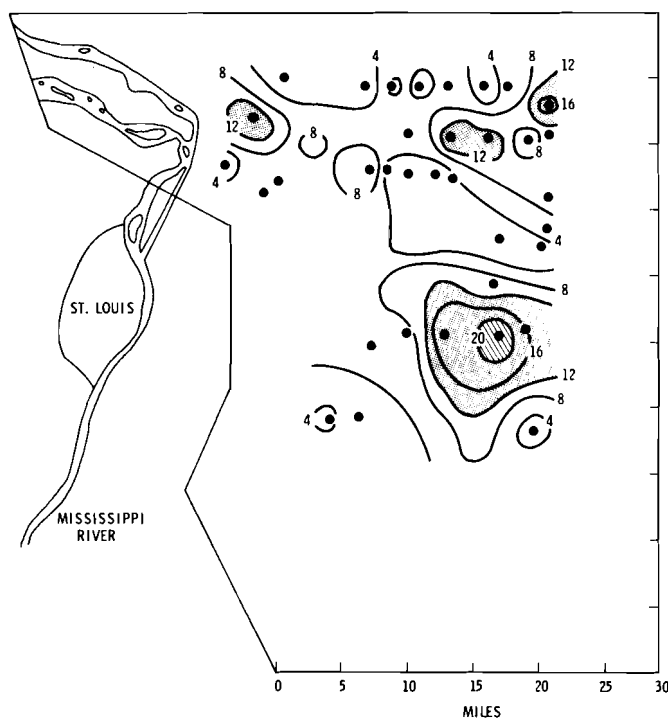
Neg 760035-3

FIGURE 4. Gold Deposition in Picograms Per cm^2 on July 19, 1975



Neg 760035-2

FIGURE 5. Europium Deposition in Picograms Per cm^2 on July 19, 1975



Neg 760035-6

FIGURE 6. Arsenic Deposition in 10^{-11} g Per cm^2 on July 19, 1975

SUMMARY

Each tracer showed a region of relatively high deposition 10-20 miles east of the tracer release. Since the convective cell was moving southeastward, each successive tracer release occurred farther southeastward, and this region of high tracer deposition was displaced a corres-

ponding distance eastward for each successive tracer. High depositions generally were not located in regions of high rainfall. A second region of high tracer deposition occurred several miles further to the southeast for each tracer in roughly the same region as a region of relatively high rainfall.

A DESIGN FOR REGIONAL PRECIPITATION CHEMISTRY
MEASUREMENT AND ANALYSIS

M. Terry Dana and J. M. Hales

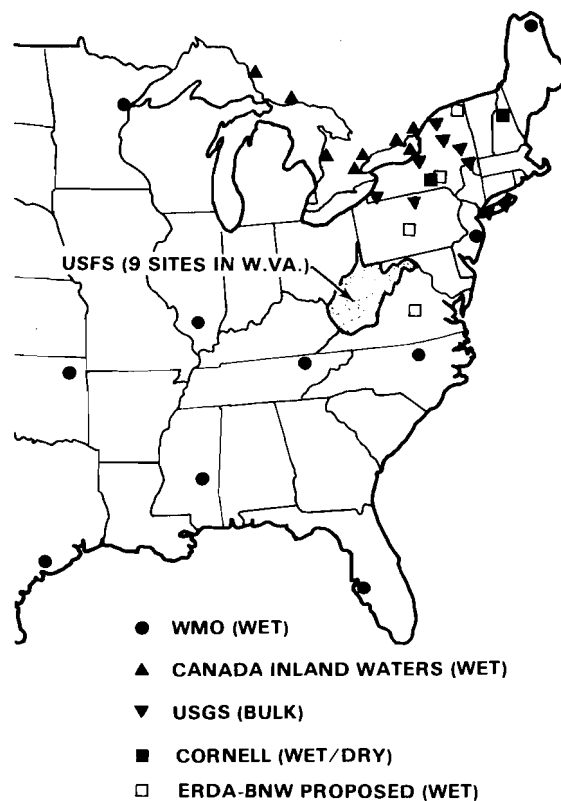
A precipitation chemistry network, utilizing a newly-designed collector, is being set up in the northeastern United States. Siting of three or four initial sites is being coordinated by NOAA, Air Resources Laboratory personnel. Chemical analyses of samples will be done for all relevant fossil-fuel pollutant species, with careful quality control and rapid dissemination of results a major goal.

INTRODUCTION

The use of fossil fuels in energy production has increased steadily; the electrical output has more than doubled in the past 10 years. This increase, and continued increasing use of fossil fuels--particularly coal in the northeastern United States, could significantly affect the environment. Recent research has indicated that a trend toward more acid precipitation in the northeast has paralleled the increased fossil fuel use; in addition, it seems that the original pollutant of most concern, SO_2 , has been supplanted by its oxidation product sulfate. The use of tall stacks to disperse SO_2 and other gaseous pollutants has apparently allowed for the existence of sulfate and other products of reactions in the atmosphere at great distances from the source.

Careful and consistent monitoring of air and precipitation quality is of great importance in assessing the impact of these trends to the affected region. Unfortunately, collection of precipitation for chemical analysis in the northeast has been rather nonuniform in terms of chronological and geographical coverage, sample collection and analysis practices. Figure 1 shows the sites of present collections in the eastern United States.

A research project, Precipitation Scavenging of Fossil-Fuel Pollutants



Neg 71941-1

FIGURE 1. Locations of Existing Precipitation Chemistry Measurements Sites in the Eastern United States and Southeast Canada, Plus Proposed Sites to be Deployed in Mid-1976

on a Multi-State Scale, has been initiated to fulfill the overall objective: the design and institution of a regional precipitation collection network which will avoid past difficulties and provide a basis for a high-quality system of monitoring and data collection for the future. The design of this system places particular emphasis on 1) developing a reliable wet deposition collector; 2) careful siting of collection stations to avoid local source influences and provide adequate coverage of the region; and 3) instituting chemical analysis procedures that provide rapid analysis of all relevant pollutant species and fast dissemination of results to interested scientists and environmental policy makers.

The first year's effort has concentrated on the development of sampler hardware (described in another report in this volume) and the institution of several initial sites in the northeastern United States. The latter effort is being coordinated by personnel of the Air Resources Laboratory of NOAA; Figure 1 shows the initial sites proposed for deployment by mid-1976. The precipitation chemistry of each locale will be examined prior to the location of the exact site, which will then be outfitted with the Battelle sampler. Progress has been made toward location of sites as follows: Ithaca, New York (in cooperation with Cornell University); Whiteface, New York (State University of New York); and State

College, Pennsylvania (Pennsylvania State University).

The chemical analysis will be performed primarily at the Richland laboratories of Battelle. Wet chemical methods will be used for analysis of these species: SO_2 (dissolved), SO_4^{2-} , NO_2^- , NO_3^- , NH_4^+ , H^+ (pH measurement and titration methods), PO_4^{3-} , and conductivity. The eventual list of species to be considered routinely will also include Cl, Na, K, Mg, Ca and carbonates. Of primary importance in this area is the institution of careful and well-defined methods for sample handling and analysis. Full documentation will be provided so that the quality of data will be subject to review at all times. The chemical analysis will be completed as soon as possible after sample collection, and a report of results will be disseminated regularly and promptly through the use of a special data output format.

The northeastern United States network will be expanded in future years in cooperation with other laboratories engaged in precipitation chemistry research. In particular, close coordination with the Illinois State Water Survey, whose plans include a midwestern network, is a goal of the present project. The expanded operating network and developed chemical analysis and quality control methods should form an important component of the Multistate Atmospheric Power Production Pollution Study.

DEVELOPMENT OF A NO-DEPOSITION PRECIPITATION SAMPLER

D. W. Glover

A new type of field sampler is being developed at Battelle-Northwest for precipitation chemistry studies under the MAP3S program. This sampler incorporates sample freezing and automatic lid opening, and will be in operation for test purposes in the near future.

A new type of no-deposition precipitation sampler is currently under development at Battelle-Northwest, and preliminary field testing is expected to take place in December 1975 or January 1976. The testing of components thus far has been satisfactory and no major problems are anticipated with the automated opening portion of the sampler. Figure 1 shows the design of the sampler.

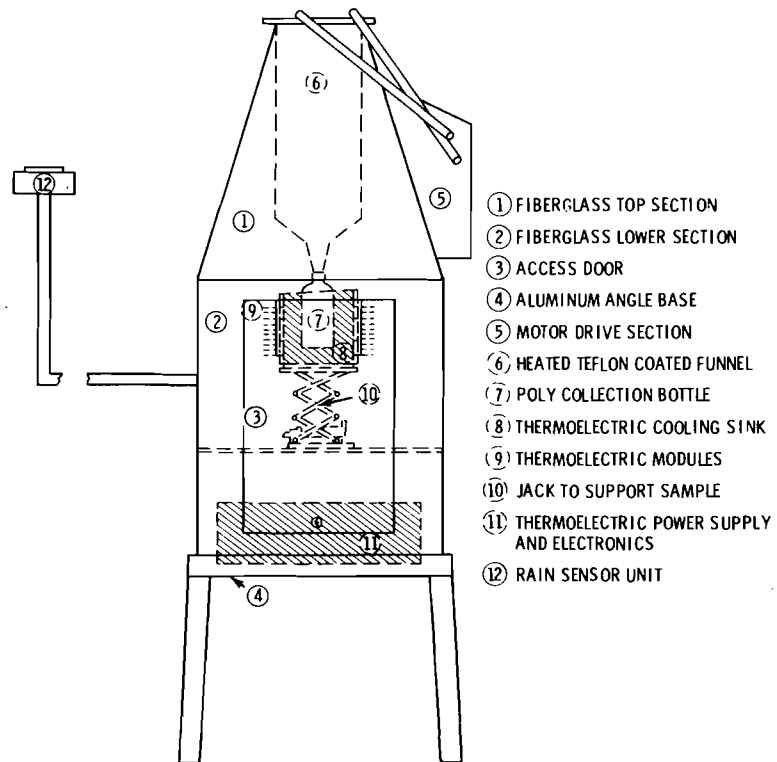
When completed the sampler will utilize a thermoelectric cooling device to freeze the rain sample within a short time after collection. This freezing portion of the sampler, however, has not yet been assembled for testing; some problems in that section are possible.

The principle of automatic lid operation of the sampler has been utilized previously, and several automatic opening samplers are currently being used and tested. However, some problems exist with almost every type that has been used. In addition to incorporating sample freezing with

such a device, we are hoping to use some of the information on the shortcomings of the others to build one as efficient and dependable as possible for implementation in the MAP3S field program.

The principle of operation is the use of a moisture-sensing electronic device to activate a small gear motor which in turn opens the sampler lid. The sensing grid is slightly heated so a quicker closing response time can be achieved. The sensitivity of the unit will be adjustable to respond to only continued precipitation not a brief light shower of only a few drops. A test system is now in operation in the laboratory but a more refined circuit is being built to replace the one now in use.

All components which can come in contact with the sample are being made from or coated with Teflon or polyethylene so as to make the collected sample usage as versatile and free of all contamination as possible.



Neg 72296-1

FIGURE 1. Design of Sampler

POLLUTANT TRANSFORMATIONS AND INTERACTIONS

It is especially important to understand the changing physical and chemical character of pollutants in the atmosphere. The biological and ecological significance can be strongly dependent on the form they assume as they are delivered to receptors. The efficiency of pollutant removal can also be strongly influenced by the pollutants physical and chemical state and the transformations that occur during transport.

The Pacific Northwest Laboratory's investigations of atmospheric pollutant transformations have centered primarily around aircraft observations of pollutant concentrations and conversion rates, which are being interpreted with the use of numerical models of the transformation processes. The primary field experiment during the past year was conducted using the DC-3 aircraft in the St. Louis region during the final term of the METROMEX series. In this experiment, extensive trace gas and aerosol data, and solar radiation measurements were recorded in a Lagrangian reference frame downwind of the metropolitan complex. This experiment was designed as a precursor to the more extensive MAP3S studies and is presently being evaluated using analytical and numerical modeling procedures. The contributions immediately following summarize the progress made during the past year in these areas.

• AEROSOL AND TRACE GAS TRANSFORMATIONS

AEROSOL BALANCE EQUATIONS: THEORETICAL STUDIES

R. L. Drake

Similarity solutions have been obtained for homogeneous and nonhomogeneous aerosol systems. The processes governing these time-evolving systems are coagulation, the condensation of vapors and gases on the particles, volumetric sources and sinks, gravitational setting, vertical velocity fields, and vertical diffusion.

Drake¹ described a similarity solution for an aerosol system evolving in a homogeneous medium. He also listed some applications and advantages of the transformed similarity systems and their resulting solutions. Here we consider both homogeneous and nonhomogeneous media.

The defining equation for the evolution of an aerosol in a nonhomogeneous medium is

$$\frac{\partial n}{\partial t} + \frac{\partial In}{\partial v} + \frac{\partial Wn}{\partial z} - \frac{\partial W_s n}{\partial z} = \frac{\partial \left[D \frac{\partial n}{\partial z} \right]}{\partial z} + \frac{1}{2} N + S, \quad (1)$$

where

$$N(z, v, t) = \int_0^v F(z, u, v-u, t) du - 2 \int_0^\infty F(z, u, v, t) du, \quad (2)$$

and

$$\begin{aligned} F(z, u, v, t) &= \beta(z, u, v, t) n(z, u, t) n(z, v, t) \\ &= \beta(z, v, u, t) n(z, v, t) n(z, u, t) \\ &= F(z, v, u, t) \geq 0. \end{aligned} \quad (3)$$

The independent variables in Eq. (1) to (3) are the particle volume u or v , the time t , and the vertical coordinate z . The only dependent variable in the system is the number density $n=n(z, v, t)$ defined as: ndv is the total number of particles in the interval $v \pm \frac{1}{2}dv$ at time t and location z per unit volume of the medium in which the particles are embedded.

The parameters that govern the system are the coagulation parameter $\beta=\beta(z, u, v, t)$, the condensational growth rate $I=I(z, v, t)$, the volumetric source or sink distribution $S=S(z, v, t)$, the vertical velocity field in the medium $W=W(z, t)$, the particle settling velocity $W_s=W_s(z, v, t)$, and the eddy diffusivity of the medium $D=D(z, t)$. The physical units of these quantities are: $\{n\}=L^{-6}$, $\{I\}=\{\beta\}=L^3T^{-1}$, $\{S\}=L^{-6}T^{-1}$, $\{W\}=\{W_s\}=LT^{-1}$, and $\{D\}=L^2T^{-1}$, where $\{A\}$ is the dimension of A , L the characteristic length and T the characteristic time.

In a series of papers we have studied the system in Eq. (1) to (3). Abstracts of these papers are given in the following paragraphs. The homogeneous system that we have studied can be obtained from Eq. (1) to (3) if the z -dependency is eliminated and if W , W_s , and D are set equal to zero.

(I) QUALITATIVE THEORY FOR HOMOGENEOUS SYSTEMS²

Abstract: This paper is concerned with the similarity transformation of an aerosol balance equation. The system describes the time evolution of an aerosol population in an homogeneous medium under the influences of sources and sinks, coagulation of particles and condensational growth. Without specifically solving for the similarity spectrum, several properties of the spectrum are derived; and several integral power moments are obtained in terms of the similarity spectrum. The relationships between the system parameters and the total volume and total number concentrations of the aerosol population are indicated.

(II) SIMILARITY SOLUTIONS FOR HOMOGENEOUS SYSTEMS³

Abstract: This paper uses direct and indirect methods to obtain several sets of similarity solutions for aerosol systems evolving in a homogeneous medium. The forms of the solutions vary with the variations in the coagulation parameter, the condensational growth mechanism, and the source-sink distribution. For several different combinations of the system parameters, the exponential "ramp" function and the Junge power law were obtained for the similarity spectra. From the results in this paper, several relationships between the system parameters and the similarity spectra were also obtained. For example, the heavier-tailed spectra correspond to the unbounded coagulation parameters and the lighter-tailed spectra correspond to the bounded kernels.

(III) SIMILARITY SOLUTIONS FOR NONHOMOGENEOUS SYSTEMS⁴

Abstract: This paper is concerned with an aerosol population evolving in a nonhomogeneous medium under the influences of a condensational growth mechanism, coagulation, a source and sink distribution, a vertical wind field, gravitational settling, and vertical diffusion. Transformation equations are derived that map the physical space (z, v, t) into a one-dimensional similarity space, defined by the θ -coordinate. The governing equation for the similarity spectrum $\psi(\theta)$ is a second order, nonlinear, ordinary, integrodifferential equation. Indirect methods are derived for the solution of this system. Using one of the indirect methods, a detailed example is worked out resulting in the similarity solution of a system representing an evolving aerosol under realistic type conditions.

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AEROSOL BALANCE EQUATIONS: NUMERICAL MODELING

R. L. Drake

This report describes the work being done on the aerosol component of a user-oriented computer code being formulated to solve the general equations of continuity for trace substances in the atmosphere. This submodel will be used extensively in the Aerosol and Trace Gases Program.

Hales¹ described a user-oriented computer code for solving the general equations of continuity for trace substances in the atmosphere. This code will eventually include components for the transport and fate of all gaseous and particulate pollutants important to the Aerosol and Trace Gases Program. Formulation of these components will include terms to account for the interaction of gases, different species of particles, and gas-to-particle conversions.

This report concerns the aerosol component of the general code. We describe the general equation for an aerosol interacting with gases and other species of aerosols; outline the current status of the submodel development; predict the future work required to complete this submodel and incorporate it into the general user-oriented computer code.¹

The governing equation for the number density of the i^{th} aerosol specie n_i is given by

The independent variables in Eq. (1) are the usual cartesian space coordinates x_m ($m=1,2,3$ where x_3 is the vertical coordinate positive upward), the particle volume or mass v , and time t . The dependent variable is the average number density of the i^{th} aerosol species $n_i = n_i(x_m, v, t)$. In Eq. (1), we assume that there are several interacting aerosol species, namely, $n_1, n_2, \dots, n_I$. The prime quantities n_i' are the fluctuating components of the number densities due to fluid turbulence. In addition, the bracket $\langle \cdot \rangle$ represents some type of statistical average.

The individual terms and the system parameters in Eq. (1) are defined as:

1. The second term on the left-hand side of Eq. (1) is the advection term and V_m is the mean velocity field in the fluid;
2. V_{si} is the gravitational settling velocity of the i^{th} aerosol;

$$\frac{\partial n_i}{\partial t} + \sum_{m=1}^3 \frac{\partial V_m n_i}{\partial x_m} - \frac{\partial V_{si} n_i}{\partial x_3} + \frac{\partial I_i n_i}{\partial v} = C(n_i) + B(n_i) + S_o(n_i) - S_1(n_i)$$

$$+ \sum_{\substack{j=1 \\ i \neq j}}^I R_{ij}(n_i, n_j) + \sum_{m=1}^3 \sum_{j=1}^3 \left[\frac{\partial}{\partial x_m} + A_{im} \frac{\partial}{\partial v} \right] K_{jm} \left[\frac{\partial}{\partial x_j} + A_{ij} \frac{\partial}{\partial v} \right] n_i$$

$$+ \langle C(n_i') \rangle + \langle B(n_i') \rangle + \langle S_o(n_i') \rangle - \langle S_1(n_i') \rangle + \sum_{\substack{j=1 \\ i \neq j}}^I \langle R_{ij}(n_i', n_j') \rangle. \quad (1)$$

3. I_i is the mean condensational growth mechanism;
4. $C(n_i)$ represents the mean particle coagulation mechanism;
5. $B(n_i)$ is the mean breakup mechanism;
6. $S_0(n_i)$ is the mean primary source of particles and $S_1(n_i)$ is the mean volumetric sink for particles;
7. $R_{ij}(n_i, n_j)$ is the mean transfer of particles from the i th species to the j th species, or vice-versa;
8. K_{jm} are the components of the eddy diffusivity tensor describing the mean effects of turbulent diffusion on the particles in the fluid; in general, the K_{jm} depend upon the volume of the particles;
9. The term A_{im} represents the partial derivatives $\partial v / \partial x_m$ for the i th aerosol distributed throughout space for a fixed time t . In cloud physics, A_{im} is a function of several parameters, such as the densities of air and water, the specific heats of air and water, the heat of condensation, and the temperature and moisture gradients;
10. The last five terms on the right-hand side of Eq. (1) represent the nonlinear, fluctuating terms in the system.

We now briefly outline the current status and future direction of our research to simplify and implement the system given in Eq. (1).

The formulations of the mean breakup term $B(n_i)$, the mean coagulation term $C(n_i)$, and the mean aerosol transfer or interaction terms R_{ij} will be patterned after the results given in Melzak.² The corresponding formulas for the fluctuating quantities, $B(n_i')$, $C(n_i')$ and $R_{ij}(n_i', n_j')$, are considered by Scott,³ Marcus,⁴ and Gillispie.⁵ It appears that the fluctuating terms can be neglected if the spectrum of n_i versus v evolves as a Poisson distribution. However, if the distribution is non-Poissonian, the $\langle C(n_i') \rangle$ appears to

be a positive contribution to the right-hand side of Eq. (1); similar statements can be made about the breakup and transformation terms. Hence, if n_i evolves in a non-Poissonian manner (which is usually the case), then the coagulation rate is faster when $\langle C(n_i') \rangle \neq 0$ than when $\langle C(n_i') \rangle = 0$. However, because of the many uncertainties in the parameters in the terms $C(n_i)$, $B(n_i)$ and R_{ij} , we will neglect the contributions from fluctuating quantities for now.

The condensational growth term I_i represents the growth of particles in the i th aerosol due to condensing vapors and gases on the particles. I_i can also be formulated so that it represents a secondary source of particles due to gas-to-particle conversions. The formulation of this term will be based on the studies of Brock^{6,7,8,9} and Heisler and Friedlander.¹⁰ The fluctuating terms involving the condensational growth are accounted for by the K_{jm} and A_{im} terms. In the field of cloud physics, Levin and Sedunov¹¹ have discussed the formulation of A_{im} . For the general field of aerosol physics, we will have to obtain a similar expression for A_{im} , which will probably vary with each species.

The ambient velocity field V_m will be postulated or obtained from field data. If the overall computer code is eventually linked to a dynamic flow model, then V_m would be obtained from the flow calculations. The gravitational settling velocity V_{si} for aerosol particles depends upon particle shape, size and density, as well as the properties of the fluid. The values for V_{si} will be obtained from the theoretical and empirical data that occur in many current reference books and papers in the field of aerosol physics.

The forms of the volumetric sources and sinks will depend upon the specific problems and areas being studied. We will also gain some information and insight from the formulation of heterogeneous and homogeneous nucleation processes by Brock,⁹ and Slinn's^{12,13} discussions concerning the sink mechanisms in the atmosphere. In general, we are rather ignorant of the details of $S_0(n_i)$ and $S_1(n_i)$; thus, at this time, the fluctuating

components of the sources and sinks will be neglected.

The determination of the components K_{jm} of the eddy diffusivity tensor will be based on the following theoretical and empirical results. Calder¹⁴ and Brutsaert¹⁵ discuss the diffusivity tensor and the relationships between the various components K_{jm} . Brutsaert finds that the non-zero components are K_{11} , $K_{13}=K_{31}$, K_{22} and K_{33} when the mean wind is along the x_1 -axis. In addition, Brutsaert relates K_{11} , K_{13} and K_{22} to K_{33} , and then relates K_{33} to the vertical diffusivity for momentum K_z through a Reynold's analogy argument. Other relationships between K_{33} (for heat transfer) and K_z are given in Smagorinsky¹⁶ and Drake¹⁷ et al., O'Brien¹⁸ and Agee¹⁹ et al. give models for K_z in the planetary boundary layer under various temperature and wind regimes.

Eq. (1) describes a system that depends on five independent variables, x_1 , x_2 , x_3 , v , t . To solve this system numerically requires a very large memory and a great investment in computation time for any reasonable real-world problem. In order to reduce these memory and time requirements and still retain realistic physics and chemistry, we propose to eliminate the v - and x_1 -dependency in Eq. (1). In eliminating the x_1 -dependency, we assume that the mean wind V_m is given by

$$V_2 = V_3 = 0, \quad V_1 = U. \quad (2)$$

The rationale for eliminating the v -dependency is based upon the "three-mode" aerosol distribution proposed by Whitby²⁰ and supported by the data collected by PNL in St. Louis.²¹ Whitby considers three distinct aerosol ranges in the normal atmospheric aerosol; they are the transient nuclei or Aitken nuclei range $d < 0.1 \mu m$, the accumulation range $0.1 < d < 1 \mu m$, and the mechanically-generated aerosol range $1 < d < 100 \mu m$, where d is the diameter of a particle. Each range has its own main sources of mass, its own principal processes, and principal removal mechanisms. In addition, there are mechanisms, such as coagulation, that transfer some particles from lower to higher ranges. Breakup mechanisms may transfer particles

from higher ranges to lower ones. In our work we will write an equation of type (1) for each of the ranges. Each equation will reflect the processes, sources and sinks representative of its particular range. The transfer terms R_{ij} will represent the mechanisms for transferring the particles from one range to another range. However, these three equations are still dependent upon v .

To eliminate v in the equations, we will postulate the variation of n with respect to v , namely

$$n(x_m, v, t) = \phi(v; \alpha, \beta, \gamma), \quad (3)$$

where ϕ is a known function of v and the parameters α, β, γ . The parameters α, β, γ are unknown functions of x_m and t ; but they can be related to the integral power moments of n ,

$$M_\ell(x_m, t) = \int_0^\infty v^\ell n(x_m, v, t) dv, \quad (4)$$

where $\ell=0,1,2$.

If we replace the n 's in our three equations for the various Whitby ranges (each n will have a different set of parameters) and then apply the integral in (4) for $\ell=0,1,2$, we will obtain the defining equations for the integral power moments in the three size ranges. The results of these operations will be the elimination of the independent variable v and the simplification of the coagulation, breakup and transfer terms.

Knowing the integral power moments, $M_\ell(x_m, t)$, $\ell=0,1,2$, we can compute α, β, γ and thus define the number density for each of the size ranges from Eq. (3)

There are three candidates for the function ϕ that are mathematically and physically appealing (see Mielke and Johnson²²). They are: the gamma distribution which has a "light tail," the lognormal distribution which has an "intermediate tail," and the "heavy-tailed" beta-kappa distribution.

The x_1 -dependency will be eliminated by replacing the (x_1, x_2, x_3, t) fixed coordinate system by a moving coordinate system (x_2, x_3, t) . The

moving system will travel along the x_1 -axis at a rate equal to U . Strictly speaking, U should be a constant in time, but we will allow it to be a slowly varying function of time. We will also allow the x_1 -path to slowly vary direction as the vertical slab (x_2, x_3, t) moves along.

However, the orientation of the vertical slab (x_2, x_3, t) must always be normal to the x_1 -direction. Hence, the aerosol submodel, as well as the complete trace substance model, will be a two-dimensional, time-varying system that is advected along in the direction of the mean wind U .

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THE GROWTH OF AEROSOL IN AN URBAN PLUME

A. J. Alkezweeny

Time changes of aerosol particle size distribution in the range of 0.01-5.0 μm diameter, concentration of O_3 , NO , NO_2 , SO_2 , several hydrocarbons, and sulfate were measured in an urban plume. The investigation was conducted in a Lagrangian frame of reference using instrumented aircraft. The air parcel trajectory was identified by the movement of a tetroon launched from the ground to an altitude within the plume. This study was carried out in metropolitan St. Louis, Missouri, U.S.A., during the METROMEX program.

A pronounced change in the aerosol particle size distribution and an increase in the total volume of the aerosol were observed. Gas-to-particle transformations involving existing nuclei cause the aerosol growth. In this paper the results of the measurement of trace gases and the aerosol particles and their chemical analyses will be presented and discussed.*

INTRODUCTION

The objective of the experiment conducted in St. Louis during the summer of 1975, was to determine physical and chemical changes of pollutant in the city plume. Emphasis was placed on particle growth by coagulation and/or gas-to-particle transformation. The investigation was carried out in a Lagrangian frame of reference using an instrumented DC-3 aircraft. The air parcel trajectory was identified by the movement of a 0.94 m^3 tetroon launched from the ground to an altitude within the plume. Portions of the data will be presented here; details of the results will be reported later.

EQUIPMENT

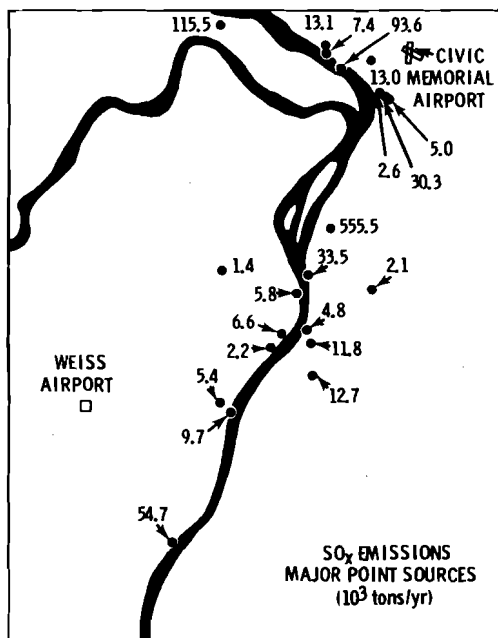
Size distribution of particles with diameters in the range of 0.01-1.0 μm was measured with an airborne Electrical Aerosol Size Analyzer,¹

* This Abstract has been accepted for presentation in the 12th International Symposium on Atmospheric Pollution, 5-7 May 1976, Paris, France.

and those from 0.3 μm to about 5 μm were obtained with a Royco Optical Sensor Model 220 interfaced with a 15-channel pulse height analyzer and printer. A General Electric Particle Counter was used to measure concentrations of Aitken nuclei. Particles were also collected on IPC filters at the rate of 50 cfm for later chemical analysis. A flame photometric detector was used to determine SO_2 concentration. NO , NO_2 and O_3 concentrations were measured with a TECO Model 14D, NO_x Analyzer and a Bendix Ozone Monitor. Dew point, and aircraft parameters were also recorded. Most of the parameters were sampled 5 times per second, and averaged over the second via a Nova computer and stored on a 7-track magnetic recorder.

SAMPLING

The tetroon was launched either from Civic Memorial Airport or Weiss Airport, located at the edge of the city (see Figure 1), depending on the wind direction. The location was chosen to prevent the tetroon from going over the city and interfering with commercial aircraft flights. The



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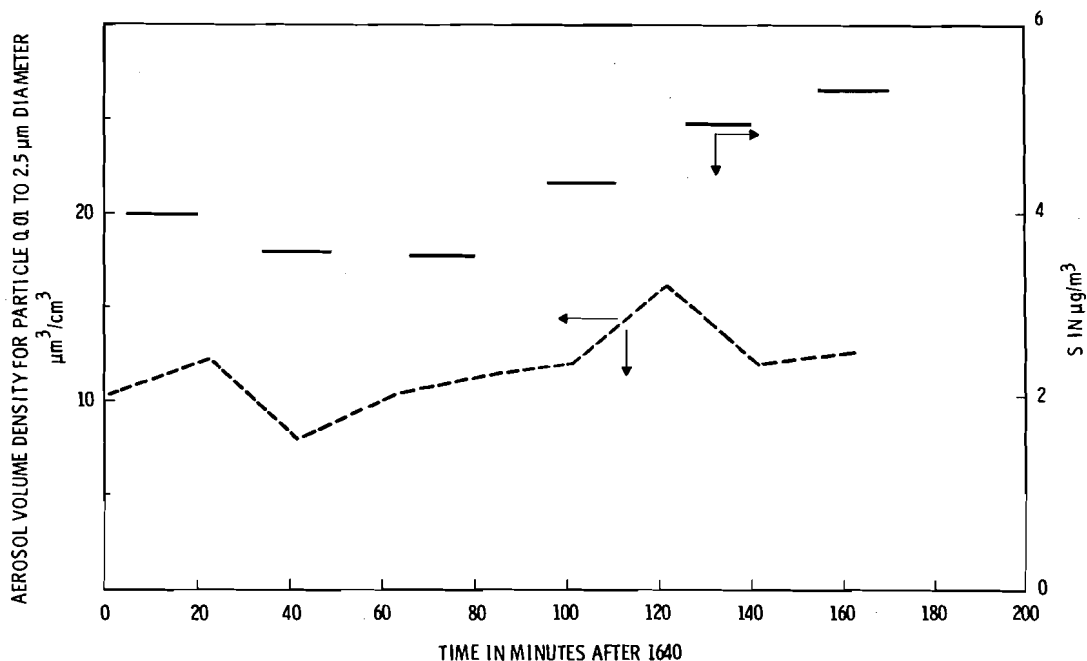
FIGURE 1. The Locations of the Tetroon Launch Sites and the Major SO_x Point Sources Emissions in the Area

procedure for deploying the tetroon is that described by Hoecker.² Before and during each flight, pilot data were collected to determine wind speed and direction. The tetroon was launched 1 hr after the aircraft took off to allow instrumentation calibration and stabilization.

On August 6, a tetroon was launched at 1615 local time from Weiss Airport to an altitude of 3000 ft MSL. The day was characterized by low overcast stratus and stratocumulus in the morning changing to broken cumulus by noon, then broken variable scattered cumulus and swelling cumulus in the afternoon; clear at sunset. The wind was from the northeast at about 17 knots. The temperature inversion at 1647 hr was located at about 8000 ft MSL. Although SO₂ data was not collected during this flight, values of 48-93 ppb were measured at 3000 ft on August 4 about 10 miles upwind of the launch site, under similar wind conditions.

RESULTS

Figure 2 shows the time changes in the particle volume concentration



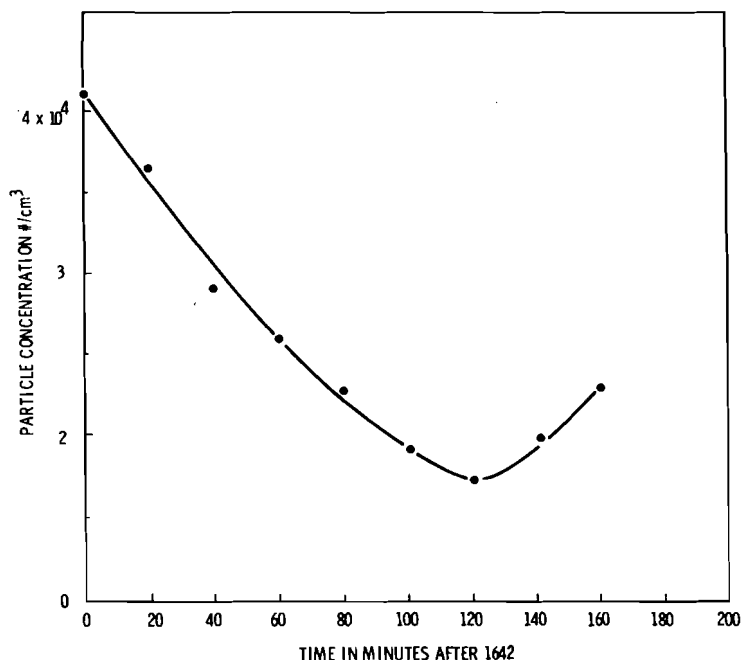
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FIGURE 2. The Variation in the Aerosol Volume Concentration and the Sulfur Content of Aerosol Measured on August 6, 1975

measured during the flight; superimposed on it is the sulfur content of the aerosol. The volume concentrations were calculated from the particle size distribution and the sulfur was obtained from the analysis of the high-volume filter by x-ray fluorescence. The volume concentration is averaged over 15 min of sampling time. The figure shows three distinct features of the aerosol mass concentration profile. The first 60 min the sulfur and particle volume concentrations decreased with time reflecting the domination of plume dilution by diffusion over particle growth. During the second 60 min, plume diffusion was less significant and the particle volume concentrations decreased. At the end of this period a new material was entrained onto the air parcel resulting in a decreased concentration. At this point the pilot reported smoke from below having reached the sampling level. Apparently this new material has a lower mass concentration but the total number of particles is higher. The latter conclusion is based on examination of the variation

of the total particle number concentration with time (Figure 3). As expected the first 2 hr were characterized by a continuous decrease in the particle count as a result of particle growth and plume diffusion. Beyond this point the count started to increase. Although the number concentration was calculated from the measured particle size distribution, a similar profile was also observed from the G.E. Aitken Nuclei Counter (Figure 4).

Figure 4 shows the variation of other parameters measured during the flight. The relative humidity value ranged 50-60% and the temperature fluctuated around 20°C. The change in altitude with time is the sampling altitude but it was very close to the tetroon level. The ozone level slightly increased with time. On the other hand, the NO/NO₂ ratio decreased and showed no correlation with the ozone. This behavior is typical for the flights we made. The NO and NO₂ levels remained always below about



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FIGURE 3. Concentration of Particles Larger than 0.01 μm in Diameter as a Function of Time Measured on August 6, 1975

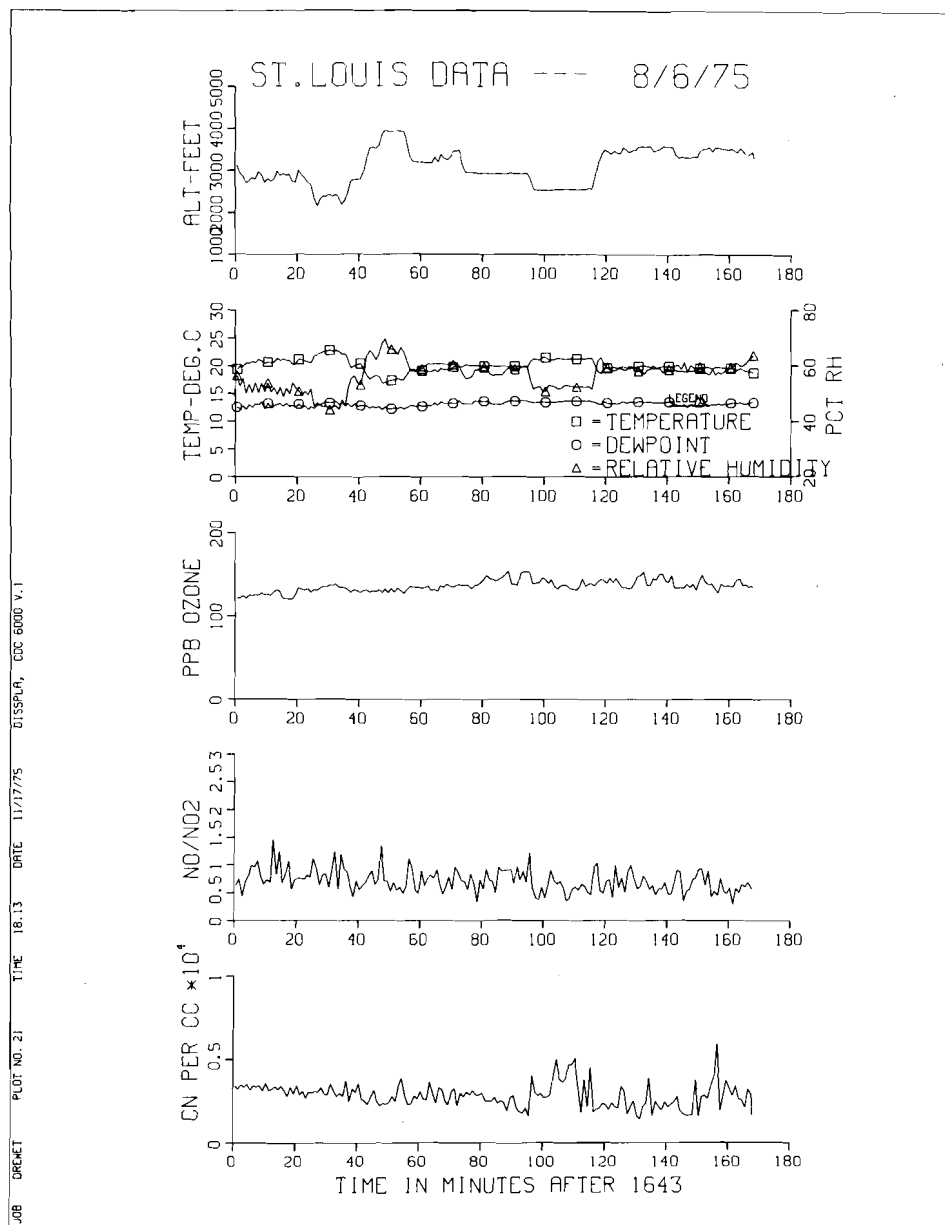
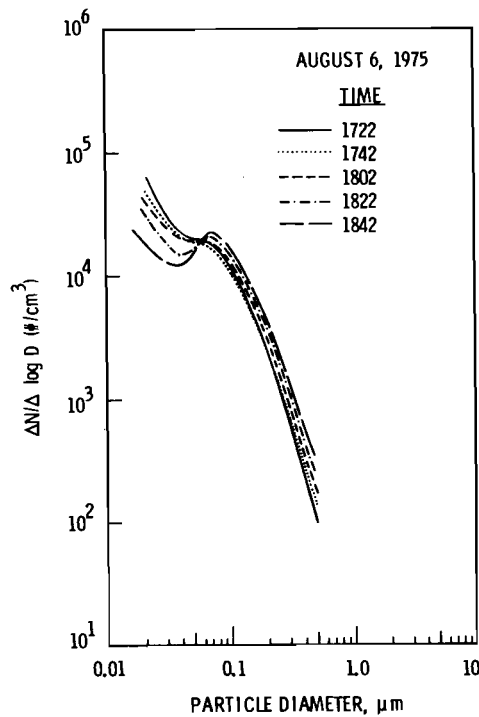


FIGURE 4. Some of the Parameters Measured During the August 6, 1975, Flight and the O_3 , NO/NO_2 Concentration

20 ppb except when a plume was encountered along the tetron trajectory. In this case a drastic drop in the O_3 level was observed associated with an increased NO/NO_2 ratio.

Figure 5 shows the changes in the particle number distribution, and the same data is plotted in Figure 6 in terms of volume distribution. Parti-

cles of sizes below about $0.06 \mu m$ were growing to sizes up to $0.9 \mu m$. Since the total number of particles was decreasing while the total volume was increasing, the particles were growing by coagulation and chemical condensation. No evidence of homogeneous nucleation of particles can be seen from this data. Similar behavior was also observed by Husar and Whitby³ when a

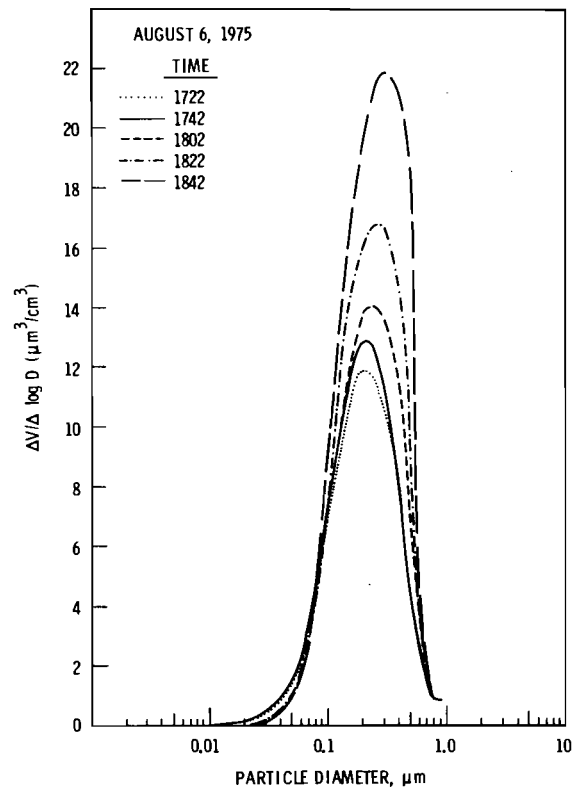


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FIGURE 5. Number Distribution of Particles Measured on August 6, 1975

large balloon was inflated with Los Angeles ambient air and exposed to solar radiation.

It is useful to compare this size distribution measurement with those measured a year earlier on August 21, 1974. For this experiment, the tetroom was released to an altitude of 3000 ft MSL, from the Civic Memorial Airport, about noon under clear sky with an average wind speed of about 8 knots. Figure 7 shows the size distributions measured. The particle growth is similar. The two sets of particle size distributions showed the bimodal nature of the distribution. The first mode roughly lies between 0.06-0.9 μm , the so-called accumulation mode.⁴ Particles in this range grow by coagulation and/or condensation. The second mode is above about 1 μm . Particles of such sizes showed no detect-



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FIGURE 6. The Same as Figure 5, Except the Data Plotted as a Value Distribution

able growth, and as can be seen from Figure 7, their number decreased with time as a result of plume diffusion. Plume diffusion does seem to dominate the growth of smaller particles as the case of August 6. However, the mass concentration measured on August 21 is much higher than that measured on August 6.

The experiment described above is part of the aerosol and trace gases transformation program aimed at understanding pollutant transformation through field experiment and modeling, with a strong interaction between the experiment and the models. The models are described elsewhere by Hales⁵ and Drake.⁶

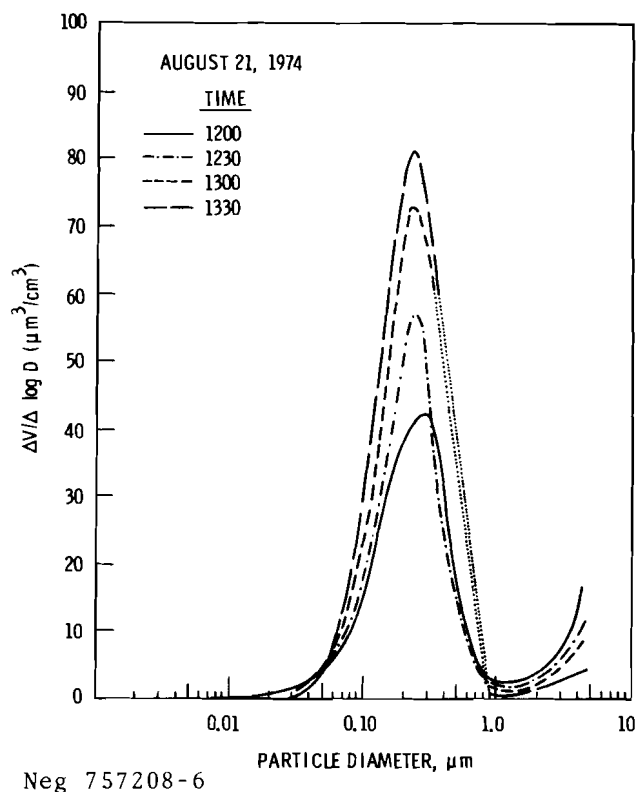


FIGURE 7. Volume Distributions of Particles Measured on August 21, 1974

ACKNOWLEDGEMENTS

The author wishes to acknowledge the contributions of K. M. Busness, R. N. Lee, N. S. Laulainen, and J. W. Thorp who participated in the field operation and data reduction;

to D. R. Drewes who developed a computer program for data analysis; to J. M. Hales, C. E. Elderkin, and R. L. Drake for their valuable discussion and remarks throughout the year; and to F. O. Gladfelder and W. R. Griffith, the pilots.

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SIMULTANEOUS AEROSOL SIZE DISTRIBUTIONS AND TURBIDITY
MEASUREMENTS OVER A METROPOLITAN AREA*

A. J. Alkezweeny and N. S. Laulainen

The particle size distribution measured in the St. Louis city plume is shown to be bimodal. Total particle volume decreases dramatically above the inversion. The peak in the volume distribution is shifted toward larger particles at higher altitudes. Aerosol extinction coefficients derived from sunphotometer optical depth measurements at 5 wavelengths are compared to those calculated from the measured size distributions, using Mie theory with two different particle refractive indices. For an entirely real refractive index of 1.5, the measured values are consistently larger by factors of 2 to 4 than the calculated values. Including a large imaginary part to the refractive index, $n = 1.5 - 0.3i$, improves the agreement, suggesting that the aerosol in the city plume may have significant absorptive characteristics.

PROCEDURE

A DC-3 instrumented aircraft was used to simultaneously measure aerosol particle size distributions and turbidities at different altitudes over metropolitan St. Louis, Missouri.

An Electrical Aerosol Analyzer¹ and a Royco Optical Sensor model 220 were used to determine the particle size distribution in the range of 0.01 μm to 5.0 μm . The sensor interfaced with a 15 channel pulse height analyzer and a printer. A multiwavelength sunphotometer measured solar intensities in 4 narrow wavelength bands (~ 10 nm, centered at 448.2, 500.7, 557.8 and 618.9 nm respectively), and in 2 broad wavelength bands (~ 60 nm, centered at 500 and 598.4 nm respectively). Using calibrated instrumented response of

* Abstract of an expanded version of this paper has been accepted for presentation in the 12th International Symposium on Atmospheric Pollution, 5-7 May 1976, Paris, France.

the photometer, turbidities were determined for each of the wavelength bands.

The flight was made on August 8, 1975, under clear and hazy sky. Wind was from the southeast at less than 10 knots. The measurements were made 12 miles northwest of Civic Memorial Airport in Alton, Illinois. The vertical temperature profile shows an inversion between 4000-5000 ft MSL (Figure 1). The particle size distributions measured during this flight are shown in Figure 2.

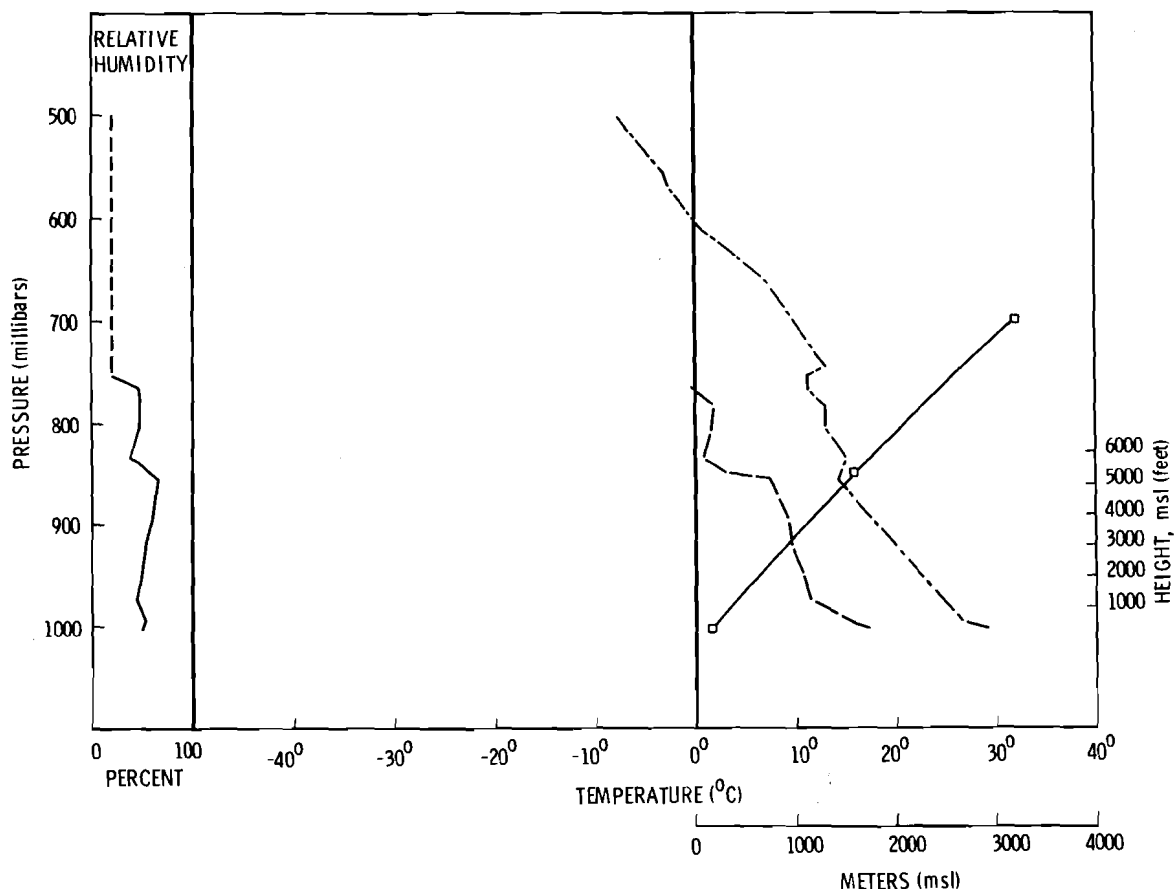
RESULTS

Clearly the distributions are bimodal. The total volume decreases dramatically above the temperature inversion. The peak in the volume distribution is shifted to larger parti-

cle sizes with increasing altitude. The figure also shows that the volume concentration just below the inversion is higher than at 2000 ft MSL, indicating that the material is accumulating beneath the inversion. This is also reflected in SO_2 measurements, which show similar behavior. The material above the inversion probably represents a well-aged pollutant trapped above the inversion. The SO_2 level was 7 ppb which is an order of magnitude lower than below the inversion.

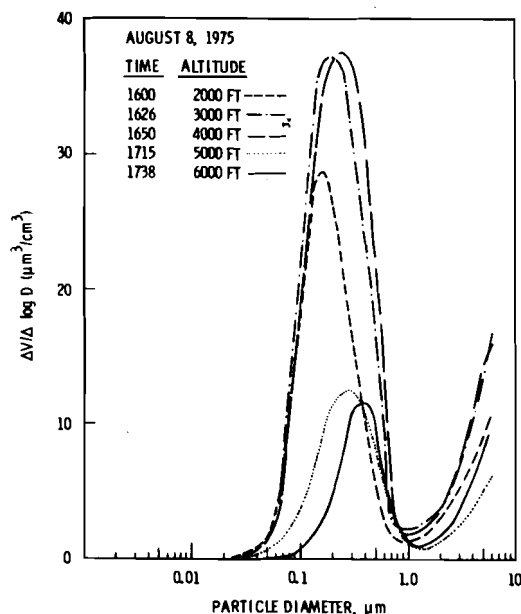
Aerosol extinction coefficients derived from the sunphotometer turbidity measurements are given in Table 1, Column A for each wavelength band. The coefficients were determined using the equation

$$b = \Delta\tau/\Delta h, \quad (1)$$



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FIGURE 1. RAPS/Alton No. 144, August 8, 1975, 1646 CDT



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FIGURE 2. Particle Size Distributions Measured During Flight

where b is the average extinction coefficient for the layer between h and $h + \Delta h$, $\Delta \tau$ is the difference between the turbidities (aerosol optical depths) measured at h and $h + \Delta h$, and h is the altitude. Since τ is unitless, b has units of $(\text{length})^{-1}$.

For comparison, aerosol extinction coefficients were calculated with Mie theory from the measured size dis-

tributions at each altitude with two different particle refractive indices using the form

$$b = \sum_i Q(\alpha_i, n) \Delta S(D_{pi}) \cdot \frac{10^{-6}}{4}, \quad (2)$$

where $Q(\alpha_i, n)$ is the unitless Mie extinction efficiency for size parameter $\alpha_i = \pi D_{pi} / \lambda$ and refractive index n , and $\Delta S(D_{pi})$ is the measured particle surface area ($\mu\text{m}^2/\text{cm}^3$) in the geometric mean particle diameter size class D_{pi} . The summation is over all size intervals from about $0.04 \mu\text{m}$ to $5 \mu\text{m}$. Calculated extinction coefficients for $n = 1.5$ are given in Table 1, Column B, while coefficients for $n = 1.5 - 0.3i$ are listed in Table 1, Column C.

For $n = 1.5$, the measured values are consistently larger, by factors ranging from 2-4, than the calculated values. Including a large imaginary term to the refractive index, $n = 1.5 - 0.3i$, improves the agreement; appreciable discrepancies still exist however. The comparison does suggest that the aerosol in the St. Louis city plume may have significant absorptive characteristics, a result found by others.² Spatial inhomogeneities in the city plume may be responsible for some of the observed discrepancies between measured and calculated extinction coefficients.

The inversion layer also has a dramatic effect on the transmission of solar radiation. Table 2 shows the solar radiation transmission

TABLE 1. Comparison of Aerosol Extinction Coefficients Derived from Sunphotometer Optical Depth Measurements at 5 Wavelengths and Extinction Coefficients Calculated from Measured Size Distributions Using 2 Different Particle Refractive Indices

Altitude (ft)	Extinction Coefficient, b , 10^{-4} m^{-1}														
	448.2 nm			500.0 nm			557.8 nm			598.4 nm			618.9 nm		
	A	B	C	A	B	C	A	B	C	A	B	C	A	B	C
2500	2.0±.7	1.20	2.30	0.9±.5	0.96	2.03	0.4±.4	0.72	1.85	0.7±.7	0.65	1.72	2.3±4.4	0.61	1.64
3500	5.1±.6	1.62	2.83	6.0±.5	1.30	2.52	4.6±.5	0.98	2.30	3.1±.6	0.90	2.15	3.1±1.0	0.85	2.05
4500	3.3±.5	1.22	1.94	1.1±.4	1.00	1.74	1.4±.4	0.75	1.60	2.2±.6	0.69	1.50	2.1±1.0	0.65	1.43
5500	1.9±.5	0.59	0.74	1.3±.3	0.50	0.68	1.3±.4	0.38	0.63	0.1±.4	0.36	0.60	.7±1.0	0.34	0.58

TABLE 2. Solar Radiation Transmission Losses Due to Aerosol Through a Turbid Atmosphere Referenced to 6000 ft and Descending to 2000 ft

Altitude (ft)	Transmission Loss Due to Aerosol				
	448.2 nm	500.0 nm	557.8 nm	598.4 nm	618.9 nm
5000	6%	4%	4%	0.3%	2%
4000	15%	7%	8%	7%	8%
3000	27%	23%	20%	15%	16%
2000	31%	25%	21%	17%	22%

losses due to aerosol through a turbid atmosphere as one descends from above the inversion (6000 ft MSL reference level) down to well below the

inversion. As expected the predominance of submicron particles has the greatest effect for the shorter wavelengths.

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TURBIDITY VARIATIONS OVER THE ALTON, ILLINOIS,
AIRPORT DURING AUGUST 1975

N. S. Laulainen

Multiwavelength turbidities were derived from sunphotometer measurements at Alton Civic Memorial Airport from July 30, 1975 to August 11, 1975. Aerosol loading varied by an order of magnitude during this time with the highest turbidities associated during fog/haze conditions. The turbidity was found to have a wavelength dependence with Ångström exponent α between 1.4 and 2.4.

In support of aircraft sunphotometer measurements, frequent ground observations were made at Alton Civic Memorial Airport from July 30, 1975 to August 11, 1975. Aerosol optical depths (turbidities) were derived from the measurements according to procedures described elsewhere.¹

Multiwavelength turbidities for 8 of the 10 observation days are shown in Figure 1. Least squares fits of the wavelength-dependent aerosol optical depth to a power law of the form

$$\tau = B\lambda^{-\alpha} \quad (1)$$

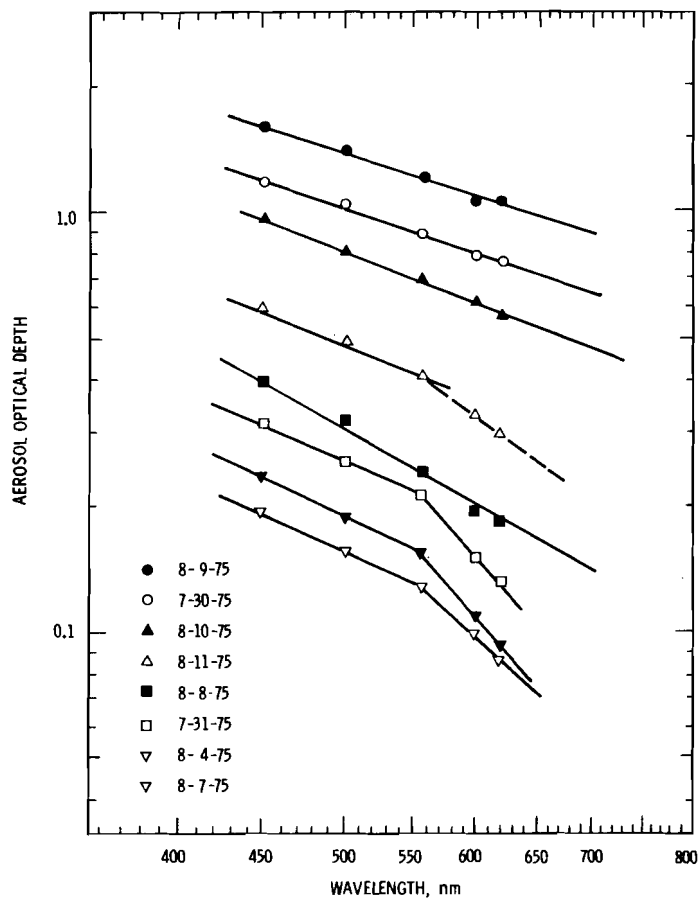
were also carried out. The coefficients B and α are listed in Table 1, where B is related to the aerosol loading and α to the aerosol size distribution. Inspection of Figure 1 and Table 1 shows that large variations in aerosol loading occurred during this time interval. The highest values were associated with fog/haze conditions. The wavelength exponent α varied between 1.4 and 2.4. The apparent discontinuity of the 598.4 and 618.9 nm turbidities for July 31, August 4, 5 and 11 may be an artifact of the sunphotometer, as environmental

TABLE 1. Power Law Fit to the Wavelength Dependent Aerosol Optical Depths Measured over Alton Civic Memorial Airport

Date	α	B
7-30-75	1.4	0.39
7-31-75(a)	1.8	0.076
8-4-75(a)	1.9	0.051
8-5-75(a)	1.9	0.070
8-6-75	2.1	0.053
8-7-75(a)	1.9	0.041
8-8-75	2.4	0.057
8-9-75	1.4	0.53
8-10-75	1.7	0.25
8-11-75(a)	1.7	0.15

a. 598.4 and 618.9 nm data are not included in fit.

temperature control was not always practical during several of the observation intervals.



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FIGURE 1. Multiwavelength Turbidities for 8 of the 10 Observations Days

Reference

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AEROSOLS AND SO₂ MEASUREMENTS
DURING THE DA VINCI BALLOON FLIGHT

A. J. Alkezweeny

Balloonborne measurements of Aitken nuclei, particles larger than 1 μ in diameter, and SO₂ concentrations were made during the da Vinci flight, launched on November 1, 1974. The concentrations of the nuclei and the large particles vary between 200-1900 nuclei/cm³, and 0.069-1.12 particles/cm³ respectively. Generally low concentrations are associated with the higher altitudes. The SO₂ level during the period 00:00 to 03:00 MST is 80 ppb, and no detectable trace of this gas was observed at any other time.

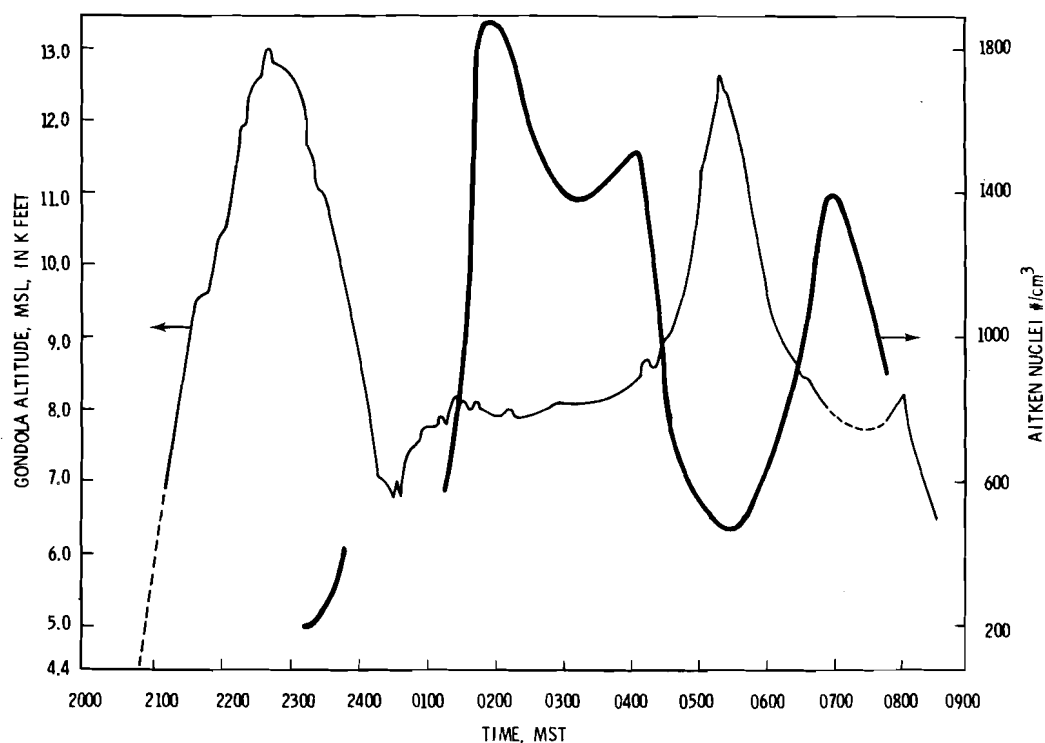
During the da Vinci manned balloon flight, launched on November 1, 1974, several measurements were made to better understand the characteristics of the lower atmosphere and the behavior of its constituents. One important feature of this study was the inter-relationship between the measured parameters. Interpreting the results of the earth's electrical field, solar radiation, and air density experiments require knowledge of atmospheric aerosol particle loading; on the other hand aerosol loading is influenced by atmospheric convection.

The Pacific Northwest Laboratories experiment was designed to measure concentrations of Aitken nuclei (particles <0.1 μ m in diameter), size distributions of particles in the range of 0.3 μ m to about 5 μ m in diameter, and SO₂ concentrations. To avoid contamination from the gondola, air samples were brought up to the instrumentation from about 30 ft below. Aitken nuclei concentration was measured by the Gardner Small Particle Detector. A Royco model 220 optical sensor was used to determine the particle size distribution. This instrument operates on the principle that right angle light scatters from a single particle. Its output pulses were transmitted to the ground and recorded on a magnetic tape recorder. The relationship between pulse height and particle size

was determined by a laboratory calibration with monodispersed latex spheres of known size. The SO₂ level was measured with a Casella Miniature SO₂ Sampler. In this instrument, the SO₂-containing air sample is bubbled through an aqueous solution of H₂O₂. The resulting change in the conductivity over the sampling period is related to the SO₂ concentration. The SO₂ sampler and the Aitken Nuclei Counter are battery and manually operated.

Figure 1 shows the Aitken nuclei concentrations measured during the flight, superimposed on the gondola altitudes. The nuclei concentrations vary between 200-1900 particles/cm³. Generally lower concentrations are associated with higher altitudes in agreement with other observations.¹ It is interesting to compare these data with those measured on the ground at the Mule Park, N.M., site by the ASTA group of White Sands Missile Range. The site elevation was about 8000 ft MSL. Their data show an average of about 1200 particles/cm³, comparable with those measured on the gondola for the same altitude and approximately the same time period.

The average concentration of SO₂ during the period of 00:00 to 03:00 MST time is 80 ppb, and no detectable trace was observed at any other time.



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FIGURE 1. Concentrations of Aitken Nuclei Measured During da Vinci Balloon Flight Superimposed on the Balloon Trajectory

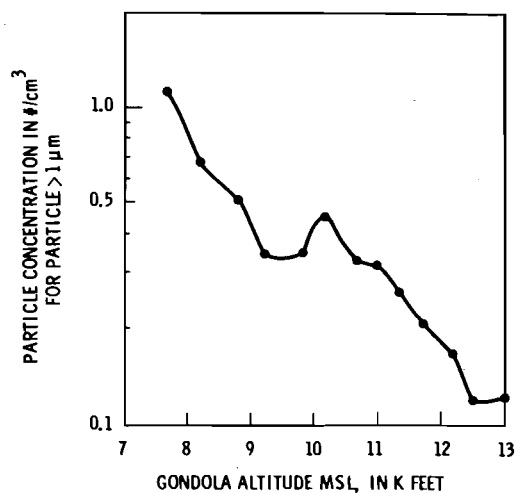
This value for a background SO_2 is high. It is possible that contamination from the gondola may have contributed to it since during the early time of this period the balloon was ascending. This is felt to be unlikely, however, since no detectable SO_2 was measured during the first ascent.

Examination of the aerosol particle data from the tape recorder revealed that the pulses representing particles less than 1μ in diameter are imbedded in noise and unfortunately could not be analyzed. However, the concentrations of particles larger than 1μ in diameter were determined. The results are shown in Table 1. During the first balloon ascent, the counts fluctuated and therefore do not show

any correlation with altitude. However, the particle concentrations measured during the first descent generally increased with the decreasing balloon altitude (Figure 2). This behavior is consistent with other particle profiles measured from aircraft.³ The figure also shows a peak in the particle concentration profile around 10,000 ft. Rawinsonde data taken at Holloman (0200 hr MST) show a peak in the relative humidity profile of about 77% at about 11,000 ft MSL. Therefore, it is possible that the rise in the particle count may have been caused by the rise in the relative humidity. A similar effect was observed by Stampfer² during aircraft aerosol sampling in a relatively unpolluted atmosphere.

TABLE 1. Concentrations of Particles Larger than 1μ in Diameter Measured During the First da Vinci Flight

Time	Altitude $\times 10^3$ ft MSL	Concentration (No./cm ³)
2200	10.4	0.069
2206	10.5	0.085
2212	10.9	0.115
2218	11.8	0.062
2224	12.0	0.093
2230	12.5	0.162
2236	12.6	0.071
2242	13.0	0.124
2306	12.5	0.119
2312	12.2	0.169
2318	11.7	0.206
2324	11.3	0.260
2330	11.0	0.321
2336	10.7	0.326
2342	10.2	0.456
2348	9.8	0.347
2354	9.2	0.342
0000	8.8	0.508
0006	8.2	0.66
0012	7.7	1.12



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FIGURE 2. Concentration of Particle $>1 \mu\text{m}$ as a Function of Gondola Altitude During Descend, Time 2242-0012 MST

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INTEGRATION OF MODELS FOR ATMOSPHERIC PROCESSES

As the pressing demand for energy compels man to consider alternative forms of power production, a careful assessment of the environmental impact of these alternatives becomes critical.

For near-term considerations the two main candidates for steam-electric power production are nuclear energy or fossil fuel combustion. One consideration in assessing the impact of either method is the behavior of potentially harmful contaminants released to the atmosphere. Because the atmospheric processes which affect, directly or indirectly, the fate of a pollutant are extremely complex, practical methods of simulating these processes must be developed to assess the environmental impact of increased or different forms of power production pollution.

Recent evaluations of energy technology development have indicated that increased utilization of high sulfur coal will be required to meet the energy demands until enough nuclear power is available to carry a significant share of the load. For this reason a strong emphasis is presently placed on assessing the health and ecological effects of sulfur dioxide and sulfates. Reported in this section are modeling efforts on this problem, which involve not only transport and dispersion calculations over a multistate area, but chemical transformation and removal by dry deposition and precipitation scavenging. The objective has been to produce a model, containing all these components, which would serve as a vehicle for improvement through sensitivity analyses and experimental verification.

- **SAFETY ANALYSIS AND ENVIRONMENTAL EFFECTS STUDIES**
- **MAP3S MODELING STUDIES**
- **PACIFIC NORTHWEST ENERGY RELATED REGIONAL ASSESSMENT PROGRAM**
- **INVESTIGATION AND MODELING OF THE METEOROLOGY OF THE HANFORD REGION**

AN ANALYSIS OF TRITIUM IN THE ATMOSPHERE
RELEASED BY A CTR*

D. S. Renné, W. F. Sandusky and M. T. Dana

Removal by atmospheric processes of routinely released tritium from a controlled thermonuclear reactor (CTR) has been investigated.

Results of this analysis show that within 50 km of the plant atmospheric concentrations of tritium will be the limiting factor for routine releases. On the regional and global scale, surface water concentrations tend to become the limiting factor. However, both air and surface water tritium concentrations are estimated to be below existing standards during normal commercial CTR operations.

INTRODUCTION

The properties of tritiated water vapor, created when tritium is released into the environment, are essentially the same as those of atmospheric water vapor. Thus tritium becomes a component of the hydrologic cycle, behaving the same as water vapor. The majority of tritium released in the troposphere is limited to the general latitude of release, although some lateral mixing is to be expected.²

The removal of tritium from the atmosphere occurs primarily through precipitation, although near the point of release some tritiated water vapor can be diffused to the ground and taken up directly by vegetation. Once deposited the tritium can infiltrate the ground water, remain as part of the surface runoff and storage, be assimilated into plants, or be transpired and evaporated back into the atmosphere.

* This paper summarizes studies conducted for Division of Controlled Thermonuclear Research of ERDA under Contract No. E(45-1)-1830.

ESTIMATES OF NORMALIZED TRITIUM CONCENTRATIONS IN THE ATMOSPHERE AND SURFACE WATER

Estimated Average Ground-Level Atmospheric Concentrations

The technique used here to compute average annual ground-level normalized concentrations of tritium involves evaluating the relation:

$$x_a/Q = \left\{ \frac{1}{\pi \sigma_y \sigma_z \bar{u}} \exp \left(-y^2/2\sigma_y^2 \right) \right\} \exp \left[-k_{eq}(x/\bar{u}) \right] \quad (1)$$

where:

x_a = atmospheric tritium concentration (Ci-m⁻³)

Q = source term to the atmosphere (Ci-year⁻¹)

σ_y = 0.5t = horizontal standard deviation of the plume (m)

$\sigma_z = 2K_z(x/\bar{u})^{1/2}$ = vertical standard deviation of the plume (m)

t = plume travel time (sec)

K_z = vertical diffusion coefficient
= $5 \text{ m}^2\text{-sec}^{-1}$

$\bar{u}$ = mean wind speed through the layer 300 m-2000 m above ground

k_{eq} = equilibrium scavenging coefficient

x = downwind distance from the source (m)

y = plume crosswind distance (m)

The term in the curly brackets in Eq. (1) was evaluated from a climatology of wind trajectories near Morris, Illinois,³ the assumed location of a CTR for this analysis.

The second exponential term in Eq. (1) represents the depletion of the plume due to scavenging of tritium by precipitation. Under equilibrium conditions, where the in-cloud and below-cloud tritium concentrations are equal, and where the height of the release is at ground-level, the equilibrium scavenging coefficient k_{eq} can be obtained from:⁴

$$k_{eq} = \frac{J}{H^*} \left(\frac{2}{\pi} \right)^{1/2} \frac{1}{\sigma_z} \quad (2)$$

where, for Morris Illinois:

σ_z' = Smith-Singer⁵ dispersion coefficient

$$= 0.63x^{0.86}, \quad x \leq 4.4 \text{ km}$$

$$= 859 \text{ m}, \quad x > 4.4 \text{ km}$$

The results of the ground-level atmospheric tritium concentration estimates near Morris, Illinois are shown in Figure 1. These concentrations have been normalized to a 1 Ci-year⁻¹ release. Regional normalized concentrations using the same conditions for Eq. (1) and (2) are shown in Figure 2.

Estimated Average Surface Water Concentrations

The average annual surface water tritium concentrations, x_g , are determined from:

$$x_g = Q \frac{1 - \exp[-k_{eq}(x/u)]}{AW} \quad (3)$$

where:

W = average annual depth of the surface or groundwater mixing layer (m-year⁻¹)

A = area of precipitation (m²)

Since this analysis is intended to provide conservative estimates of tritium concentrations that would show the maximum expected impacts for normal operations, the value of W was assumed to be determined from the mean annual net rainfall:

$$\frac{AW}{\text{year}} = P - E \quad (4)$$

where:

P = amount of precipitation (m³-year⁻¹)

E = amount of evaporation (m³-year⁻¹)

Equation (4) shows that estimates of surface water concentrations of tritium are made by determining the amount of the source term that is depleted by washout. Thus, estimates of surface water concentrations were made for each concentric areal ring around a CTR located in Morris, Illinois from the relation:

$$x_g \Big|_{x=j-i} = Q \left[(1 - f_{x=j}) - (1 - f_{x=i}) \right] \quad (5)$$

where:

$$f = \exp \left[-k_{eq} \left(\frac{x}{\bar{u}} \right) \right]$$

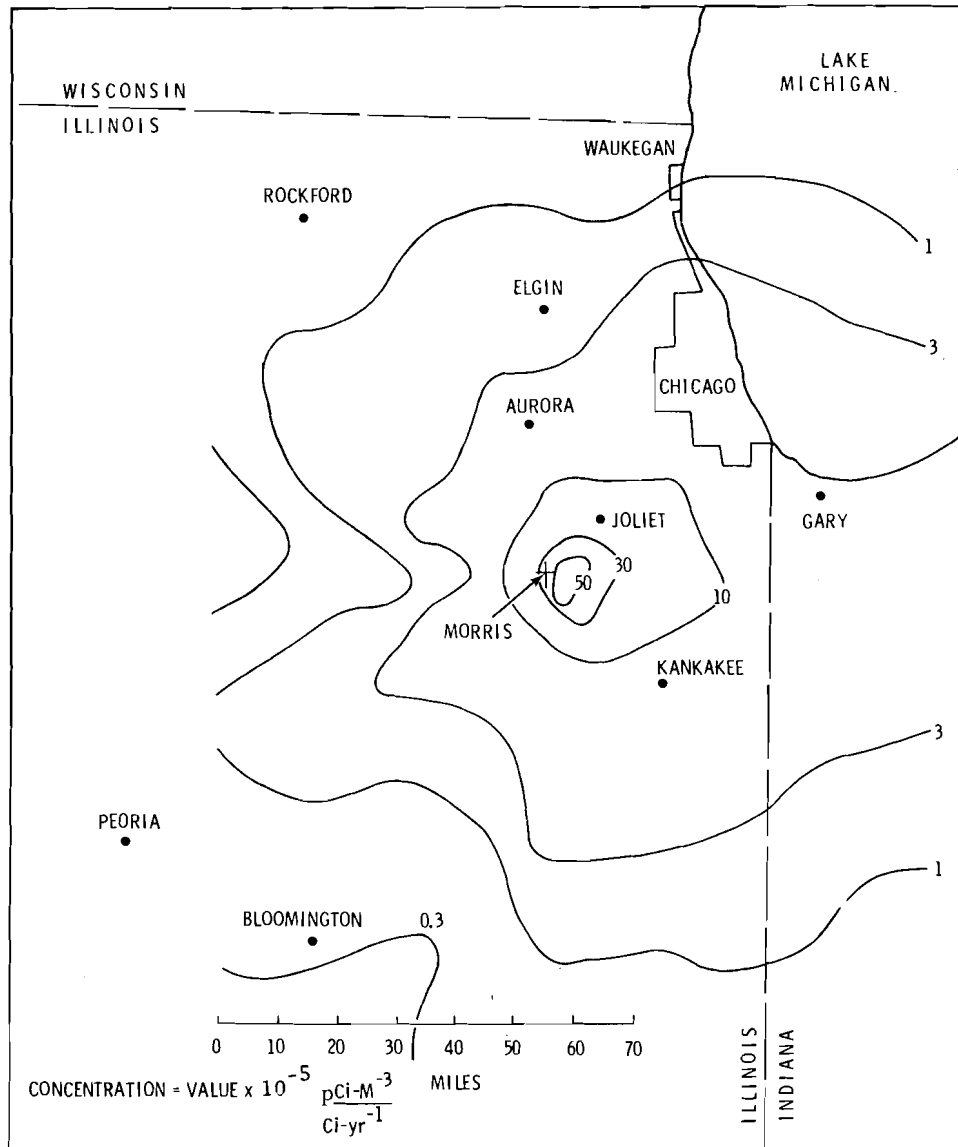


FIGURE 1. Ground-Level Air Concentration Values of Tritium for an Area Near Morris, Illinois

- i = radial distance to inner circumference of a concentric areal ring
- j = radial distance to outer circumference of a concentric areal ring

This technique has the disadvantage of assuming that all precipitation systems are distributed equally around a CTR, along with the distribution of $^3\text{H}_2\text{O}$.

The result of the surface water tritium concentrations using an iteration of Eq. (5) and (3) are shown in Table 1. Again normalized concentrations are provided, based on a normal release of 1 Ci-year^{-1} . Figure 3 shows the variation of surface water concentrations with distance from the source using the hydrologic conditions at Morris. The value of σ_z' during precipitation episodes were obtained from the LMFBR analysis.⁷ For

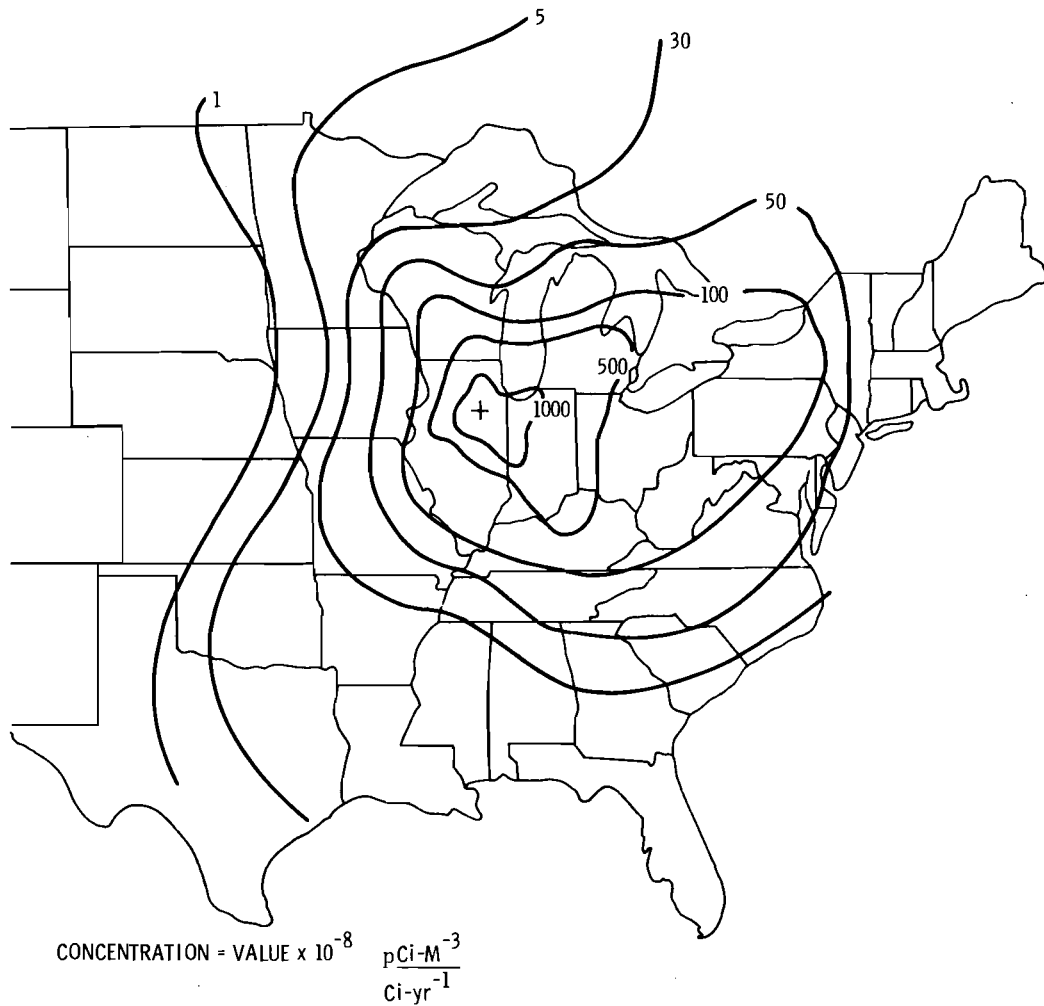


FIGURE 2. Ground-Level Air Concentration Values of Tritium for the Eastern United States

TABLE 1. Surface Water Tritium Concentrations (x_g) for Various Distances from the Source

Radial Distance (Km)	Δf^*	Area (m^2)	$\frac{x_g/Q}{\text{pCi-l}^{-1} \text{ ci-yr}^{-1}}$
0-50	.0317	$.785 \times 10^{10}$	10.9×10^{-3}
50-100	.0307	2.36×10^{10}	3.52
100-150	.0298	3.93×10^{10}	2.05
150-200	.0287	5.50×10^{10}	1.41
200-250	.0279	7.06×10^{10}	1.07
250-300	.0270	8.64×10^{10}	.844
300-500	.0997	5.02×10^{11}	.537
500-1000	.1995	2.36×10^{12}	.228

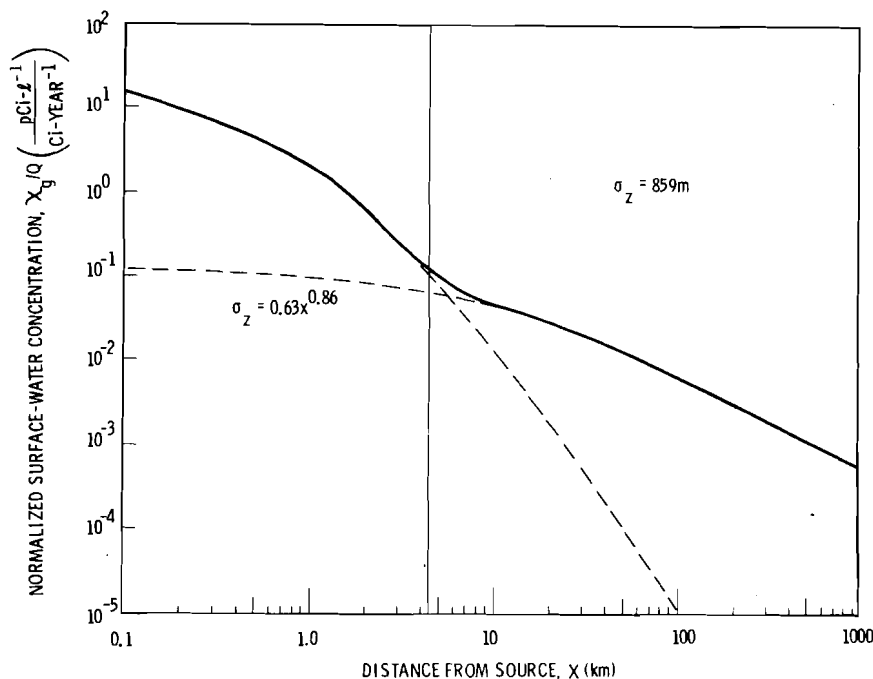


FIGURE 3. Normalized Surface Water Concentration (x_g/Q) as a Function of Distance (x)

Morris, the value of σ_z' increases to distance of 4.4 km, and is constant beyond that point.

Estimated Latitudinal and Global Average Atmospheric and Surface Water Concentrations

A technique was developed to analyze the estimated concentrations of tritium in the atmosphere and surface water supplies in the northern hemisphere based on a continuous normal operation of CTR plants distributed throughout the 30°-50° latitude belt. The analysis is based on Reference 8 with modifications to account for sources near ground level and for the transport of hydrologic components across latitude belts.⁴

Table 2 summarizes model results of estimated atmospheric and surface water concentrations.

SUMMARY

The information in Figures 1 and 2 and in Tables 1 and 2 allow computations of the maximum allowable emissions (MAE) of a CTR based on the analysis procedure used here and the

maximum permissible concentrations (MPC) that have been developed for $^3\text{H}_2\text{O}$. For atmospheric tritium, the MPC is $4 \times 10^{-6} \text{ Ci-m}^{-3}$, and for tritium in the drinking water supply the MPC is $1 \times 10^{-4} \text{ Ci-l}^{-1}$. Thus a value for the maximum allowable emission for a CTR is the ratio of the MPC to the normalized atmospheric or surface water concentration estimate:

$$\text{MAE} = \frac{\text{MPC}}{x_a/Q \text{ or } x_g/Q} \quad (6)$$

A summary of maximum allowable emissions based on atmospheric and surface water estimates is given in Table 3. It is evident from this table that atmospheric concentrations represent the limiting factors for a CTR operation near the plant, while surface water concentrations represent the limiting factors at great distances from the plant. However, it is also evident that large quantities of tritium releases could be tolerated according to this analysis--quantities that far exceed the emission currently expected by a CTR technology.

TABLE 2. Average Ground-Level Atmospheric and Surface Water Concentrations by Latitude Belt for Land and Ocean Areas Incorporating the Global "Background"

Latitude (°N)	LAND (W=0.5m)		OCEANS (W=75m)	
	$\frac{x_a}{Q} \frac{\text{PCi-m}^{-3}}{\text{Ci-yr}^{-1}}$	$\frac{x_g}{Q} \frac{\text{PCi-l}^{-1}}{\text{Ci-yr}^{-1}}$	$\frac{x_a}{Q}$	$\frac{x_g}{Q}$
0-10	2.72×10^{-6}	2.42×10^{-4}	3.84×10^{-8}	6.65×10^{-7}
10-20	2.72	2.59	4.00	6.79
20-30	2.82	2.64	6.46	8.63
30-50(a)	3.03	2.69	18.96	32.42
50-60	2.83	2.47	7.50	7.14
60-70	2.74	2.43	4.29	6.65
70-80	2.70	2.42	3.80	6.65
80-90	2.69	2.42	2.74	6.65
Global(b)	2.68	2.42	3.71	6.65

a. Source latitude belt.

b. "Background."

TABLE 3. Summary of Maximum Allowable Emissions (MAE) of Tritium to the Atmosphere by a Normal CTR Operation Based on Atmospheric (x_a) and Surface Water (x_g) Concentrations

	$\frac{x_a}{(\text{Ci-yr}^{-1})}$	$\frac{x_g}{(\text{Ci-yr}^{-1})}$
Local (near Morris, Ill.)	6.2×10^9	9.2×10^9
Regional (eastern U.S.)	5.0×10^{10}	2.8×10^{10}
Global	1.6×10^{12}	3.7×10^{11}

CONCLUSIONS

This analysis has shown that the normal operation of a commercial CTR in the vicinity of Morris, Illinois should have a small impact on the environment in terms of increased tritium concentrations in the air and surface water. This, of course, assumes that CTR operations do not emit quantities of tritium exceeding 10^3 kg/year.

Several topics requiring additional research have been identified during the completion of this analy-

sis. More sophisticated modeling techniques for transport over several hundred km are required, particularly if complex terrain and land-sea contrasts are to be included. For example, some success has been obtained using numerical boundary layer models to estimate tritium concentrations in the vicinity of the Savannah River Laboratory.⁹ The manner in which $^3\text{H}_2\text{O}$ enters the hydrologic cycle, and the similarities between $^3\text{H}_2\text{O}$ and H_2O need further quantification. This becomes particularly important when examining the scavenging of $^3\text{H}_2\text{O}$ by pre-

precipitation, and the subsequent return to the atmosphere via evapotranspiration. Research into the probabilities

of large accidental releases during unfavorable meteorological episodes needs to be undertaken.

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A REGIONAL SCALE MODEL FOR COMPUTING DEPOSITION
AND GROUND LEVEL AIR CONCENTRATION
OF SO₂ AND SO₄ FROM AN ELEVATED SOURCE

L. L. Wendell, D. C. Powell and R. L. Drake

A computer model has been developed which utilizes time-changing gridded wind field data and gridded hourly precipitation data to compute average air concentrations and amounts of SO₂ and sulfate deposited from a continuous elevated release of SO₂. The model simulates atmospheric transport, chemical transformation, and removal by dry deposition and precipitation scavenging.

INTRODUCTION

During the past year the most urgent regional pollution problem associated with power generation has been generation of sulfur compounds by fossil fuel combustion. Both the government and private industry are sponsoring programs to investigate the environmental impact of the SO₂-sulfate cycle as the demand for energy requires continued and increased use of sulfur-bearing fuels.

The seriousness and complexity of this problem have been pointed out in a lengthy report prepared for the Committee on Public Works of the United States Senate.¹ The report indicates that oxidation products of SO₂, including sulfuric acid and suspended particulate sulfates are more toxic than the parent compound. Thus the problem takes on multistate proportion, introducing formidable challenges to the development of a realistic assessment model.

The initial considerations and beginnings of this modeling effort have been previously described.² A model has now been developed and subjected to initial testing. The model is designed to use time-and space-varying gridded wind and precipitation data, along with user-specified source locations and source strengths to compute average air concentrations and deposited amounts of SO₂ and sulfate. The

model simulates transport and dispersion, chemical transformation, and wet and dry removal. In this first version of the model the simulations of the transformation and removal processes are of a simple linear nature. They do, however, allow some investigation of the sensitivity of the overall process to its interacting parts. Options have been left open to allow more sophisticated treatment when the appropriate knowledge becomes available from experimental programs. The description of the model in this report will be brief; more detailed documentation for formal presentation is in progress.

TRANSPORT AND DISPERSION

The transport of material in this model is purely kinematic and is accomplished using similar techniques as developed for mesoscale purposes. The model is designed to accept a time series of gridded winds, assumed to be representative of the layer carrying the contaminant. The gridded wind fields for this work are currently generated from 12-hourly radiosonde data. The model approximates a continuous plume centerline by a series of particles released from selected source locations, at 1-hr intervals, into wind fields obtained by hourly interpolation between the 12-hourly observations. An example of the result of this procedure is displayed graphically in

Figure 1. The positions of the particle are computed on an hourly basis.

The plumes depicted in Figure 1 are analogous to a satellite picture 12 and 24 hr after release. Despite the fact that this type of representation only approximates what is happening in the atmosphere, it provides some interesting depictions of plume stretching and overlapping in a time- and space-varying flow on this scale.

Dispersion of the mass of material associated with each particle is allowed to develop in a Gaussian fashion. The variable σ_z is approximated

as described previously.² The variable σ_y is approximated with the form

$$\sigma_y = ax^b$$

where x is distance traveled by the particle of concern and the values of a and b are selected according to stability class to reproduce the Pasquill-Gifford curves.⁴ The plume widths shown in Figure 1 are determined by assuming the plume edges to be $2\sigma_y$ from the centerlines, making a total width of $4\sigma_y$.

The average surface air concentrations and the deposition quantities are calculated by sampling the approximated plumes over a network of grid squares, approximately 34×34 km. The model has been constructed with the option of neglecting horizontal diffusion about the plume centerline. This is an assumption made by others in long-term assessment models on a similar spatial scale.⁵ Preliminary tests comparing runs with and without horizontal plume diffusion indicate that the extra expense (factor of 4) is probably not justified for long-term assessments.

TRANSFORMATION

The chemical processes involved in the transformation of SO_2 to sulfates, including sulfuric acid, in the atmosphere are complex and the subject of considerable investigation.¹ However, for the current version of the model the oxidation reaction has been assumed to be linear with a user-specified fraction per hr.* This rate could be made to vary with relative humidity or the presence of other chemical species, but the present assumption is probably in line with the assumptions regarding diffusion.

DRY DEPOSITION

The dry removal processes involve 1) the absorption of gaseous SO_2 by

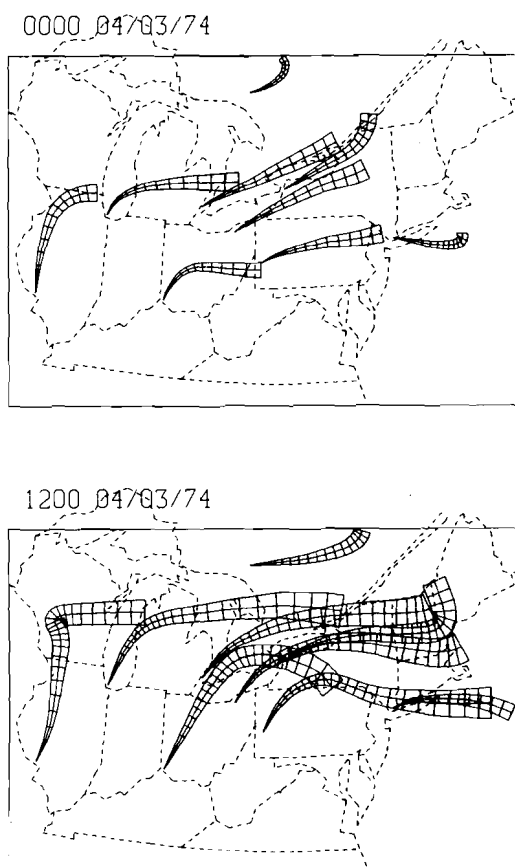


FIGURE 1. Twelve-Hourly Plume Plots from Nine Sites with Continuous Releases Beginning at 1200 GMT April 2, 1974. The lines perpendicular to the centerlines indicate plume segments generated during 1 hr of release time.

* See accompanying article, "Chemical Deposition and Transformation Parameterizations for Regional Models," by R. L. Drake, this volume.

the soil or vegetation and the 2) deposition of particles containing sulfates. These processes have also been assumed linear with user-specified deposition velocities.* No accounting is made at present for spatial variation of roughness or vegetation type but the computer code can easily be adapted to account for this when experimental programs provide adequate data to do so. The current version of the model uses source depletion instead of a surface depletion in the dry deposition calculations. For a deposition velocity of 1 cm/sec and a transport velocity of about 10 m/s the discrepancy would be on the order of 30%.⁶ The error would increase for lower transport velocities. Methods for correcting this problem are being examined and evaluated; an appropriate method will be incorporated into the model to more realistically approximate the surface depletion.

PRECIPITATION SCAVENGING

The processes of wet removal are also very complex and the subject of much study.⁷ Most investigators developing assessment models either avoid the problem completely or use a blanket depletion using climatological average precipitation rates. One of the reasons for this is that hourly precipitation records are stored in a format not readily converted for use in a real-time assessment model.* An important question to be answered in this regard is whether the use of real time precipitation is essential to making a realistic assessment.

The model in its present form will accept either gridded hourly precipitation rates or average precipitation rates. An example of the effects of the real-time precipitation data is shown in Figure 2. For the purposes of this graphical display any portion of the plume encountering a precipitation rate greater than 0.01 in.-hr⁻¹ was immediately dropped. Precipitation scavenging is incorporated into the assessment model through the application of a washout

coefficient which is directly proportional to the precipitation rate.⁷ In-cloud or below-cloud scavenging are not included in the present version of the model, however methods are under investigation.

ILLUSTRATION OF COMBINED TRANSFORMATION AND REMOVAL EFFECTS

The simultaneous occurrence of transformation of SO₂ to sulfate with wet and dry removal of SO₂ and sulfate at different rates for each can be difficult to visualize. The effects of these combined processes as they are approximated in the model for a single plume element are shown in Figure 3. The main feature which is apparent in the diagram is the limiting effect the removal processes have on the SO₂ available for conversion to sulfate which, because of its postulated lower removal rates, can be carried long distances from the source. This is especially apparent in comparing Figure 3b and 3c. The relative magnitudes of the fractions of the original SO₂ in the air as sulfate and deposited at the surface, are dramatically reversed as wet removal is added as a continuous process. It is interesting to note that in the wet removal by an intermittent process the effect on the amount of SO₂ available for conversion to sulfate would depend strongly on the travel time available before precipitation is encountered. This effect is currently under investigation.*

DISCUSSION

The present model contains the essential elements, in varying degrees of approximation, to begin validation and assessment exercises. Our approach in this modeling effort is to maintain a balance between development and application. Our objective is to produce a model which will serve as a vehicle to be improved

* See accompanying article, "Preparation of Hourly Maps of Gridded Precipitation Scavenging Calculations," L. L. Wendell, and C. E. Hane, this report.

* See accompanying article, "An Examination of the Effects of Real Time Versus Time Averaged Precipitation Data on SO₂ and SO₄ Removal in the Regional Assessment Model." L. L. Wendell and D. C. Powell, this report.

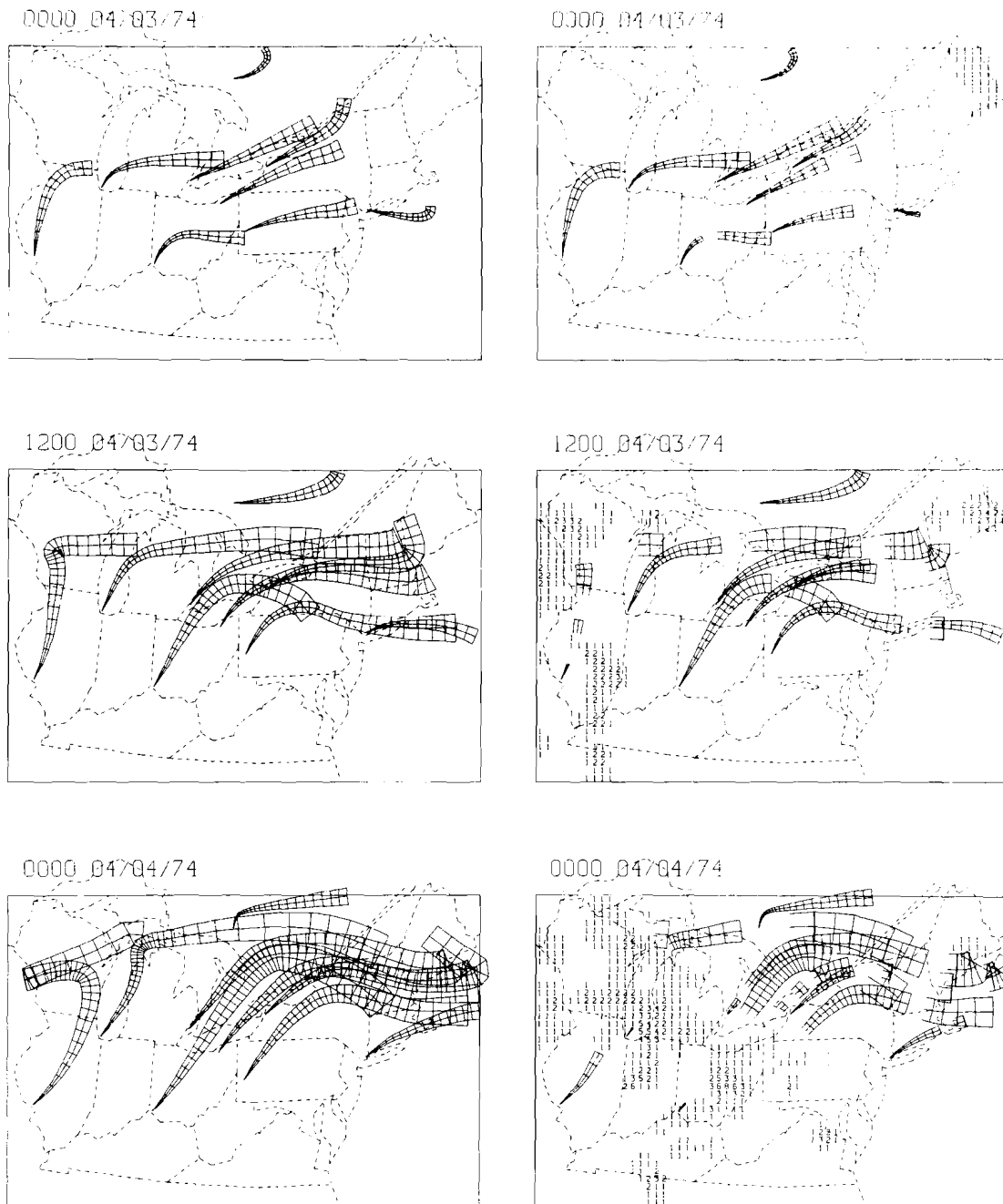
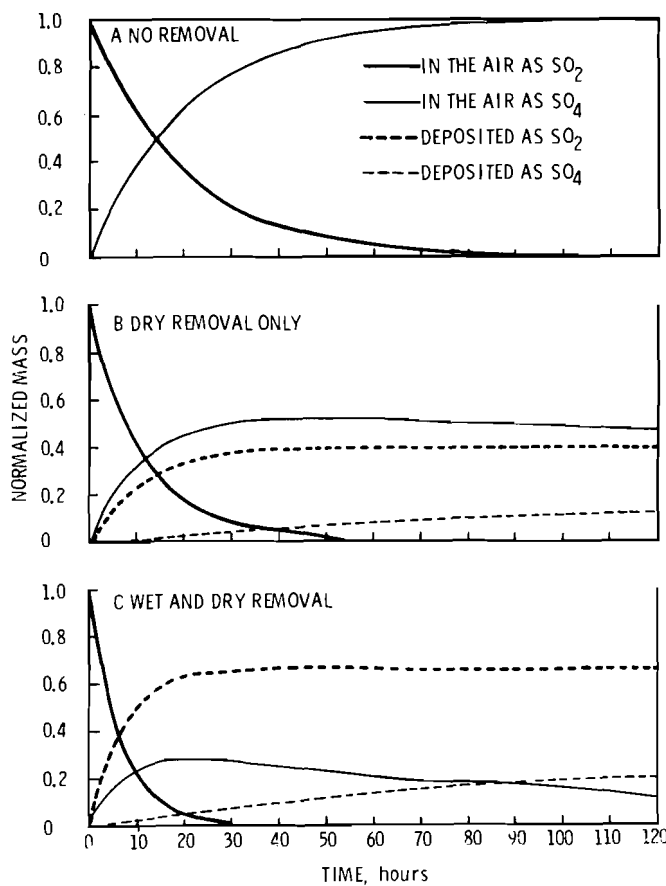


FIGURE 2. Plume Positions Shown at 12 hr Intervals for a Release Starting at 1200 Z April 2, 1974. In the plots on the left (a) Dry - no precipitation is considered. In the plots on the right (b) Wet - hourly precipitation is allowed to eliminate those portions of the plumes encountered. The numbers on the plots in b) are coded as follows: 1: 0.01-0.10 in., 2: 0.11 - 0.20 in., 3: 0.21 - 0.30 in., etc.



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FIGURE 3. The Fate of the Normalized Mass of SO_2 in an Air Parcel Is Depicted as a Function of Time for a) No Removal, b) Dry Removal only and c) Wet and Dry Removal simultaneously. The coefficients applied are as follows: $\text{SO}_2 \rightarrow \text{SO}_4$ transformation, 0.05 hr^{-1} ; dry deposition velocities, 1 cm sec^{-1} for SO_2 and 0.1 cm sec^{-1} for SO_4 ; wet removal coefficients, 0.065 hr^{-1} for SO_2 and 0.0065 for SO_4 .

upon. This approach allows the validation efforts, the assessment exercises and comparisons with other models to suggest the priority of the required improvements. These efforts,

coordinated with the efforts of other ERDA-supported laboratories should provide an essential tool to aid in making the decision required to establish fossil fuel energy policy.

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AN EXAMINATION OF THE EFFECTS OF REAL TIME VERSUS TIME-AVERAGED
PRECIPITATION DATA ON SO₂ AND SO₄ REMOVAL
IN THE REGIONAL ASSESSMENT MODEL

L. L. Wendell and D. C. Powell

The regional scale diffusion model was exercised to produce deposition patterns of SO₂ and sulfate for an 18-day period, using for one run the hourly precipitation as it occurred during the period, and for another run the average precipitation pattern for the same period. A comparison of the results indicated more total removal by the average precipitation pattern but significantly greater deposition in localized areas by the hourly precipitation.

INTRODUCTION

Including the precipitation scavenging process for SO₂ and SO₄ into an assessment model has taken several forms. The simplest approach is to apply a wet removal rate that is constant both in time and over the area of interest.¹ A more sophisticated approach has been to apply a washout coefficient which is proportional to the climatological rainfall rate.² In both of these approaches the removal by precipitation scavenging is a continuous process as if rain were falling, however lightly, over the entire area of concern for the whole period of the assessment. This assumption is obviously not realistic and would tend to remove more material than stronger, but intermittent rains, and do so in a more uniform manner.

One could argue that from a statistical point of view that over a long enough period of time the differences in wet deposition due to the intermittency of real time precipitation would average out. This argument, however, assumes little or no correlation between the air flow and precipitation patterns. The cyclical nature of the frontal storms through latitudes indicates such a correlation is possible and makes the problem worthy of closer examination.

A sensitivity analysis was proposed utilizing the Regional Model

for transport transformation and removal developed for SO₂-sulfate assessment.* The simple transformation and removal parameterizations of the current model were not modified for this analysis. Complexities such as a transformation rate varying with relative humidity³ or SO₂ coming out of solution in falling rain drops and reentering the air⁴ are left for future analyses.

DEFINITIONS OF REMOVAL MODES UNDER CONSIDERATION

The model was run with three removal alternatives for purposes of comparison. First the dry removal process was modeled exclusively. Second, both dry and wet removal were included; wet removal being modeled as the result of time-averaged precipitation. For this process the precipitation rate used in each grid square was the time-averaged amount occurring during the specified 18-day interval. Third, a real-time precipitation was substituted for the time-averaged values. In this process new rainfall rates for each grid square were entered hourly into the computation process. By comparison of the concentration and deposition patterns predicted by these last two

* See accompanying article, "A Regional Scale Model for Computing Deposition and Ground Level Air Concentration of SO₂ and SO₄ from an Elevated Source."

processes, it is possible to assess the effects of the correlation between wind and precipitation patterns--also the effects of locally heavy precipitation through a densely polluted layer.

DESCRIPTION OF DATA AND PARAMETERS

The meteorological data used for this test included a set of 12 hourly gridded wind fields from 1200 GMT April 2, 1974 through 1200 GMT April 20, 1974. The gridded winds were obtained through a random to grid interpolation of radiosonde winds averaged through a layer from 100 m to 1000 m. Hourly precipitation data from 777 reporting stations were gridded from this same period for the precipitation scavenging calculations.

Since our main concern for this test was the effect of real time versus average precipitation, the parameters were kept as simple as possible. A neutral stability class was maintained for the diffusion calculations throughout the period. The growth of σ_z was not limited by a varying mixing depth.

The continuous release was prescribed to be from a single location just east of St. Louis, Missouri at a rate of 100,000 tons/year⁻¹. The effective release height was held at 400 m above the surface for the entire period.

The rate constant for the transformation of SO₂ to sulfate was set

at 0.05 hr⁻¹. The dry removal deposition velocity was set at 1 cm sec⁻¹ for SO₂ and 0.1 cm sec⁻¹ for SO₄. The parameters for wet removal were specified as

$$\Lambda_2 = \frac{P}{2} \text{ hr}^{-1} \text{ for SO}_2$$

$$\Lambda_4 = \frac{P}{20} \text{ hr}^{-1} \text{ for SO}_4$$

where P is the rainfall rate in mm hr⁻¹. For the relatively light rainfall rate of 0.1 mm hr⁻¹, $\Lambda_2 = 0.05 \text{ hr}^{-1}$ and $\Lambda_4 = 0.005 \text{ hr}^{-1}$. For a moderate rain of 5 mm hr⁻¹, $\Lambda_2 = 2.5 \text{ hr}^{-1}$ and $\Lambda_4 = 0.25 \text{ hr}^{-1}$.

During the 18-day period of the test, six frontal storms passed through the area covered by the computational grid with precipitation amounts varying from light (<2.5 mm hr⁻¹) to heavy (>25 mm hr⁻¹) and a high degree of spatial variability. However, when the rainfall was averaged in each grid square for the entire period the resulting pattern was relatively uniform.

TEST RESULTS

Total Budget

To show the effects of real time versus average precipitation on the model results, the total SO₂-sulfate budgets at the end of the 18-day test are presented for the three modes of removal in Table 1. The fractions of

TABLE 1. Fate of SO₂ Released for 18 Days from a 400 m Effective Height (%)

Precip. cond.	Left in air over grid		Deposited on surface of grid		Transported beyond grid		Total
	SO ₂	SO ₄	SO ₂	SO ₄	SO ₂	SO ₄	
Dry	2.7	7.7	33.6	4.6	10.7	41.3	100.0
18-day average	1.4	3.7	62.3	7.8	4.1	20.7	100.0
Real time	2.0	6.1	50.0	8.8	5.1	28.0	100.0
Real time - average	+0.6	+2.4	-12.3	+1.0	+1.0	+7.3	0.0

the total SO_2 released which remain in the air as SO_2 and SO_4 at the end of the period are presented only to show the complete budget. These fractions will decrease as the duration of the release increases.

The quantities of interest are the amounts removed and the amounts escaping the computational grid. Note that amounts removed in the form of SO_2 and SO_4 reflect the larger removal parameters for SO_2 than for SO_4 . The difference in removal rates also appears in the amounts which escape the grid, 4-5 times more of the original SO_2 escape the grid as SO_4 than in the original form.

In comparing the results for the three modes of removal we find some distinct differences. The case with no precipitation was included to serve as a reference when comparing the two modes of wet removal. However, this is only a guide because the results from dry removal cannot be directly extracted from the combined wet and dry removal, especially in the case of real time precipitation. It is apparent that using the 18-day average precipitation rate continuously in each grid square produces a significantly higher removal of SO_2 than applying the precipitation rates as actually measured during the period. When both modes of wet removal for SO_2 are compared to the dry removal the increase affected by the real time precipitation is about 50% while that effected by the 18-day average is about 90%. This over-removal by the average precipitation is reflected in the fact that 35% more SO_4 escaped the grid when real time precipitation was used. That more SO_2 is available for transformation in the real time precipitation case is also evident from the higher removal of SO_4 than for the average precipitation case.

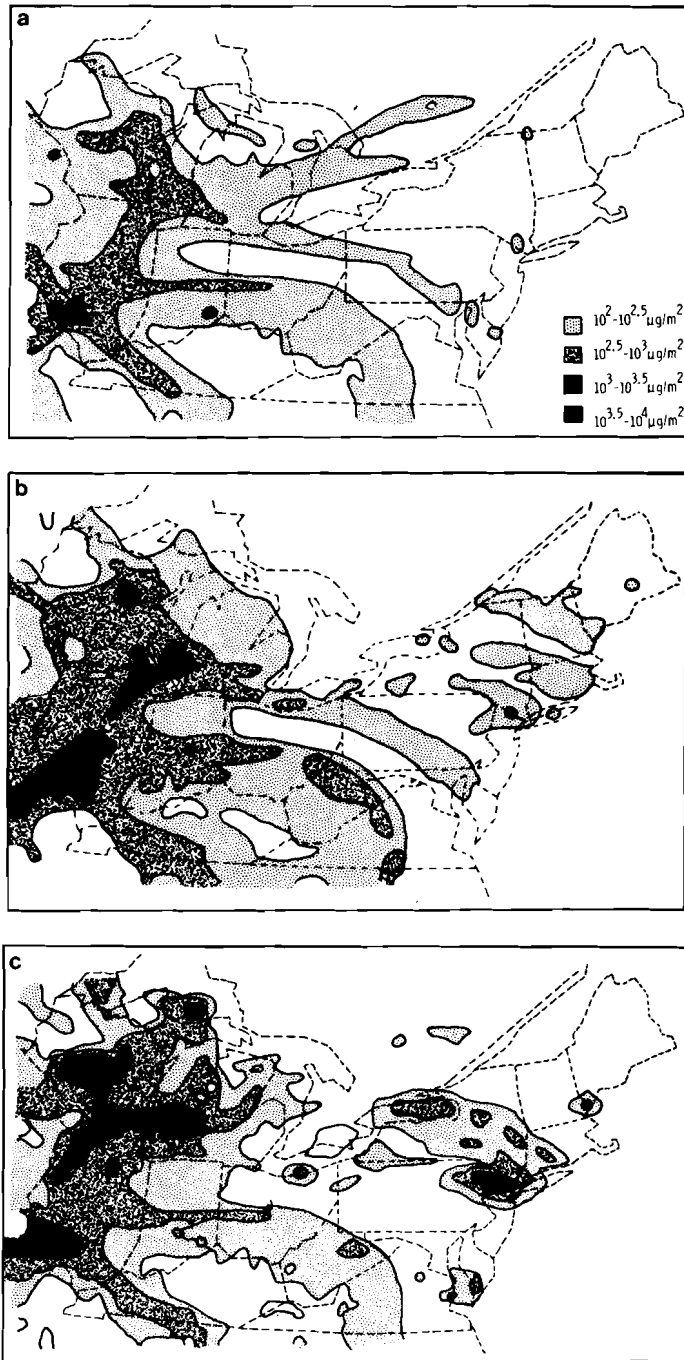
Spatial Distribution

Maps were produced for the three modes of removal which displayed the spatial distribution of the average air concentrations for SO_2 and SO_4 as well as the deposited amounts of each. The major points from those maps (not shown here) are:

- The highest 18-day average air concentrations for SO_2 were on the order of $1 \mu\text{g m}^{-3}$ within 50-100 km of the source, dropping off rapidly with distance. There was no significant difference in the spatial variability between the three removal modes.
- The highest 18-day average air concentrations for SO_4 were on the order of $0.3 \mu\text{g m}^{-3}$ within 50-200 km of the source depending on the removal mode. The SO_4 patterns contrasted sharply with the SO_2 patterns in that the concentrations dropped off much more slowly with distance from the source and showed a greater variation in distribution with removal mode.
- The patterns for the SO_2 deposited over the 18-day period were similar to the patterns for the SO_2 average air concentrations, with major amounts deposited within 100-200 km of the source. Deposited amounts near the source were on the order of $10,000 \mu\text{g m}^{-2}$ for the dry removal mode and $30,000 \mu\text{g m}^{-2}$ for the two modes including wet removal. Again the patterns for all three modes were very similar with deposited amounts dropping to less than $100 \mu\text{g m}^{-2}$ beyond a 1000 km radius from the source.

The patterns of deposited SO_4 for the three modes of removal are shown in Figures 1a through 1c. The pattern for the dry mode shown in Figure 1a is similar to the pattern for the average air concentration of SO_4 and should reflect the major paths of the material over the area during the period. The second mode of removal including precipitation scavenging on a continuous basis with the 18-day average rainfall rate is shown in Figure 1b. The pattern for this case is very similar to the one for the dry mode, except for the amplification which reflects the 70% increase shown for the total budget in Table 1. There are additional amounts shown deposited in New England which would be expected from the pattern on the air concentration map.

The pattern of SO_4 deposition produced using the removal mode, including real time precipitation is shown in Figure 1c. The pattern for the



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FIGURE 1. SO_4 Deposition in $\mu\text{g m}^{-2}$ for a) Dry Removal Only, b) Wet and Dry Removal Using Average Precipitation, c) Wet and Dry Removal Using Real Time Precipitation.

lower half of the grid is recognizable from the patterns of the first two removal modes but the pattern in the upper half of the grid has some striking differences. In the northwestern portion of the grid the major difference is the order of magnitude greater deposit in west central Wisconsin. The other major difference in pattern is in the New York, New Jersey and New England area. For the real time precipitation mode there is an order of magnitude more SO_4 deposited in the New York and New Jersey but significantly less in the New England states.

Probably the most striking feature of the SO_4 deposition, for the parameters used, is that the $1000 \mu\text{g m}^{-2}$ being deposited 1500 miles from the source is of the same order of magnitude as the amount being deposited no more than 50-100 km from the source.

DISCUSSION AND RECOMMENDATIONS

Considering the complexities of the problem and the crude parameterizations in the model during this single sensitivity analysis, a fair degree of caution must be exercised in drawing conclusions from the results. In this test we have considered the case of a single source of SO_2 which emits 100,000 tons of SO_4 per year under the conditions including a slow to average rate of conversion and removal efficiencies about an order of magnitude greater for SO_2 than for SO_4 . Under these conditions and simplifying assumptions with respect to stability conditions and mixing depth, the model has produced results for an 18-day period from which we would make the following observations:

- A single plant of this size in the designated area does not add a significant amount of SO_2 or SO_4 to the background level of average

air concentration beyond the immediate vicinity of the source.

- Almost 60% of the SO_2 emitted was deposited within the northeastern United States as SO_2 or SO_4
- About 33% of the SO_2 emitted leaves the northeastern United States in the form of SO_4 to be deposited elsewhere.
- The use of time average precipitation in the model causes a significant over-removal of SO_2 , primarily near the source. This allows less SO_4 to be removed at more distant locations within the computational grid or to escape the grid altogether.

The major conclusion with regard to the use of real time or average precipitation is that the most significant discrepancy appears in the spatial distribution of SO_4 deposition. This discrepancy cannot be corrected by a simple change in magnitude of the washout coefficient. It is a discrepancy which would be especially important in the area of control strategy for fuel combustion. Whether the spatial discrepancy in pattern would cancel out over a long term assessment run is still subject to determining a distinct correlation between flow and precipitation patterns. The case we have selected is typical of meteorological conditions in the spring. More cases should be examined during other seasons of the year and for longer periods of time.

As we pointed out, the results obtained in this test depend on the parameterizations of the transformation and removal processes. Because of the wide range and the uncertainty of the results obtained by different investigators, it would be worthwhile to conduct further sensitivity analyses with various combinations of parameters as well as perform validations with experimental data.

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PREPARATION OF HOURLY MAPS OF GRIDDED PRECIPITATION DATA FOR USE IN PRECIPITATION SCAVENGING CALCULATIONS

L. L. Wendell and C. E. Hane

A method for producing hourly fields of gridded precipitation data from randomly spaced reporting stations is developed. The method is tested on 20 days of data from a network of 777 stations in the MAP3S study area in the northeastern United States.

INTRODUCTION

It is generally accepted that precipitation is the principal route by which sulfates are returned to earth.¹ Any analysis of the sulfur cycle, therefore, should contain information about the precipitation over the area of concern. A question has been raised about the importance of reconstructing the time changing fields of precipitation as opposed to using average climatological data in assessment models which include precipitation scavenging as a removal process.*

Precipitation scavenging of atmospheric contaminants involves many

complex processes.² Precipitation rate measured at the ground at a specific location may not completely represent these processes, but it should provide a better first estimate of the wet removal, at any particular time, than the climatological average rate for that location.

To provide precipitation data in a form which may be utilized in a numerical simulation of the processes affecting an atmospheric contaminant some prior analyses of the data from the randomly spaced collection of reporting stations is required. This report briefly describes a technique developed for these analyses.

DATA

Hourly precipitation data for stations throughout the United States is available from the National Climatic Center, at Ashville, North Carolina.

* See "An Examination of the Effects of Real Time Versus Time Averaged Precipitation Data on SO₂ and SO₄ Removal in the Regional Assessment Model," by L. L. Wendell and D. C. Powell, this volume.

The density of hourly precipitation-reporting stations is somewhat greater than for stations reporting standard meteorological information. The station density for the computational grid in the northeastern U.S. is shown in Figure 1. The few stations shown for the segment of Canada which is on the grid were obtained from the Atmospheric Environment Service in Downsview, Ontario.

The hourly data at Ashville may be obtained by state in printed documents or on magnetic tape. The tapes contain long time series (20-25 years) data for each station. Sorting and merging to get simultaneous data for over 800 stations would be a major project in itself. To avoid this difficulty for a preliminary test, the data for April 1 through April 20, 1975 were extracted from the printed documents, punched on cards, edited, sorted by day and written to tape for future analysis.

RANDOM-TO-GRID-DATA ANALYSIS

The hourly data from the randomly spaced collection of stations shown

in Figure 1 were utilized to provide hourly precipitation rates in the grid squares shown in Figure 2. This will allow material to be removed from plumes and deposited in each grid square according to the amount of contaminant as well as the precipitation rate in the grid square.

The basis for the interpolation technique was simply to compute the weighted average of the 3-5 stations closest to the center of each grid square. The weighting factor selected was the inverse square of the distance of the stations from the center of the grid square.³ The second power of inverted distance was considered appropriate because, for many of the grid squares, most of the stations in the interpolation for a given square lie outside the boundaries of the square. For this reason, some preference for the data at the closer stations was deemed appropriate, but higher powers of the inverted distance seemed to allow more preference than necessary to the closer stations especially in areas of high station density.

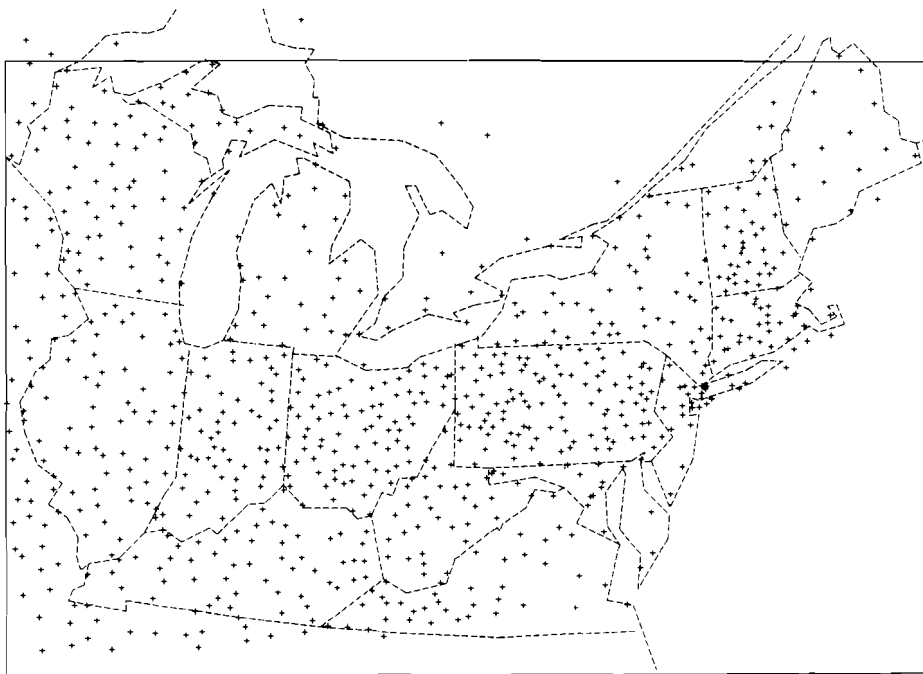


FIGURE 1. Distribution of Hourly Rainfall Measurement Stations over the Computational Period

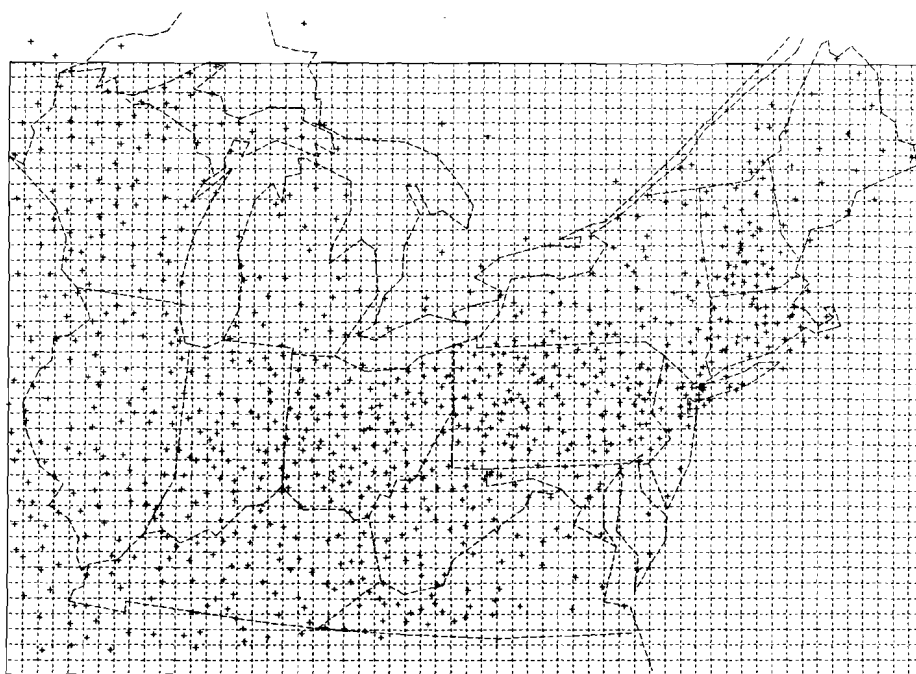


FIGURE 2. Grid Network Used to Digitize Rainfall Data from the Randomly Spaced Stations

A 60 x 40 matrix of gridded precipitation rates was produced for each hour of the 20-day test period and written to tape for use in assessment models.*

DISCUSSION AND RECOMMENDATIONS

Hourly precipitation analyses were examined at 6-hr intervals throughout the 20-day sample. Highly localized patterns, which moved across the grid with the frontal storms, were observed. These patterns bore little resemblance to the broad relatively smooth pattern of the average rainfall

over the grid during the period. The contrast is striking enough to overshadow differences which might be introduced through different interpolation weights or the catch deficiencies of raingauges.⁴

No distinction was made between types of precipitation in this data set. The distinction between rain and snow would have to be made from available temperature data. The difference between rain from stratiform or convective clouds might be deduced from digitized radar data. Whether the scavenging is taking place in or below the clouds is also important to the amount of removal. These are refinements, however, which may be incorporated when validation experiments indicate that they are needed.

* See "A Regional Scale Model for Computing Deposition and Ground Level Air Concentration of SO₂ and SO₄ from an Elevated Source," by L. L. Wendell, D. C. Powell, and R. L. Drake, this volume.

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EXAMINATION OF THE WIND SHEAR IN REGIONAL SCALE FLOW LAYERS DERIVED FROM RADIOSONDE DATA

L. L. Wendell and T. D. Fox

Recent regional scale atmospheric transport models have used average winds derived for layers near the earth's surface or standard pressure levels. Significant shear effects are evident when transport is calculated with 100 m-1000 m layer averaged winds or with winds at the 850 mb level. Shear effects are also investigated within the 100 m-1000 m layer.

INTRODUCTION

One of the fundamental considerations in an assessment model is the atmospheric transport. For regional-to large-scale considerations trajectory computations have been kinematic in nature based on measured wind data.^{1,2,3*} The two main sources of wind data are the analyzed northern hemispheric gridded data at the standard pressure levels produced by NOAA's Numerical Meteorological Center (NMC) and the basic radiosonde data which are used to produce the analyzed fields. The NMC analyses have been used in areas where much of the computational area is over the ocean and observations are very sparse.¹ The resolution of the data in these analyses is 381 km at 60° N,

and the first level of analyzed winds is 850 mb (about 1500 m). The radiosonde data have been used to compute layer average winds from about 100 m-1000 m at the measurement locations.² The layered average winds are used directly to compute the trajectories^{2,3} or to produce gridded winds for trajectory calculations.*

The wind shear which can occur in the lower layers of the atmosphere

* See "An Examination of the Effects of Real Time Versus Time Averaged Precipitation Data on SO₂ and SO₄ Removal in the Regional Assessment Model," by L. L. Wendell and D. C. Powell, this volume,

raises questions about transport accuracy as well as the diffusion simulation. The discrepancies which can arise when different trajectory methods are applied have been comprehensively reviewed by Danielsen.⁴ The trajectory methods used for long term assessments for practical reasons must compromise between sophistication and economy. This is the reason for the simple kinematic approach of most investigators. This report presents the results of a preliminary comparison of plume transport in different layers of the lower atmosphere.

DATA

The data used for this comparison were the gridded 850 mb winds of the NMC analyses and the layer averaged winds computed from radiosonde data. The layer averaged winds were interpolated to a grid with one half the NMC mesh size for the transport calculations.*

Single Level Winds Versus Layer Averaged Winds

The 850 mb winds represent the flow at about the 1500 m level and in general would be expected to exhibit different characteristics than winds close to the surface. In order to examine these differences in a way which is pertinent to atmospheric pollutant transport, a set of 12-hourly plume plots were generated from NMC 850 mb winds for comparison with a set of plots generated from 100 m-1000 m layer averaged winds for a release beginning 0000 GMT April 19, 1974. Both sets of plots are shown in Figure 1. As would be expected, the higher speeds at the 850 mb level are indicated by the longer plumes. The more regular character of the plumes carried by the 850 mb winds is probably a reflection of smoothing and filtering of the NMC analyses.

The most striking feature of this comparison is the indication of strong directional shear at several

of the release locations, particularly Chicago and St. Louis during the first 12 hr of the release. It may be seen in Figure 1 that the 850 mb plumes from these two locations affect almost completely different geographical areas than the plumes carried by the layer averaged winds.

Shear in Layer Averaged Winds

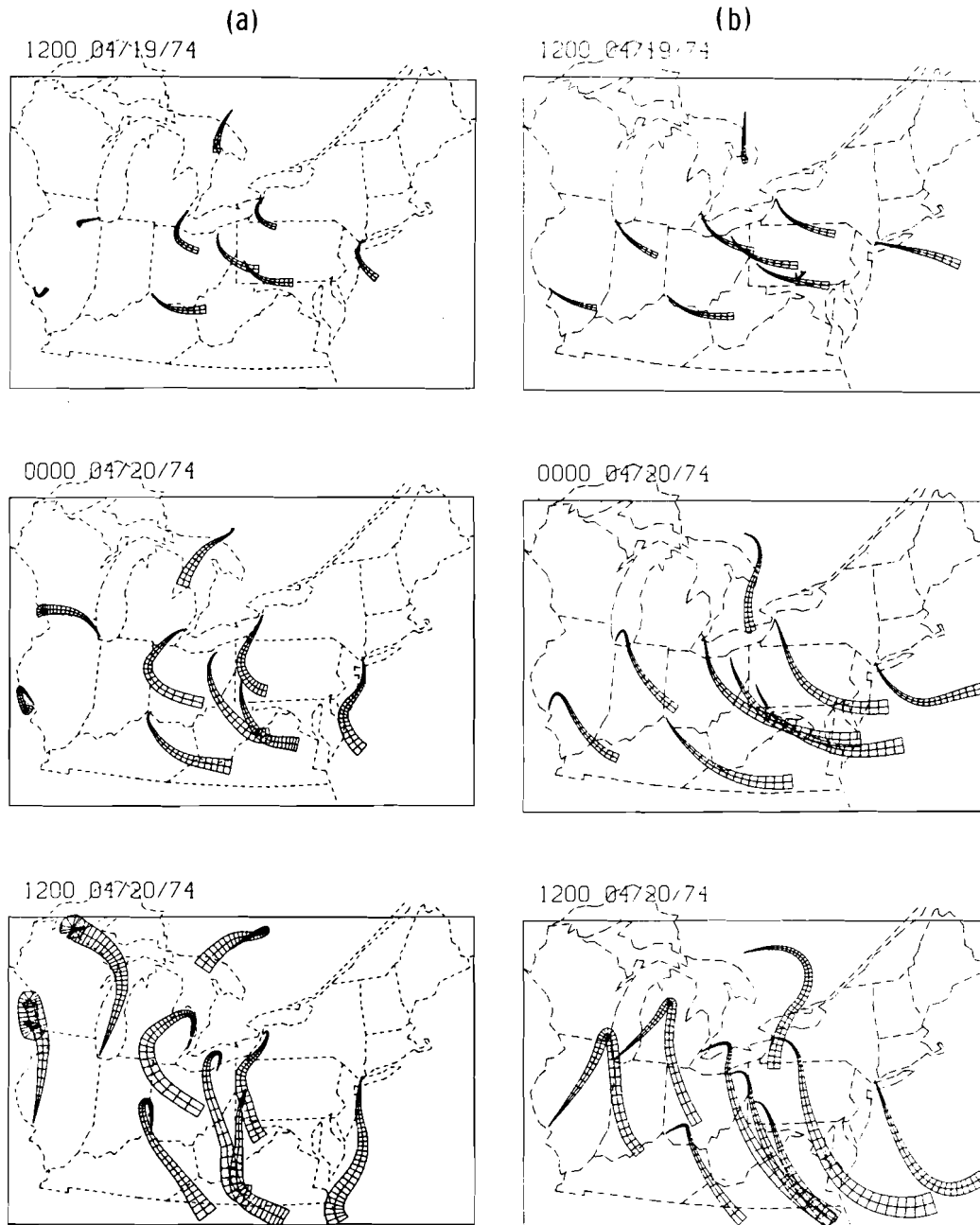
To investigate the significance of the shear in the flow in the lowest 1000 m layer, averaged winds were determined for the layer 100 m-500 m and the layer 500 m-1000 m at each sounding location. These winds were then gridded as shown in Figure 2. The main difference between wind fields for the two layers is that winds in the upper layer are stronger. There is some direction shear evident in Illinois at 1200 April 10, 1974. The grid spacing shown is about 170 km and is the same as that used in the plume transport calculations.

The 12-hourly plume plots for the 48 hr release are shown in Figure 3. The advection of the points defining the plume centerlines was carried out in the two layers as if the plumes were originating in the separate layers above each release point. No interaction between layers was allowed.

The effects of the speed and direction shear over a period of time are much easier to discern from the plume plots than from the wind field plots. During the first 12 hr of the release, beginning at 0000 GMT April 10, 1974, the resulting plumes in the two layers indicate direction shear in the western portion of the area, speed shear in the central portion and very little shear in the eastern portion. As time progresses the direction shear seems to propagate eastward. The synoptic weather maps for the period of consideration showed the computation area initially in the high pressure pattern between two frontal storms. Toward the end of the period the flow in the eastern portion of the area is reflecting the approach of the frontal storm from the west.

The shear, for the case indicated in these two layers, appears to be enough for the St. Louis and Cincinnati sources to cause significant differences in the areas exposed at

* See "A Regional Scale Model for Computing Deposition and Ground Level Air Concentration of SO₂ and SO from an Elevated Source," by L. L. Wendell, D. C. Powell and R. L. Drake, this volume.



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FIGURE 1. Plume Positions Shown at 12-Hourly Intervals for a Release Starting at 0000 GMT April 19, 1974 Computed with a) Layer Averaged Winds Between 100 and 1000 m and b) Winds at the 850 mb Surface

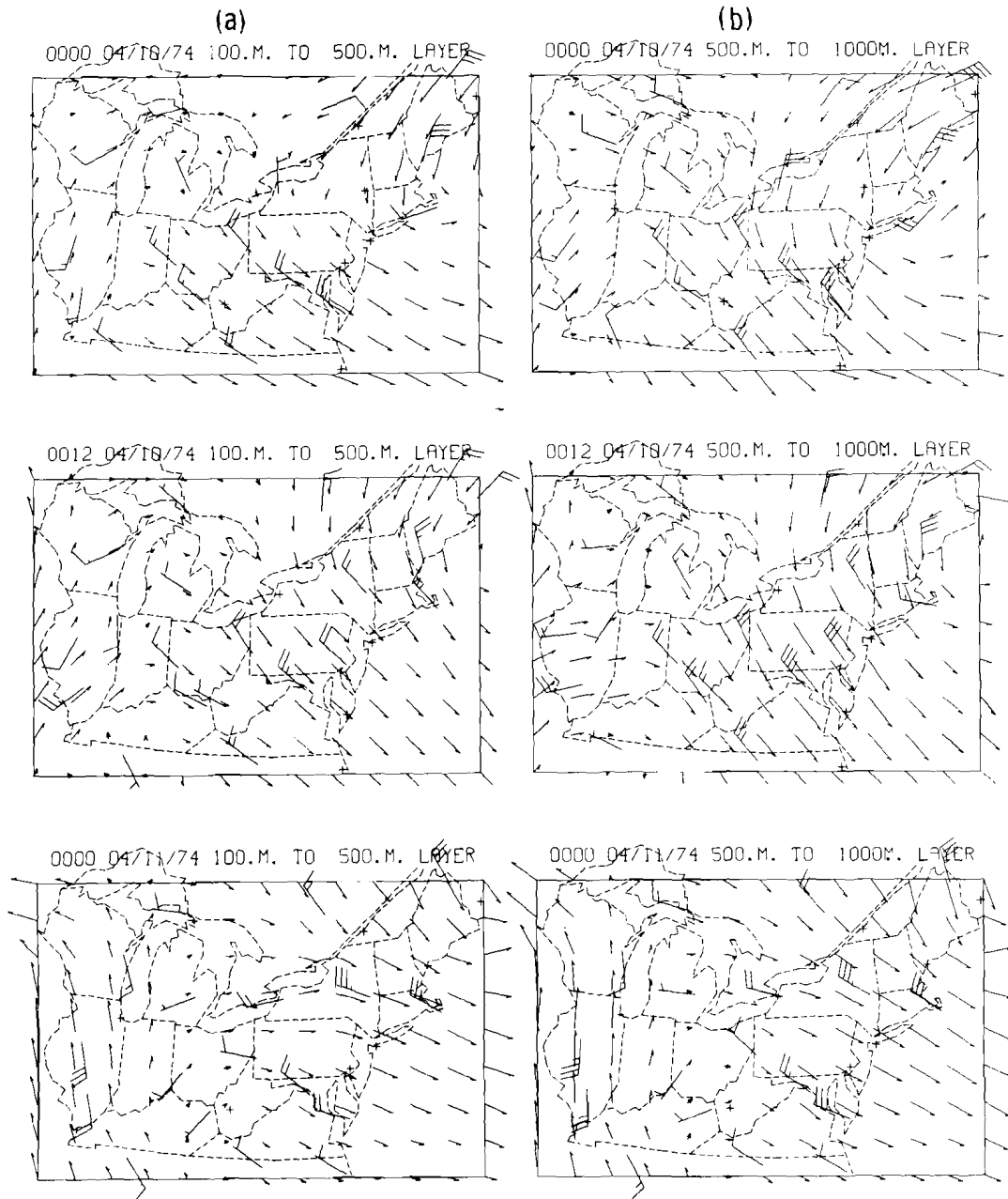


FIGURE 2. Wind Field Plots for Layer Averaged Winds in
a) the 100 m-500 m Layer and b) 500 m-1000 m Layer.

least for short term considerations. The deposition by precipitation scavenging could be quite different if real time precipitation data were used in the computations.

DISCUSSION AND RECOMMENDATIONS

The main implications from these few comparisons are that shear in the lowest 1500 m of the atmosphere is a factor which can strongly influence

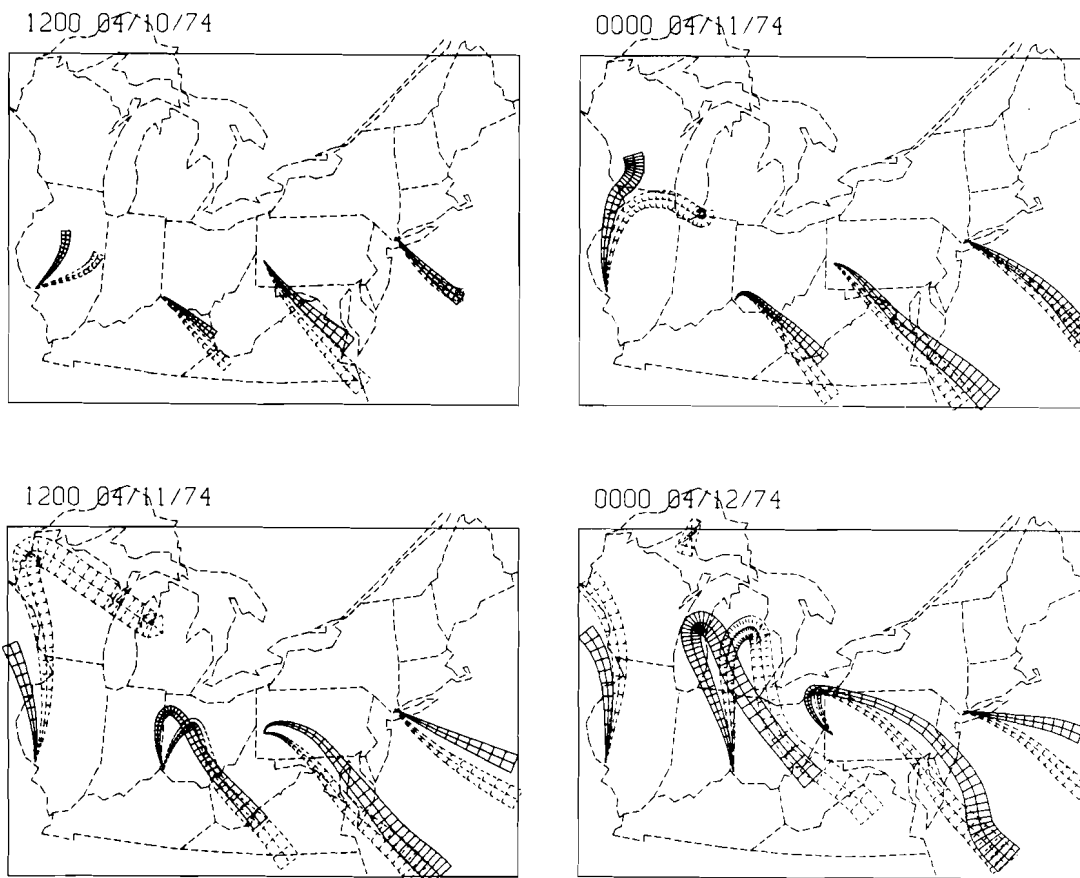


FIGURE 3. Plume Positions Shown at 12-hr Intervals for a Release Starting at 0000 GMT April 10, 1974. The solid plumes were generated with the 100-500 m layer winds. The dashed plumes were generated with the 500-1000 m winds.

the results of transport calculations. Selection of the transporting winds to be used should be carefully considered in terms of the application intended.

Other topics that must be addressed when considering the shear problem are vertical transfer of the contaminant by bulk vertical motion or by a diffusive process; the effect of these processes on the horizontal spread of the contaminant for the two

layers considered may be deduced by visualizing a plume which is some combination of the pairs shown in Figure 3. This points up the need for careful consideration of stability conditions and the energetics of the flow. To determine whether all this sophistication is necessary for long term assessments will require sensitivity testing in the models under development as well as some statistical considerations.

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CHEMICAL DEPOSITION AND TRANSFORMATION PARAMETERIZATIONS FOR REGIONAL MODELS

R. L. Drake

This report discusses the deposition and transformation components of the regional scale model being developed by PNL for the MAP3S program. The processes of chemical transformation and removal by dry deposition and precipitation scavenging are based on the total mass of an advecting, pseudo-Gaussian puff and not on the vertical concentration profile of the pollutant. The results from this formulation are compared with the source, surface and base depletion models of Horst.¹

INTRODUCTION

The overall description of a regional scale model for computing deposition and ground level air concentrations of SO₂ and SO₄ from elevated sources is given in Wendell et al.² This regional model will be an integral part of the MAP3S program, currently being planned and implemented by the national laboratories. The present report only concerns the parameterizations of the chemical transformations and removal processes in the regional model.

The current parameterizations are based on the total mass of an advecting, pseudo-Gaussian puff and not on

the more realistic concept of the vertical distribution of the pollutant. The application of this "integrated" approach may cause inaccurate transformation and deposition rates. For example, the dry deposition rate of both SO₂ and SO₄ may be too large since the usual deposition mechanism is based on the concentration of the pollutants at a level close to the ground ($Z = Z_d$) and not on the total mass of the pollutant in the vertical column. For this reason, we compare the current formulations for dry deposition with the more realistic surface depletion model of Horst.^{1,3} Hence, the outline of this report is: 1) the description of the time-evolution of the total mass of a puff of

pollutant (SO_2 plus SO_4) undergoing a transformation process ($\text{SO}_2 \rightarrow \text{SO}_4$) and experiencing losses due to dry deposition and precipitation scavenging; 2) descriptions of Horst's source, surface and base depletion models; 3) a comparison between the total mass and the Horst models; and 4) proposed corrections to the total mass model.

THE TOTAL MASS MODEL

In the total mass model we assume that a puff of pollutant is being advected along a path (of length s) under the influence of the mean wind $U(t)$.⁴ The origin of this path is at an elevated source of effective height h . The total mass of SO_2 in the advecting puff is given by $M = M(s, h)$ and the mass of SO_4 is $N = N(s, h)$. If we assume that the source location is defined by $(s, t) = (0, 0)$, then the relationship between t and s is given by

$$s = s(t) = \int_0^t U(\tau) d\tau. \quad (1)$$

As the puff is advected along, it is under the influence of a transformation from SO_2 to SO_4 and removal of SO_2 and SO_4 by dry deposition and precipitation scavenging. The dry deposition rates for SO_2 and SO_4 are by $\alpha_1 = \alpha_1(t)$ and $\alpha_2 = \alpha_2(t)$, respectively; similarly, the scavenging rates are $\beta_1 = \beta_1(t)$ and $\beta_2 = \beta_2(t)$; and the transformation rate is given by $\gamma_{12} = \gamma_{12}(t)$. Hence, the defining equations for the advecting pollutants are given by

$$\frac{dM}{dt} = - [\alpha_1 + \beta_1 + \gamma_{12}] M, \quad (2)$$

$$\frac{dN}{dt} = - [\alpha_2 + \beta_2] N + \frac{3}{2} \gamma_{12} M. \quad (3)$$

The $3/2$ in Equation (3) accounts for the differences in molecular weights between SO_2 and SO_4 .

Given the initial values

$$\begin{aligned} M_* &= M[s_*, h] = M[s(t_*), h], \\ N_* &= N[s_*, h] = N[s(t_*), h], \end{aligned} \quad (4)$$

we can easily solve Equations (2) and (3). The results are

$$M = M_* A(t), \quad (5)$$

$$N = \left[N_* + \frac{3}{2} M_* \int_{t_*}^t \frac{\gamma_{12}(\tau) A(\tau)}{B(\tau)} d\tau \right] B(t), \quad (6)$$

where

$$A(t) =$$

$$\exp \left[- \int_{t_*}^t \{ \alpha_1(\eta) + \beta_1(\eta) + \gamma_{12}(\eta) \} d\eta \right], \quad (7)$$

$$B(t) = \exp \left[- \int_{t_*}^t \{ \alpha_2(\eta) + \beta_2(\eta) \} d\eta \right] \quad (8)$$

In order to determine the total mass budget from s_* to s , we introduce the following notation: let the total dry deposition of SO_2 and SO_4 be denoted by D_1 and D_2 , respectively; denote the material scavenged by precipitation as W_1 and W_2 ; denote the amount of SO_2 transferred to SO_4 as T_1 ; and denote the amount of SO_4 formed as T_2 . Thus, given the quantities N and M in Equations (5) and (6), we can determine the following:

$$D_1 = \int_{t_*}^t \alpha_1(\eta) M d\eta, \quad D_2 = \int_{t_*}^t \alpha_2(\eta) N d\eta, \quad (9)$$

$$W_1 = \int_{t_*}^t \beta_1(\eta) M d\eta, \quad W_2 = \int_{t_*}^t \beta_2(\eta) N d\eta, \quad (10)$$

$$T_1 = \int_{t_*}^t \gamma_{12}(\eta) M d\eta, \quad T_2 = - \frac{3}{2} T_1, \quad (11)$$

where

$$D_1 + W_1 + T_1 = M_* - M, \quad (12a)$$

$$D_2 + W_2 + T_2 = N_* - N. \quad (12b)$$

In the MAP3S, regional model, the time interval $(t - t_*)$ is taken sufficiently small, usually 1 hr, so that that the parameters $\alpha_1, \alpha_2, \beta_1, \beta_2$ and γ_{12} can be replaced by constants. That is, the time-dependent parameters are assumed to be piecewise constant functions of t , where the step in the functions occur at every hour. The parameters α_i and $\beta_i, i = 1, 2$, are given in terms of other physical parameters, namely,

$$\alpha_i = \sqrt{\frac{2}{\pi}} \frac{V_{di}}{\sigma_z(s)} \exp \left[-\frac{1}{2} \left(\frac{h}{\sigma_z(s)} \right)^2 \right], \quad (13)$$

$$\beta_i = \frac{\Lambda(t) E_i}{d}, \quad (14)$$

where V_{di} is the deposition velocity (in m/hr), $\sigma_z(s)$ the vertical dispersion coefficient (in m), $\Lambda(t)$ the rainfall rate (in m/hr), E_i the collection efficiency, and d is a mean raindrop diameter (in m). Representative values of some of these parameters are $E_1 = 1$, $E_2 = 1/10$, $V_{d1} = 36$ m/hr, $V_{d2} = 4$ m/hr, and $\gamma_{12} = 0.05$ hr⁻¹; and the value for d lies in the interval from 0.002 m to 0.004 m. In addition, the rainfall rates are obtained from meteorological records and the $\sigma_z(s)$'s are obtained from a set of standard-type formulas for the various Pasquill stability classes.

THE HORST MODELS

The Horst models² are concerned with a single specie (say SO₂) puff evolving in time under the influences of the mean wind $U(t)$, turbulent diffusion and dry deposition. In all of these models a central role is played by a diffusion function defined by

$$D(s, z, h) =$$

$$\frac{\exp \left[-\frac{1}{2} \left(\frac{z - h}{\sigma_z(s)} \right)^2 \right] + \exp \left[-\frac{1}{2} \left(\frac{z + h}{\sigma_z(s)} \right)^2 \right]}{\sqrt{2\pi} \sigma_z(s)}. \quad (15)$$

This function can be interpreted in the following way: $Q_0 D(s, z, h)$, $Q_0 = \text{con-}$

stant, is the integrated concentration of SO₂ in an advecting puff, integrated with respect to the horizontal plane. The total mass of the puff is Q_0 and the puff was released from an elevated source of effective height h at time $t = 0$ (or $s = 0$). The boundary $z = 0$ is a reflective boundary and thus, no deposition occurs. The following result is consistent with the above statements:

$$\int_0^\infty D(s, z, h) dz = 1. \quad (16)$$

The usual expression for the deposition flux of a gas or particles to the surface, $z = 0$, is given by

$$F_d = V_d C(s, z_d, h), \quad (17)$$

where $C(s, z_d, h)$ is the integrated concentration of the puff of the pollutant evaluated at a level close to the surface, $z = z_d$.

Using this relationship, we can express the total mass of the advecting puff of SO₂ as follows:

$$M(s, h) = Q_0 - \int_0^s \frac{V_{d1}}{U} C(\xi, z_d, h) d\xi, \quad (18)$$

where the original mass of the puff was Q_0 . The Horst models are concerned with the approximation or evaluation of the quantity $C(\xi, z_d, h)$ in Equation (18).

The most elementary model is the source depletion model. This model approximates the integrand in (18) by

$$C(\xi, z_d, h) = M(\xi, h) D(\xi, z_d, h). \quad (19)$$

Substituting (19) into (18) and then differentiating with respect to s , results in the following approximation for $M(s, h)$:

$$M(s, h) = Q_0 \exp \left[- \int_0^s D(\xi) d\xi \right], \quad (20)$$

where

$$E(\xi) = \frac{V_{d1}}{U} D(\xi, z_d, h). \quad (21)$$

The most physically realistic model is the surface depletion model. In this case, the integrand in (18) is obtained from the following integral equation:

$$C(s, Z_d, h) = Q_0 D(s, Z_d, h) - \int_0^s \frac{V_{d1}}{U} C(\xi, Z_d, h) D(s - \xi, Z_d, Z_d) d\xi. \quad (22)$$

Equation (22) is solved by Laplace transforms and the solution is substituted into (18) giving the following:

$$M(s, h) = Q_0 \left[1 - \int_0^s \left\{ E(\xi) + \sum_{n=1}^{\infty} (-1)^n E(\xi) * \prod_{\ell=1}^n F^\ell(\xi) \right\} d\xi \right], \quad (23)$$

where "*" is the convolution operator and

$$F(\xi) = \frac{V_{d1}}{U} D(\xi, Z_d, Z_d)$$

$$\prod_{\ell=1}^n F^\ell(\xi) = F * F * \dots * F. \quad (24)$$

Horst describes the physical significance of these two models.² Since the surface depletion model takes into account the concentration of SO₂ at Z = Z_d, it is superior to the source depletion model. However, the computation effort involved in evaluating the multiple convolution integrals in (23) severely limits the application of (23) in the regional MAP3S modeling program. In fact, if the "strict" surface depletion model is going to be applied, the integral equation in (22) should be used in place of (23).

Horst's third model, called the base depletion model, "falls" somewhere in between the surface and

source model. It is obtained by replacing the (s - ξ) argument in the integral in (22) by just the argument (s). This eliminates the "convolution" character of Equation (22) and allows us to write M(s, h) as

$$M(s, h) = Q_0 \left[1 - \int_0^s \left\{ E(\xi) - F(\xi) \int_0^\xi E(\eta) \exp \left[- \int_\eta^\xi F(\alpha) d\alpha \right] d\eta \right\} d\xi \right]. \quad (25)$$

Comparing Equation (25) with Equations (20) and (23) shows that the base model is somewhat more complex than the source model, and considerably simpler than the surface model with reference to numerical computation.

TOTAL MASS MODEL VERSUS THE HORST MODELS

For comparative purposes, let θ₁ = γ₁₂ = t_{*} = 0 and M_{*} = Q₀ in Equations (5) and (7); hence, the total mass model becomes

$$M(s, h) = Q_0 \left[\exp - \int_0^s \frac{1}{U} \alpha_1(\xi/U) d\xi \right]. \quad (26)$$

Comparing Equation (26) with Equation (20) we see that the total mass model is identical to the source model if

$$\alpha_1(\xi/U) = V_{d1} D(\xi, Z_d, h). \quad (27)$$

Equation (27) is identical to (13) if Z_d = 0 and i = 1. Since Z_d ≅ 1m and Z_d << σ_z, then it is reasonable to replace (27) by (13).

Looking at Equations (23), (25) and (20), or (26), we see that all three of the Horst models have the same "leading terms" in a series expansion, namely,

$$M(s, h) = Q_0 \left[1 - \int_0^s E(\xi) d\xi + \dots \right]. \quad (28)$$

However, beyond the first two terms, the comparison of the three "series" becomes difficult. Horst² compares the source model with the surface model for a pseudo-Gaussian plume under various stability conditions and for various values of h and V_{d1}/U . Rather than repeating Horst's computations for the puff, we will compare the three formulas for certain simplifying assumptions.

In Equations (20), (23) and (25), let $Z_d = 0$, $U = \text{constant}$, and $\sigma_z = \text{constant}$. Thus, Equation (20) reduces to

$$M(s, h) = Q_0 \exp[-Es]; \quad (29)$$

and Equations (23) and (25) reduce to

$$M(s, h) = Q_0 \left[1 - \frac{E}{F} + \frac{E}{F} \exp(-Fs) \right], \quad (30)$$

where

$$E = F \exp \left[-\frac{1}{2} \left(\frac{h}{\sigma_z} \right)^2 \right], \quad (31)$$

$$F = \sqrt{\frac{2}{\pi}} \frac{V_{d1}}{U \sigma_z}. \quad (32)$$

Looking at (30), we note that the base and surface models give identical results. This is expected since the approximation used to get (25) is trivially true when $D(s, z, h)$ is a constant function of s . When $h = 0$, all three models give the same result, namely

$$M(s, 0) = Q_0 \exp[-Fs]. \quad (33)$$

If $Z_d = 0$ and $h = 0$, then $E(\xi) = F(\xi)$ and (25) reduces to (20). That is the reason that Horst calls (25) the base depletion model since it is equivalent to the source model for $h = 0$. That is, the material is "depleted from the base of the stack." The base model is equal to the surface model for constant σ_z and U , and is equal to the source model for $h = 0$. Thus, the base model is "in between" the source and surface models.

A CORRECTION TO THE TOTAL MASS MODEL

In the MAP3S regional model the usual time increment is 1 hr ($\Delta t = 1$ hr). During any given Δt , it is assumed that σ_z and U are both constant. Thus, the mass of an evolving puff of SO_2 under the influence of dry deposition is given by (20):

$$M(s_{i+1}, h) = M(s_i, h) \exp \left[-E_{i+1/2}(s_{i+1} - s_i) \right] \quad (34)$$

where the intervals $s_0 = 0$ to s_1 , s_1 to $s_2, \dots, s_i$ to $s_{i+1}, \dots$ correspond to the time intervals 0 to Δt , Δt to $2\Delta t, \dots, i\Delta t$ to $(i+1)\Delta t, \dots$. The quantity $E_{i+1/2}$ is equal to $E(s)$ evaluated at the midpoint of the $(s_{i+1} - s_i)$ interval; and $M(s_0, h) = M(0, h) = Q_0$. Equation (34) is based on the source model

For most of the Pasquill stability classes, we can replace (34) by

$$\begin{aligned} M(s_{i+1}, h) &= M(s_i, h) \left[1 - E_{i+1/2}(s_{i+1} - s_i) \right. \\ &\quad \left. + 1/2 E_{i+1/2}^2 (s_{i+1} - s_i)^2 \right]. \end{aligned} \quad (35)$$

If more accuracy is required, we can use more terms in the series expansion.

An analogous type approximation for (23) is impossible because of the multiple convolution integrals. However, we can approximate (25) in the following way:

$$\begin{aligned} M(s_{i+1}, h) &= M(s_i, h) \\ &- Q_0 G_{i+1/2}(s_{i+1} - s_i), \end{aligned} \quad (36)$$

where

$$\begin{aligned} G_{i+1/2} &= E_{i+1/2} - F_{i+1/2} \\ \exp \left[-\int_0^{s_{i+1/2}} F(\alpha) d\alpha \right] &\int_0^{s_{i+1/2}} H(\eta) d\eta. \end{aligned} \quad (37)$$

The quantity $H(\eta)$ is given by

$$H(\eta) = E(\eta) \exp \left[\int_0^\eta F(\alpha) d\alpha \right]. \quad (38)$$

The integrals in (37) can be approximated by

$$\begin{aligned} \int_0^{s_{i+1/2}} A(\eta) d\eta &= A_{1/2}(s_1) + \\ &A_{3/2}(s_2 - s_1) + \dots + A_{i-1/2}(s_i - s_{i-1}) \\ &+ A_{i+1/4}(s_{i+1/2} - s_i) \end{aligned} \quad (39)$$

where $A(\eta)$ is equal to $F(\eta)$ or $H(\eta)$. The integral in (38) is approximated in a similar fashion.

Thus, one improvement of the total mass model is to use the base model given by (36) to (39). This set of equations requires more computation and memory than (34) or (35) but it is more realistic. If the surface model is desired, we recommend the use of the integral equation in (22).

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REGIONAL AIR QUALITY ASSESSMENT FOR
NORTHWEST ENERGY DEVELOPMENT SCENARIOS

D^W. S. Renne and D^W. L. Elliott
375-6161 375-6161

Using a regional scale transport and dispersion model, work has begun on an assessment of the impacts on air quality in the Northwest from alternative energy development scenarios. Dry deposition, precipitation scavenging, and chemical transformation are incorporated into the model. A preliminary investigation of regional transport, based on winds averaged through the 100 m - 1000 m layer, has been made for several large coal-fired power plants.

INTRODUCTION

The Atmospheric Sciences Department is engaged in a regional air quality assessment of the Pacific Northwest in conjunction with DBER's Integrated Assessment Program. Our objective is to estimate the air quality in the Pacific Northwest, and provide information needed to assess the effects on terrestrial environments caused by deposition and wash-out of airborne pollutants, from development of the region's energy resources. A thorough assessment of these impacts requires detailed knowledge of the strength and geographic distribution of present and future energy-related emissions throughout the region (including emissions from energy facilities themselves, as well as secondary emissions resulting from the community and transportation growth that will inevitably occur as resources are developed) and knowledge of the atmospheric transport and diffusion properties throughout the region.

EMISSIONS

The energy-related pollutant emissions will be obtained as part of the output of the energy/economic sectors of a regional model being developed in the Regional Studies Program. Emission factors obtained from the literature^{1,2} will be incorporated into this model to convert the amount

of fuel burned or converted in various categories (such as electrical or coal conversion, transportation and space heating) and the amount of particulates released by process industries and agricultural activities, into total emissions of sulfur and nitrogen oxides, particulates, trace elements, carbon monoxide, and hydrocarbons. The emission inventory can be divided into two basic categories: 1) primary or point source emissions from large fuel burning or conversion facilities such as power plants and gasification plants, and 2) secondary or area source emissions from space heating, transportation and related industries.

TRANSPORT AND DIFFUSION MODEL

A regional scale transport and diffusion model which accounts for the spatial and temporal variations in the wind field^{3,4} is being used to estimate ground level air quality impacts of present and future energy development activities. This model interpolates the mean winds between 100 m and 1000 m above the ground, measured every 12 hr at approximately 34 sites in or adjacent to the area of the Pacific Northwest covered in the Regional Studies Program (Figure 1). Trajectories of pollutant transport are then determined from these wind fields. Standard diffusion relations are used to estimate

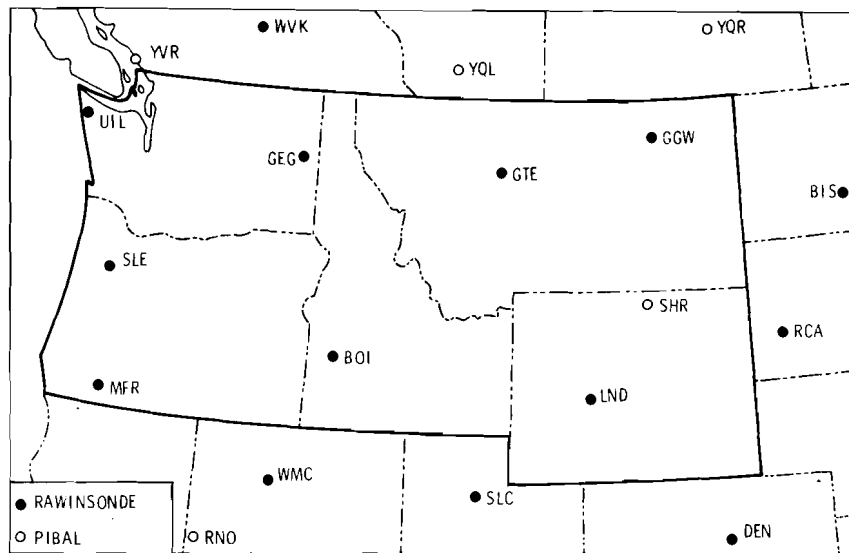


FIGURE 1. Pacific Northwest Meteorological Stations Utilized in the Interpolated Wind Fields. Additional stations off the grid used for interpolating winds near the grid boundaries.

plume dispersion. Removal processes such as dry deposition, precipitation scavenging, and chemical transformations are incorporated into the model.

For incorporation into the Pacific Northwest Regional Assessment model, calculations will be made from a long term set of meteorological data to develop annual average air quality concentrations, on a grid of receptor points normalized to the composite emissions occurring throughout the region. This matrix of normalized concentrations can then be coupled to the other sectors of the Regional Assessment model to calculate air quality and deposition processes for exogenously specified scenarios. The rate and cumulative amount of surface-deposited pollutants will be made available to the bioproductivity sector for computation of effects over time.

PROGRAM STATUS

A preliminary investigation of transport has been made for the six large existing or proposed coal-fired power plants listed in Table 1. Figure 2 shows the plume trajectories for these plants for a 4-day period

TABLE 1. Coal-Fired Power Plants for which Plume Trajectories were Computed

Name	Location	Megawatts
Centralia	SW Washington	1400 Existing
Boardman	NE Oregon	500+ Proposed
Pioneer	SW Idaho	1000 Proposed
Jim Bridger	SW Wyoming	2000 Existing
Wheatland	SE Wyoming	1500 Proposed
Colstrip	SE Montana	2060 Proposed

in April 1974 computed from the trajectory model. The existing wind data are influenced, to some extent, by large topographic features such as the Rocky Mountains. Analytic methods are being considered to incorporate the effect of topographic features such as the Cascade Mountains, Columbia Basin, and Willamette Valley.

Preliminary computations of ground level concentration, deposition, and scavenging of sulfur dioxides and sulfates from large coal-fired power plants in the Northwest have also been made. Precipitation scavenging

is computed using mean monthly precipitation data extrapolated to a fine-mesh grid. Eventually, estimates of annual average ground level concentrations and surface deposition

of sulfur dioxides and sulfates will be made on a grid with roughly 2000 "receptor points" spaced approximately 38 km apart.

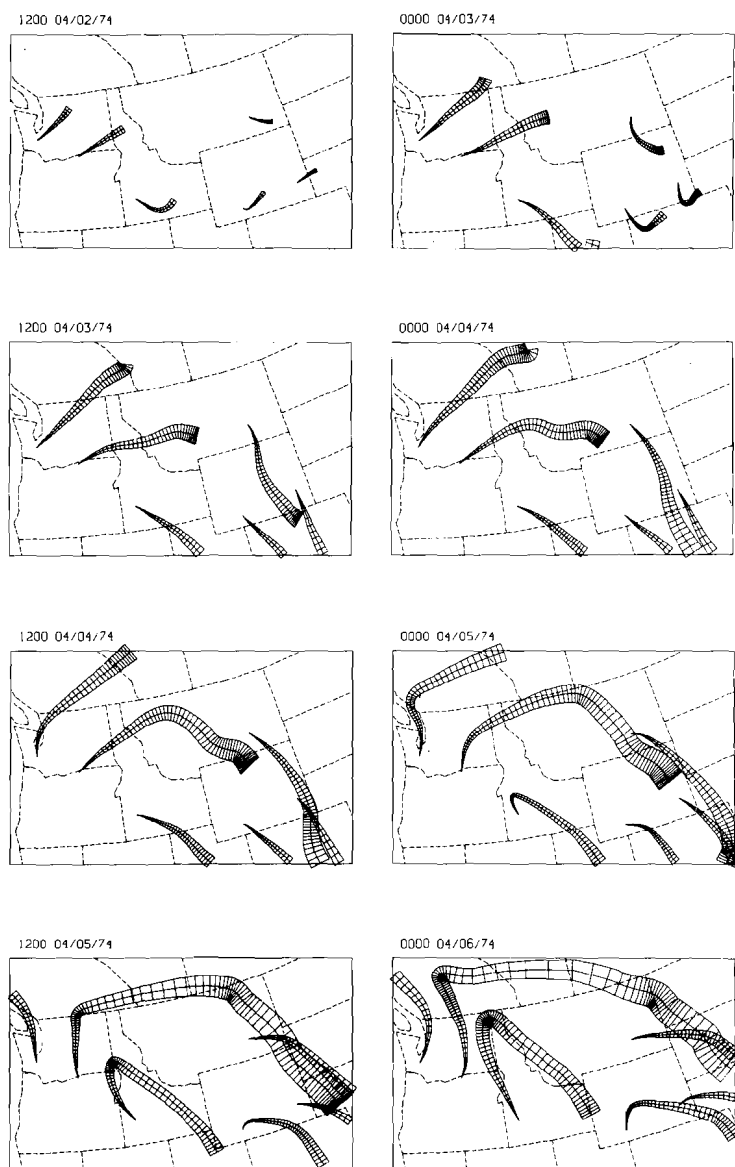


FIGURE 2. Plume Trajectories for Six Existing and Proposed Coal-Fired Power Plants. Trajectory calculations begin at 0000 GMT on April 2, 1974.

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AN INVESTIGATION OF REGIONAL FLOW PATTERNS OVER AN AREA OF COMPLEX TERRAIN

D. L. Elliott, W. F. Sandusky and R. K. Woodruff

Regional wind flow patterns and transport in the lower planetary boundary layer are examined from a network of 29 wind recording stations for a 170 by 190 km area over the Mid-Columbia region of Washington and Oregon. From an investigation of the nonfrontal situations, several different types of recurring flow patterns were observed, and each flow type appeared to be characterized by similar synoptic (surface) pressure patterns and weather conditions. Techniques for improving estimates of regional transport, through an improved data acquisition system with an extended data network, a classification of flow patterns, and the utilization and modification of existing models, are examined.

INTRODUCTION

The proper assessment and control of environmental impacts to a region from atmospheric releases (whether from single or multiple sources) requires a detailed meteorological

knowledge of the region and a framework within which this knowledge can be most effectively used. As emphasis on environmental and health protection, nuclear safety, and competition for land use and industrial development have increased,

it has become increasingly necessary to adapt and utilize current methods and techniques to assess the environmental impacts of existing and proposed developments.

Previous studies have shown the importance of considering spatial and temporal variations in the wind field in estimating mesoscale transport and diffusion.^{1,2,3} Case studies of wind-flow patterns and transport over an area which included the Hanford Reservation and immediate vicinity have previously been examined. However, this scale needed to be extended to examine the environmental impacts on the surrounding region.

The primary objectives of this study were: 1) to better understand the regional flow patterns and their relationship with synoptic weather situations, and 2) to examine techniques for improving regional transport and dose estimates for emergency response and routine assessment. Wind data were obtained from surface stations throughout the region and an analysis was performed for November 1974 data. For nonfrontal situations, an effort was made to classify wind-flow patterns over the region with synoptic-scale features, i.e., synoptic pressure patterns. This information would be very useful for both operational forecasting and assessment purposes.

The report given here is primarily a summary of a more complete and inclusive report.⁴

DESCRIPTION OF THE AREA

The study area, shown in Figure 1, is centered over the Mid-Columbia region in southeastern Washington. This region includes the Hanford Reservation. Major variations in the smoothed topography can be seen in Figure 2.

The wind data network consisted of 29 stations scattered throughout the region, with the most dense coverage in the vicinity of the Hanford Reservation. All but 3 of the 29 stations normally had hourly wind data for 24 hr, recorded on strip charts, observed, or telemetered. Heights of the wind sensors above the ground ranged from 10 ft to

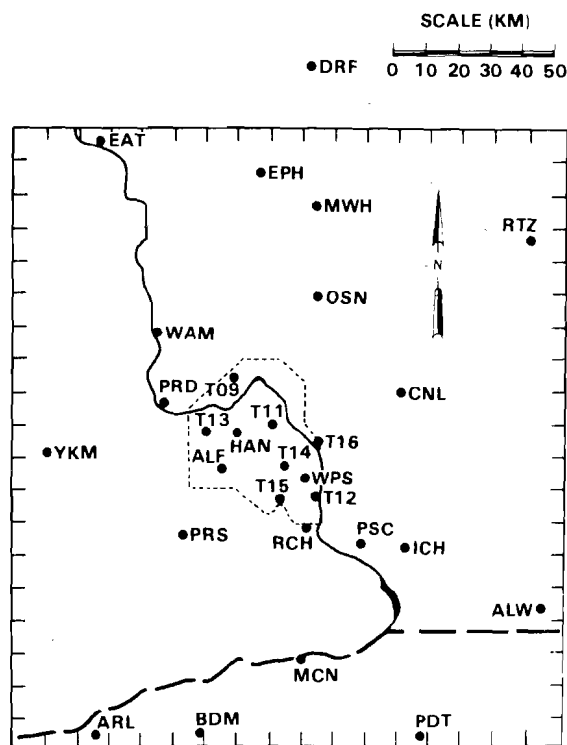


FIGURE 1. Map of the Mid-Columbia Region and Wind Recording Stations

50 ft, with the mean height at approximately 30 ft. Much of the data was obtained through the cooperation of local agencies specifically for this study.

REGIONAL WIND-FLOW PATTERNS

An objective interpolation scheme, developed by Wendell,¹ was used to obtain wind components on a regularly spaced grid from the irregularly spaced wind stations. These techniques were adapted to the Mid-Columbia region for producing wind field plots for selected periods in November 1974.

From an analysis of these plots, several recurring flow patterns were noted. Excluding days (or periods) when frontal systems were passing through or lying within the region, the flow patterns have been categorized into several main types. In all, these flow types accounted for the prevailing flow patterns observed on 19 days of the month. The results

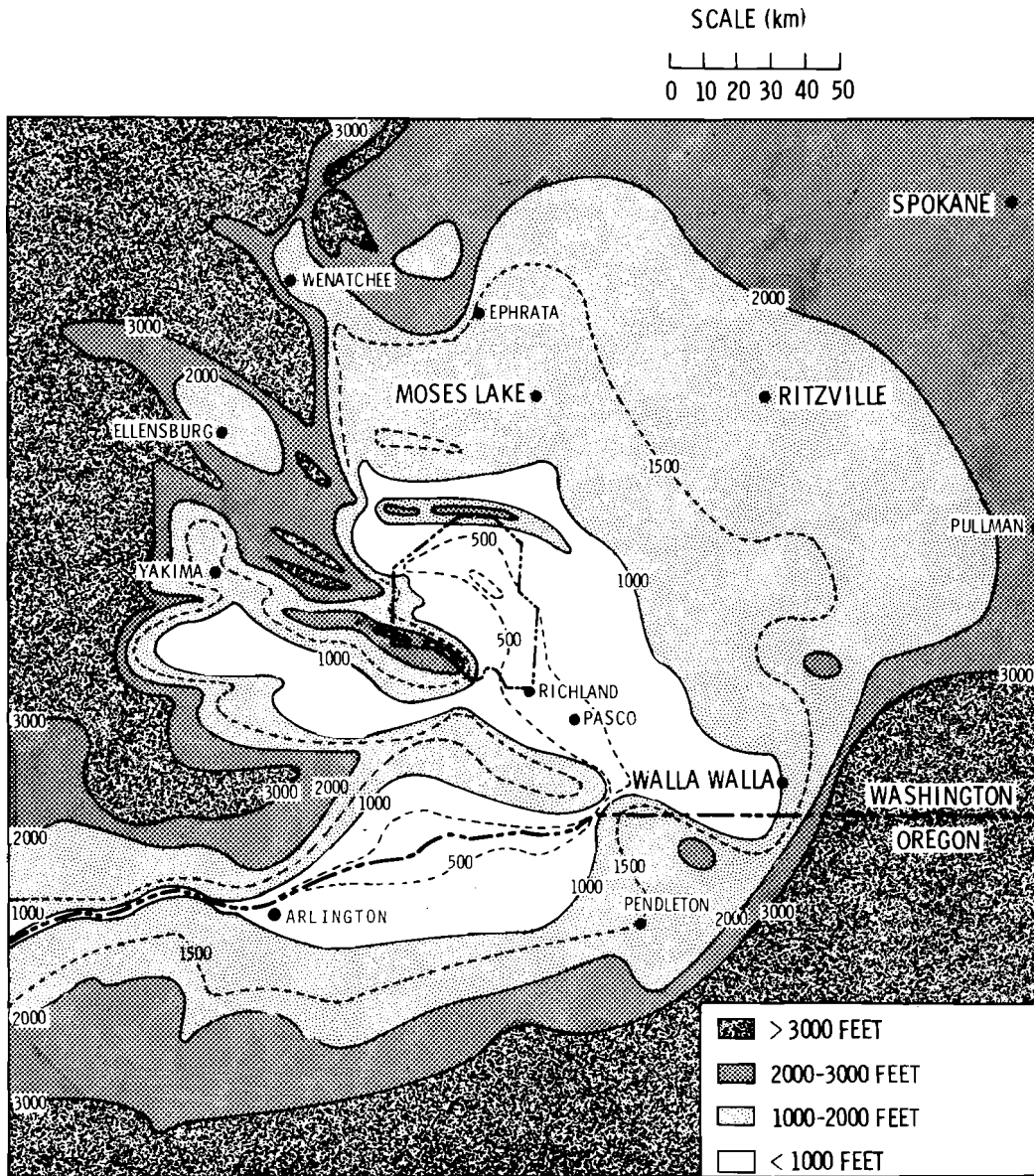


FIGURE 2. Contour Map of the Smoothed Topography

obtained demonstrate the relationships between the wind flow patterns over the region, topographic features, and synoptic weather situations. Some of these flow patterns are quite different from that expected for the existing large-scale meteorological conditions.

An important example of this is the flow pattern shown in Figure 3, which emphasizes the effect of con-

trasting transport regimes throughout the region; i.e., NW flow over Pasco, and E flow over Pullman and Arlington. High pressure centered to the east and/or northeast of the region is normally associated with this flow regime. Figure 4 shows the surface pressure analysis for November 11, 1974. Pressure gradients over the study area were primarily north-south. This type of flow pattern persisted for approximately 20 hr, with only

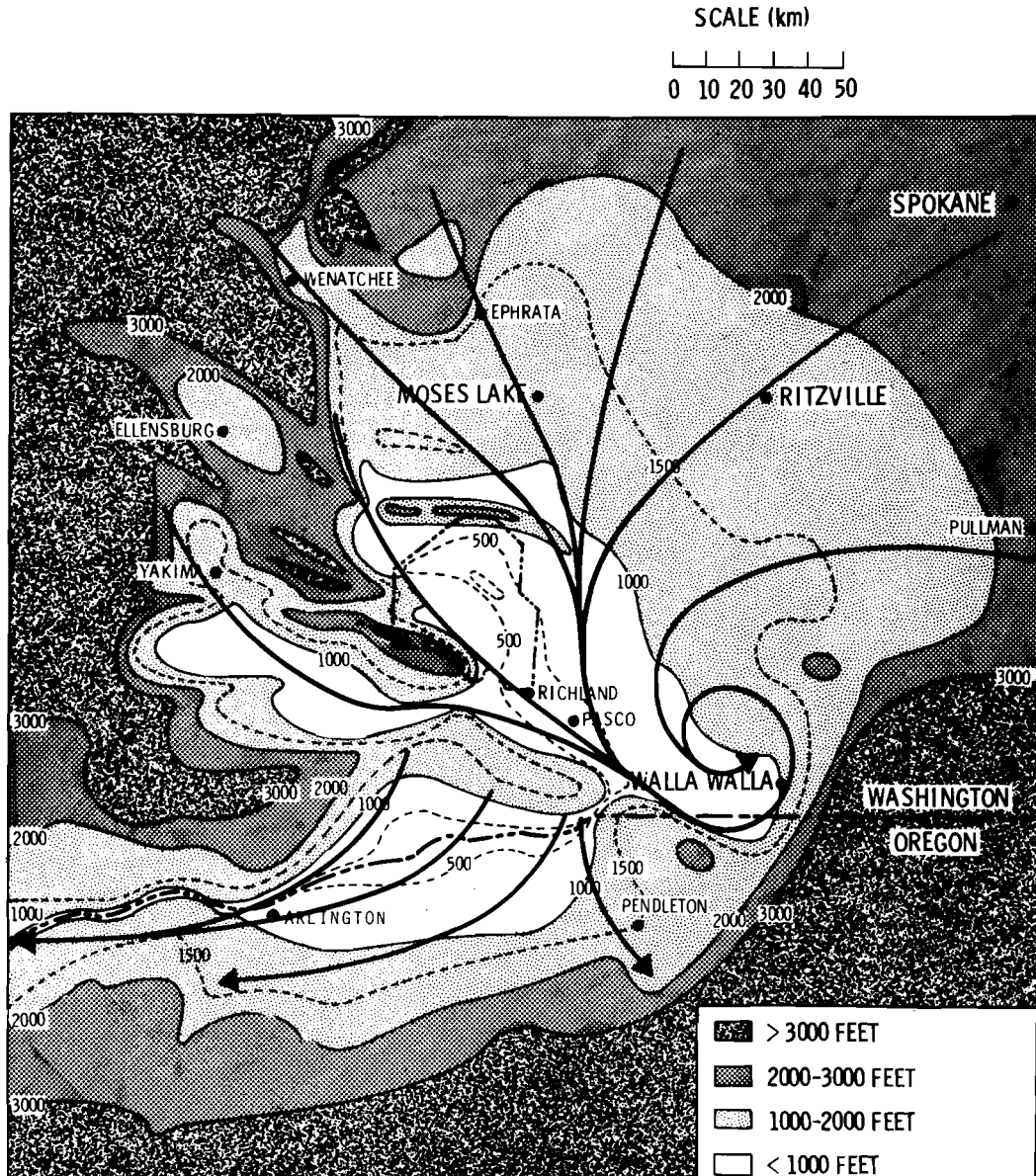


FIGURE 3. Schematic Flow Pattern Associated with a Surface High Centered East of the Region (see Figure 4)

minor deviations in the overall pattern. Mostly cloudy skies and stable conditions prevailed. The vertical profile of wind at 1300 PST, November 11, 1974 over Hanford showed NW winds up to about 700 m, switching to SW aloft. Thus the surface pressure gradient force appears to be the primary reason for the northerly component of wind over much of

the Mid-Columbia region, as upper level winds were southerly over the entire region. The variation in direction over the region can be explained, to a large extent, by topography effects. Knowledge of this regional flow variability and the large-scale meteorological situations for which it occurs, would provide substantially different and improved

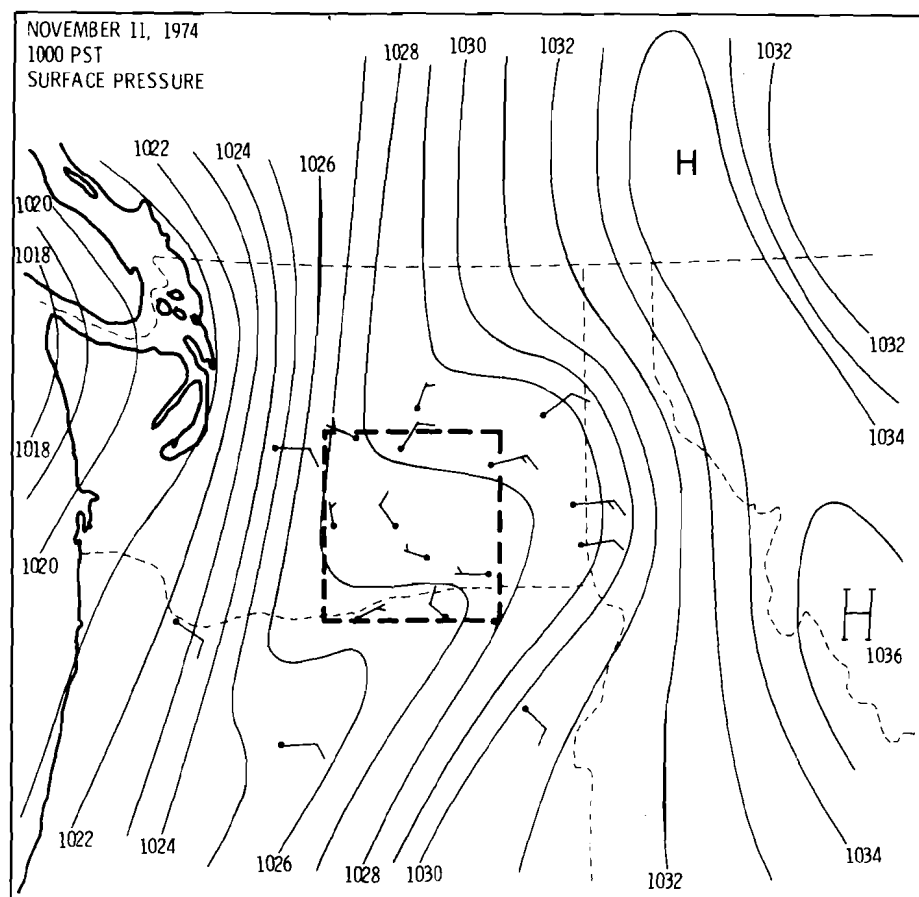


FIGURE 4. Detailed Surface Pressure Analysis for 1000 PST, November 11, 1974. The dashed box is the Mid-Columbia region.

emergency preparedness information from that available from local site data and large scale weather charts.

This type of flow pattern occurred on 4 days in November, each characterized by very similar surface pressure patterns and stable atmospheric conditions. The depth of the boundary layer flow ranged from about 500 to 1000 m over Hanford. Assuming this depth exists over the entire region, most of the effluent emissions (buoyant and nonbuoyant) would be transported within this layer.

Figure 5 demonstrates the flow pattern typically associated with high pressure west and low pressure east of the region. Surface isobars were aligned generally NW-SE, a condition which often follows the pas-

sage of a low pressure system. A description of some of the other regional flow patterns which occurred is given by Elliott, et al.⁴

CONCLUSIONS AND RECOMMENDATIONS

An investigation of the nonfrontal cases for November 1974 has revealed quite complex, recurring flow patterns over the Mid-Columbia region. There is evidence that these flow types could be classified, to some extent, by synoptic pressure patterns and atmospheric conditions. Background information of this nature would be useful in a quasi-predictive mode for the evaluation of regional transport. However, from this limited study, one should not infer that the regional wind flow pattern can be

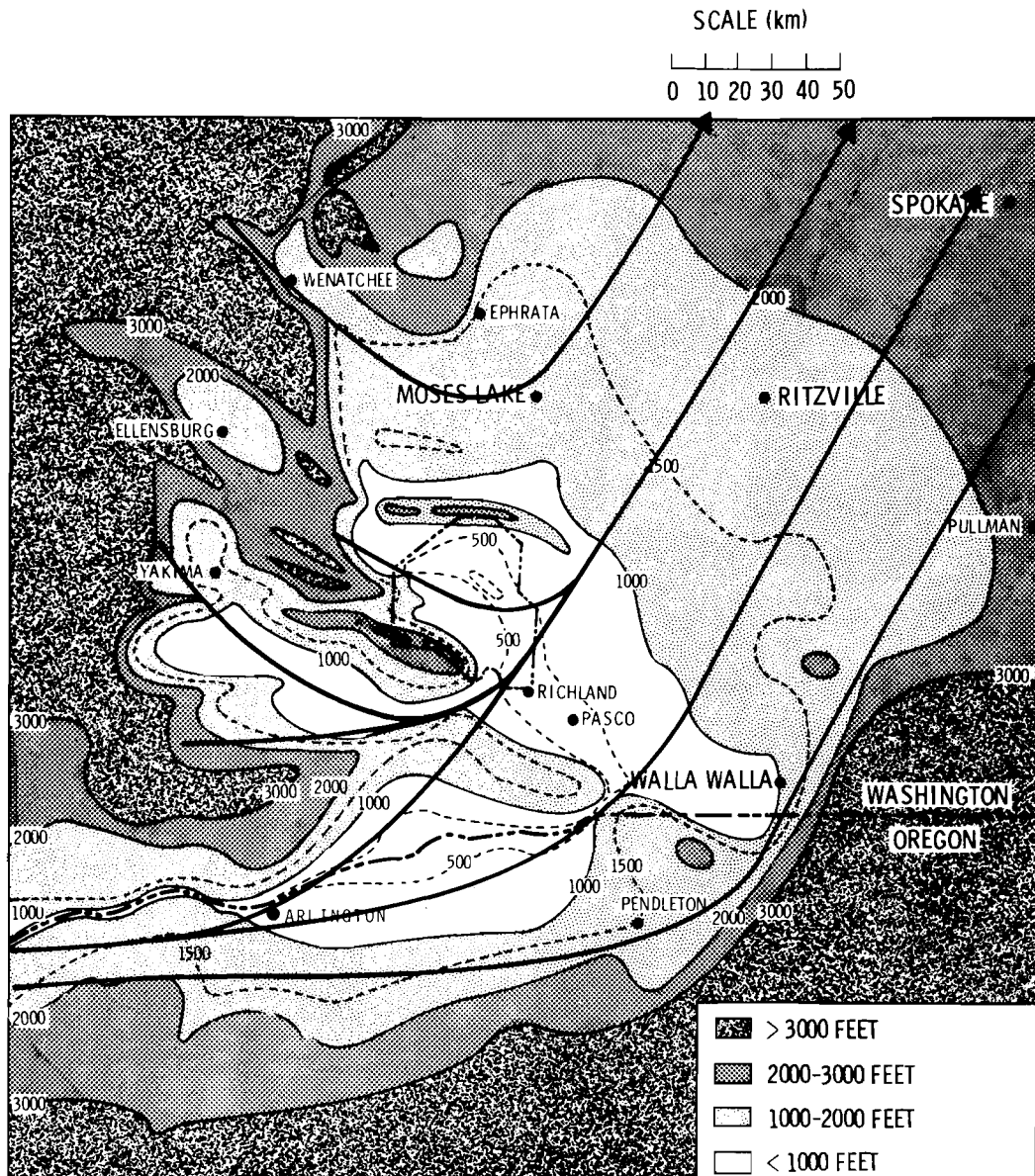


FIGURE 5. Schematic Flow Pattern Typically Associated with High Pressure to the West and Low Pressure to the East of the Region

deduced entirely from a given synoptic pressure pattern and local area wind data. The establishment of a few regional stations, in selected locations with data transmissions available for real-time use, is essential in the validation of an expected flow regime.

Through the adaptation and modification of regional transport and dif-

fusion models,^{5,6} an improved data acquisition system with an extended data network, and a classification of flow patterns, techniques exist for substantially improving estimates of transport and diffusion over the Mid-Columbia region. They could be used for real-time analysis and prediction as well as for assessing the individual or integrated impact of current or future operations in the

region. Applications could be extended to cover many other facets including pesticide and herbicide drift, safety analysis, and range fire control.

Future efforts should include:

1. Further study at the regional flow patterns to examine seasonal variations and frequency of occurrence of various flow types
2. Experiments to validate the regional model
3. Continued and accelerated refurbishment of the existing on-plant data telemetry system, and improvement in the availability of offsite data.

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WEATHER AND CLIMATE MODIFICATION

Human activity can change the average atmospheric temperature and humidity values, as well as the chemical composition of the air. These changes affect local and regional weather and climate, and may have a significant influence on global climate. Examples of human activity that produce these changes are the increase of CO₂ content of the atmosphere from the combustion of fossil fuel; the production of aerosols by industry, automobiles, home heating units and agricultural practices; the releases of large quantities of heat and water vapor from the cooling facilities of large fossil and nuclear power plants; and the modification of the earth's albedo due to urbanization, agriculture, deforestation, reservoirs and soil spills.

The Atmospheric Sciences Department has undertaken a modest effort in this area during the past year. The research activities have resulted in the following findings:

- An overview of the important natural and anthropogenic perturbations of weather and climate, and the resulting need for further climatic research;
- A theoretical study that may contribute to a better understanding of atmospheric electricity;
- Another year of atmospheric turbidity data at the Hanford site and their relationships to changes in the aerosol size distribution;
- Initial efforts in determining the optical properties of aerosols such that we can better understand the radiative properties of the atmosphere; and
- The characterization of large power plant cooling tower plumes through the use of long exposure photography and instrumented aircraft.

- **AEROSOL AND TRACE GAS TRANSFORMATIONS**

- **COOLING TOWER AND COOLING POND ATMOSPHERIC IMPACT**

- **ERDA/RL SPECIAL STUDIES**

CLIMATE AND WEATHER:
NATURAL AND ANTHROPOGENIC PERTURBATIONS

R. L. Drake

This report is concerned with perturbations and variations in weather and climate. The report discusses the definition of climate, why climatic studies are important, the natural variations of weather and climate, inadvertent and advertent climatic modifications due to humans and the need for further climatic research.

INTRODUCTION

This report is the result of a special studies program for the analysis of inadvertent weather and climate modification. The program was initially concerned with the following class of problems:

1. Identification of the weather and climatic systems that are sufficiently unstable that human activities can trigger a change in conditions through some process; for example, the change in the regional radiation balance caused by the decrease in the stratospheric ozone (O_3) layer from NO_x emissions of high flying jet air aircraft.
2. Identification of the ecosystems that are sufficiently unstable that human activities, in conjunction with normal climatic conditions may produce severe climatic effects; for example, the burn-and-slash agricultural techniques that are being used in some of the tropical rain forests.
3. Identification of those areas of the world where humans have responded or have the potential of responding adversely to normal climatic variations; for example, the overgrazing and the overpopulation of the arid Sahel region of Africa during the recent drought.

4. Identification of natural phenomena that influence local and regional climate, such as the occurrence of snowbelts to the lee of the Great Lakes, and the occurrences of arid and semi-arid regions in the lee of mountain ranges (the rain shadow).
5. Determining if the items in 4. have an affect on global climate, and if humans can produce comparable results with their urban heat islands, nuclear energy centers, and large agricultural operations.
6. Investigating the technical feasibility of a global monitoring system to measure such quantities as atmospheric turbidity, CO_2 , SO_x , sulfates, O_3 , radioactivity, fluorocarbons, chlorocarbons, heavy metals, etc.

Early in the study we began to spend a considerable amount of time looking at the weather and climatic variations that may result from the large releases of heat and moisture from energy centers, both nuclear and fossil fuel. A smaller amount of time was spent in considering the regional impacts of burning fossil fuel in the northeastern United States. Hence, we did not complete the study outlined in the previous paragraph. However, the remainder of

this report briefly describes some of the phenomena considered in our study.

WHAT IS CLIMATE?

Climate is often defined as the sum of weather variations, or as the mean course of the weather. To define the climate at any given place on the globe, we require both the mean values and the time distributions of various weather phenomena. Some of the phenomena and data that are required to define the climate at a given place are: latitude and longitude; altitude; proximity to the sea, to lakes, and mountains; mean temperature and pressure; moisture in the atmosphere and the mean precipitation; hours of sunshine and the incidence of cloudiness; mean winds; visibility and the chemical composition of the atmosphere; amount of ice and snow cover; amount and type of vegetation; nature of the ground surface; and inclination and aspects of the slopes of the land.

WHY STUDY CLIMATE?

A valid reason for the scientific investigation of any phenomenon is the desire of the human mind to understand completely the processes that produce, maintain and influence the phenomenon. Hence, we study climatic processes to satisfy our scientific curiosity.

Another reason for the study of climate, and a more important one from the humanitarian point of view, is that climate affects, or is affected by, the processes required to meet all material needs of man. Our major material needs are air, water, soil, food, portable and stationary sources of energy, recreation and wildlife, metals, fibers, shelter, clothing, and other raw materials. In obtaining these needs, climate is often a governing factor; or weather and climate are adversely affected by the processes required to meet the needs.

With an expanding world population and increasing energy requirements for world agriculture, the demand for food is greater than the current world capabilities for producing that food. This fact, combined with recent adverse weather conditions and poor governmental policies, has greatly

reduced the world's grain reserve. Without adequate reserves, weather variability is especially critical and a future sequence of low grain-producing years could bring widespread shortages of food, and starvation and serious pressures in national and international politics and economics. Thus, it is very important that we quickly develop a better understanding of the variability of weather and climate and their significance to food production. Since modern agriculture is very energy intensive, a better understanding of climate will help alleviate both the world's food and energy crises.

CLIMATIC VARIATIONS

Climatic variations seem to be associated with variations in the vigor of the whole global circulation; but causes of global system variations are still largely unknown. Hence, the fundamental need in the study of climatic change is the quantitative understanding of the general circulation of the atmosphere. Since three-fourths of the heat that moves the atmosphere comes from the ocean surface, a quantitative understanding of oceanic heat transport and the ocean-atmospheric heat exchange is especially important. Even though we do not have a detailed explanation of the many cause-and-effect linkages that must be taken into account in a comprehensive climate theory, we can identify some of the causes of climatic variations. These variations operate over a wide variety of space and time scales; the time scales range from billions of years to seasons, and the space scales from the size of the solar system to the size of a farm. The following paragraphs list some important natural causes of climatic variations over various time and space scales.

Long-term climate changes may be due to the evolution of the sun, the presence of galactic dust between the earth and the sun, the mass and state of the atmosphere (excluding CO_2 , H_2O , O_3), the shifting of the poles on the earth, the geometry of the earth-sun system, the drifting of the continents, and the orogenesis of continents. Shorter term variations in climate may be due to the amount of CO_2 in the air, the atmosphere-ocean-ice cap feedback influences,

deep ocean circulation, solar variability (sunspot activity), volcanic dust in the stratosphere, other interactions of the atmosphere and the ocean, atmospheric particles from other sources, changes in the character of the land surfaces, and changes in the chemical composition of the atmosphere.

The causes and characteristics of deserts differ depending on their location and the general flow of air. Low-latitude deserts are caused by the descent of hot, dry air from aloft in areas of subtropical high pressure; examples are the Sahara, northern Mexico, Kalahari Desert of South Africa, and Australia. The mid-latitude deserts are caused by the precipitation of moisture on the windward sides of mountain ranges creating moisture deficit areas on the leeward sides; examples are the Gobi Desert of Central Asia and the deserts of the western United States. Cool, coastal deserts are produced by air that is chilled by the passage across cold coastal currents and then is warmed on contact with the hot land. Thus, its moisture capacity is increased and it becomes a drying wind. Examples are the west coast of lower California and Mexico, the coasts of Chile and Peru; and the Namib Desert of Southwest Africa.

The Great Lakes have a considerable influence on the weather near them. However, there is a serious lack of supporting evidence for this statement.¹ In spite of this deficiency, it was found that over the lakes and their downwind shore areas, the lake-caused average changes in cloud and precipitation amounts represent 5-15% reductions in summer and 5-45% increases in winter in comparison with upwind values. In addition there are well-defined snow belts in the lee of the Great Lakes.

Satellite and aircraft photography proves that 15% less rain falls over Lake Okeechobee, Florida than over the adjacent land areas.² The cooling influence of this 1694 km² lake acts to suppress the sea breeze convergence zone and thus produces a rainfall deficit over the lake.

Islands also modify local and mesoscale climate. For example, extensive areas of reduced waves on

the sea surface occur on the leeward side of the Lesser Antilles. The relatively calm seas are caused by the disruption of air flow over the islands. Using ERTS photographs, other investigators have found cloud-free areas in the lee of islands.

INADVERTENT WEATHER AND CLIMATE MODIFICATION

Human activity can change the average atmospheric temperature slightly and thus affect climate in several ways. For example, humans can

1. Increase the CO₂ content of the atmosphere by burning fossil fuels;
2. Decrease atmospheric transparency with aerosols from industry, automobiles and home heating units;
3. Decrease atmospheric transparency by introducing dust from agricultural practices;
4. Alter the thermal properties of the stratosphere with water, NO_x and SO_x effluents from high-flying jet aircraft;
5. Heat the atmosphere by burning fossil and nuclear fuels;
6. Change the ability of the earth's surface to reflect the sun's radiation back into space through urbanization, agriculture, deforestation and reservoirs;
7. Alter the rate of transfer of thermal energy and momentum between the oceans and the atmosphere by spilling oil on the water's surface, and
8. Irrigate farmland and cause more sunlight to be absorbed by the surface.

Most investigators feel that Items 1 and 4-8 will tend to warm the surface of the earth, while Items 2 and 3 may cool or warm the earth, depending on the location of the particles and their optical properties.

Many current reports and reviews discuss the inadvertent modification of weather and climate. Some of these references are as follows:

- (1) Reference [3] is a report of a study group concerned with the climatic and ecological effects of human activities, and the implications of change and remedial action;
- (2) Reference [4] is concerned with inadvertent climate modification and considers the general theory of climate and climate modeling, the fundamentals of climate change and man's activities that influence climate;
- (3) Reference [5] is a report that discusses climatic effects of a fleet of SST's flying in the stratosphere;
- (4) References [6] and [7] are concerned with both natural and anthropogenic causes for climate change;
- (5) References [8] to [12] are concerned with the climate of cities and the mechanisms by which urban areas can effect precipitation; five of these potential mechanisms are updrafts due to heating and frictional barrier effects, the addition of water vapor, the addition of condensation and freezing nuclei, and aerosol-induced electrification; and
- (6) Reference [13] deals with the overall environmental and ecological impacts of human activity.

ADVERTENT WEATHER AND CLIMATE MODIFICATION

In a previous section we briefly discussed the need for better climate theories and information for agriculture purposes. For the same reason, there is also a great need for reliable, advertent climate modification techniques. This latter need increases in importance as more and more marginal land is used to feed the world's hungry people.

Reference [14] discusses the current problems and progress in the

field of weather and climate modification. This report considers the modification techniques used on orographic and convective clouds, other precipitating cloud systems, fogs, lightning, hailstorms, hurricanes and tornadoes.

Reference [15] is a more extensive work on weather and climate modification. Finally, Fletcher¹⁶ outlines several proposed methods of modifying the global climate, such as creating a Bering Strait Dam to deflect the Gulf and Kuroshio Currents and create a Siberian Sea, and creating African seas from the Congo River System.

RESEARCH NEEDS

Reference [17] outlines a national climatic research program for the United States and considers the possibility of a similar program for the international scene. The major components of the program are as follows:

1. Monitoring: What is now going on?
2. Empirical Studies: How does the system work?
3. Climatic Data Analysis: What has happened in the past?
4. Future Climates: How and when is the climate going to change?
5. Numerical Models: What is shown by climatic simulations?
6. Theoretical Studies: How much do we really understand?
7. Climatic Impacts: What does it all mean to man?

This reference outlines the research needs in the major components and gives the interrelationships between these components. If this program is implemented by the US Government, then the environmental activities of ERDA that are related to the climatic impacts of energy production and use should be coordinated with this new program.

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A MODIFIED SET OF MAXWELL'S EQUATIONS THAT POSSESS
GALILEAN INVARIANCE

W. G. N. Slinn

For its possible pedagogic value, a self-consistent and Galilean invariant set of modified Maxwell's equations is presented by changing the usual time derivatives. The equations can be used to interpret the null in the Michelson-Morley experiment, but they are, of course, inconsistent with the second principle of special relativity.

INTRODUCTION

As is well known, Maxwell's equations of electrodynamics are not invariant under the Galilean transformation, but are under the Lorentz transformation. It is frequently stated in discussions of the special theory of relativity^{1,2} that first attempts to obtain Galilean invariance or interpret the null result of the Michelson-Morley experiment involved searching for appropriate modifications of Maxwell's equations. I have as yet not found illustrations of how Maxwell's equations might be modified to preserve Galilean invariance. The purpose of this report is to demonstrate such a modification for its possible pedagogic value.

Modified Maxwell's Equations

The set of equations to be analyzed is

$$\nabla \cdot \vec{D} = \rho_s \quad (1)$$

$$\nabla \cdot \vec{B} = 0 \quad (2)$$

$$\nabla \times \vec{E} = -D\vec{B}/Dt \quad (3)$$

$$\nabla \times \vec{H} = \vec{j}_s + D\vec{D}/Dt \quad (4)$$

in which the only change from the usual set of Maxwell's equations (and customary notation) is the introduction of the special time derivative,

$D/D = \partial/\partial t + \vec{v}' \cdot \nabla$, where $\vec{v}'$ is the (assumed constant) velocity of the source of the fields with respect to the observer. The subscripts on ρ and $\vec{j}$ will be identified immediately. Thus this formalism singles out as special, the one observer for whom the source appears stationary. For him, the equations reduce to the familiar set of Maxwell's equations

$$\nabla \cdot \vec{D}_s = \rho_s \quad (5)$$

$$\nabla \cdot \vec{B}_s = 0 \quad (6)$$

$$\nabla \times \vec{E}_s = -\partial \vec{B}_s / \partial t \quad (7)$$

$$\nabla \times \vec{H}_s = \vec{j}_s + \partial \vec{D}_s / \partial t \quad (8)$$

where, as an interim measure, subscripts have been added to the field variables in case they might be different for different observers. Eqs. (5)-(8), of course, describe an electromagnetic wave traveling in free space at a speed $c = (\epsilon_0 \mu_0)^{-1/2}$ with respect to the wave source.

SELF-CONSISTENCY

Before demonstrating the Galilean invariance of Eqs. (1)-(4), it is perhaps of some interest to demonstrate that they are self-consistent and consistent with the continuity equation for charge. Thus, taking the divergence of Eq. (3), then using

Eq. (2), leads to an identity, and the divergence of Eq. (4) with Eq. (1) gives

$$\frac{D\rho_S}{Dt} + \nabla \cdot \vec{j}_S = 0. \quad (9)$$

For $\vec{v}'$ constant, Eq. (5) can also be written as

$$\frac{\partial \rho_S}{\partial t} + \nabla \cdot (\vec{j}_S - \rho_S \vec{v}) = 0 \quad (10)$$

where $\vec{v} = -\vec{v}'$ is the velocity of the observer with respect to the source. Now in Eq. (10), we recognize $\vec{j}_S - \rho_S \vec{v}$ as the current density, say $\vec{j}$, as measured by the moving observer if $\rho = \rho_S$. Thus Eq. (10) can also be written as

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \vec{j} = 0 \quad (11)$$

which is the desired continuity equation for charge.

GALILEAN INVARIANCE

To demonstrate the Galilean invariance of Eqs. (1)-(4) we identify two observers moving at uniform velocities $\vec{v}_1$ and $\vec{v}_2$ with respect to the source, and relate their coordinates by

$$\vec{r}_2 + \vec{v}_2 t = \vec{r}_1 + \vec{v}_1 t_1 \quad (12)$$

and

$$t_2 = t_1 = t. \quad (13)$$

It is easy to see that observers 1 and 2 agree on spatial derivatives since, for example,

$$\left. \frac{\partial \vec{B}_1}{\partial x_1} \right|_{t_1} = \left[\frac{\partial \vec{B}_1}{\partial x_2} \frac{\partial x_2}{\partial x_1} + \frac{\partial \vec{B}_1}{\partial t_1} \frac{\partial t_1}{\partial x_1} \right]_{t_1} \quad (14)$$

and using $x_2 = x_1 - v_x t$ where $\vec{v} = \vec{v}_2 - \vec{v}_1$ is the relative velocity,

then $\partial x_2 / \partial x_1 = 1$ and $\partial t / \partial x_1 = 0$, so that x-space derivatives are the same. On the other hand, time derivatives are related, for example, by

$$\left. \frac{\partial \vec{B}_1}{\partial t_1} \right|_{x_1} = \left[\frac{\partial \vec{B}_1}{\partial t_2} \frac{\partial t_2}{\partial t_1} + \frac{\partial \vec{B}_1}{\partial x_2^i} \frac{\partial x_2^i}{\partial t_1} \right]_{x_1} \quad (15)$$

(sum over coordinates x_2^i). Since $\partial t_2 / \partial t_1 = 1$ and $\partial x_2^i / \partial t_1 = -v^i$, then Eq. (15) yields

$$\frac{\partial \vec{B}_1}{\partial t_1} = \frac{\partial \vec{B}_1}{\partial t_2} - \vec{v}_2 \cdot \nabla_2 \vec{B}_1 + \vec{v}_1 \cdot \nabla_2 \vec{B}_1. \quad (16)$$

If the above transformations are substituted into Eqs. (1)-(4) written for observer 1, then observer 1, in effect, suggests to observer 2 the use of the equations:

$$\nabla_2 \cdot \vec{D}_1 = \rho \quad (17)$$

$$\nabla_2 \cdot \vec{B}_1 = 0 \quad (18)$$

$$\begin{aligned} \nabla_2 \times \vec{E}_1 = & - \left(\frac{\partial \vec{B}_1}{\partial t} - \vec{v}_2 \cdot \nabla_2 \vec{B}_1 + \vec{v}_1 \cdot \nabla_2 \vec{B}_1 \right) \\ & + \vec{v}_1 \cdot \nabla_2 \vec{B}_1 \end{aligned} \quad (19)$$

$$\begin{aligned} \nabla_2 \times \vec{H}_1 = & \vec{j}_S + \left(\frac{\partial \vec{D}_1}{\partial t} - \vec{v}_2 \cdot \nabla_2 \vec{D}_1 \right. \\ & \left. + \vec{v}_1 \cdot \nabla_2 \vec{D}_1 \right) - \vec{v}_1 \cdot \nabla_2 \vec{D}_1. \end{aligned} \quad (20)$$

But upon cancelling equal terms, we see that this set of equations is the set which observer 2 would have chosen for himself from Eqs. (1)-(4) if invariance of the field variables $\vec{D}$, $\vec{B}$, $\vec{E}$ and $\vec{H}$ is assumed.

Wave Equation

From Eqs. (1)-(4) one can obtain a wave equation for the field in a vacuum using $\rho = \rho_S = 0 = \vec{j}_S$ and $\vec{D} = \epsilon_0 \vec{E}$, $\vec{B} = \mu_0 \vec{H}$. For example, the curl of Eq. (3) and use of Eq. (4) lead to

$$\nabla^2 \vec{E} = \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} - \vec{\nabla} \cdot \nabla \left[\frac{2}{c^2} \frac{\partial \vec{E}}{\partial t} - \frac{1}{c^2} \vec{\nabla} \cdot \nabla \vec{E} \right] \quad (21)$$

where $c^2 = (\epsilon_0 \mu_0)^{-1}$. For a plane wave Eq. (21) becomes

$$\frac{\partial^2 \vec{E}}{\partial t^2} = (c^2 - v^2) \frac{\partial^2 \vec{E}}{\partial x^2} + 2v \frac{\partial^2 \vec{E}}{\partial x \partial t} \quad (22)$$

which describes waves traveling at $\pm c$ with respect to the source or $(\pm c - v)$ with respect to an observer moving at speed v relative to the source. Although it is easy to see that Eq. (22) can be used to explain the null in the Michelson-Morley experiment, it is inconsistent with the second principle of Einstein's special theory of relativity which specifies that light travels at the same speed c , regardless of the speed of the source relative to the observer.

CONCLUDING REMARKS

It might be of some convenience to present also the set of modified Maxwell equations in a slightly different form. From Eqs. (1)-(4) and the use of a few vector identities one can obtain

$$\nabla \cdot \vec{D} = \rho \quad (23)$$

$$\nabla \cdot \vec{B} = 0 \quad (24)$$

$$\nabla \times (\vec{E} + \vec{\nabla} \times \vec{B}) = - \dot{\vec{B}} \quad (25)$$

$$\nabla \times (\vec{H} + \vec{\nabla} \times \vec{D}) = \vec{j} + \dot{\vec{D}} \quad (26)$$

where all quantities are measured by the moving observer ($\vec{v}' = -\vec{v}$) and $\dot{}$ is the partial derivative with respect to time. It is noted that if effective $\vec{E}$ and $\vec{H}$ fields are defined in an obvious manner then these definitions agree with the customary ones associated with the Lorentz transformed Maxwell's equations,³ at least to order v/c .

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CALIBRATION OF SUNPHOTOMETERS

N. S. Laulainen and E. W. Kleckner

Multiwavelength sunphotometers may be calibrated using either a standard laboratory light source or using the sun itself. Here we discuss the procedures used for calibration by these methods. The solar method leads to smaller uncertainties in the calibration constants for each wavelength; however, the standard lamp method is essential to check photometer stability. A combination of the two methods is recommended.

INTRODUCTION

The measurement of solar radiation at the earth's surface provides important information about suspended particles and trace gases in the atmosphere as well as providing quantitative data on insolation required for various solar energy conversion schemes. Several types of optical instruments may be employed for measurements of this type and each must be calibrated against some standard to interpret the observations correctly. In this report we discuss the calibration of one type of solar radiation instrument--the multiwavelength sunphotometer. Tests of several different sunphotometers have been carried out during 1975, with special emphasis on verifying the accuracy of turbidity data obtained during the 1975 METROMEX program.

CALIBRATION PROCEDURES

Several methods are available for calibrating sunphotometers. One method uses the sun itself as a standard light source while the other uses a laboratory standard lamp whose spectral irradiance is known, or at least known relative to the solar spectral irradiance. In practice a combination of the two methods is usually required.

In order to discuss the procedure for the solar radiation method, it is necessary to examine the basic equation relating the measured light intensity at the earth's surface to the

intensity value which would be obtained without attenuation in the atmosphere. This relationship is usually expressed as

$$J_{\lambda} = J_{\lambda 0} \exp[-\tau_{\lambda} m] / F, \quad (1)$$

where

J_{λ} is the actual photometer reading for wavelength band λ ,

$J_{\lambda 0}$ is the outside-the-atmosphere photometer reading or calibration constant for wavelength band λ ,

F is a correction factor depending on solar distance,

τ_{λ} is the total atmospheric optical depth through the zenith at wavelength λ , and

m is the relative airmass thickness in units of the atmospheric thickness through the zenith.

The optical depth, τ_{λ} , is the sum of attenuation contributions due to molecular (Rayleigh) scattering, molecular (usually ozone) absorption and aerosol scattering and absorption.

A more convenient form of Equation (1) from the standpoint of calibration constant determination is:

$$\ln J_{\lambda} = \ln J_{\lambda 0} - \ln F - \tau_{\lambda} m. \quad (2)$$

This form clearly shows the relation between the dependent variable, $\ln J_{\lambda}$,

and the independent variable, m . A standard procedure for determining the zero airmass intercept, $\ln J_{\lambda 0} - \ln F$, and the optical depth (slope), τ_{λ} , is a least squares solution to a set of measurements $\{\ln J, m\}$. This procedure is valid only if $\ln J_{\lambda 0}$ and τ_{λ} are constant over the time interval during which the observations are made. If either of these quantities are variable, then their determined values will be in error. A more complete discussion of instrument stability and the effects of optical depth variations on instrument calibration is given elsewhere.¹ By choosing observation runs of 1-2 hr during the times of day when the relative airmass is changing rapidly, and using solar radiation measurements only for the cleanest days, both the effects of instrumental variation and optical depth variation (changes in aerosol loading) can be minimized. Moreover any instrumental variation due to changes in temperature should be negligible if the photometer is stored at room temperature between observations. The observations themselves require only 1 or 2 min to complete.

In principle, sunphotometer calibration using a standard lamp is a straightforward procedure. Because the radiant flux decreases as the inverse square of the distance from the lamp, a method for checking sunphotometer linearity is also available. Some precautions must be taken to insure accurate results. To avoid the possibility of sudden changes in standard lamp irradiance from aging effects, intercalibration with two or more standard lamps is recommended. Operation of the standard lamps according to the manufacturer's specifications requires a stable, constant current supply, otherwise current variations can also cause lamp radiance changes. Minimizing spurious reflections from walls, ceilings and components in the calibration room is also necessary for accurate and reproducible calibrations.

The calibration of the photometer is determined by pointing the photometer at the standard lamp at a minimum distance where the solid angle of the aperture subtends the entire filament and lamp cavity. The observed photometer response $J_{\lambda 0}$ depends upon the spectral irradiance of the source

$F(\lambda)$, photometer gain, G , and the filter/detector response, $R(\lambda)$ through the equation

$$J_{\lambda 0} = k \int_{\Delta\lambda} G F(\lambda') R(\lambda') d\lambda', \quad (3)$$

where the integration is essentially over the wavelength bandpass $\Delta\lambda$ of the filter and k is a constant of proportionality. For both the sun and standard lamp, the spectral irradiance is a slowly varying function of wavelength, while in most cases the filter/detector response is nearly like a delta function (for broadband filters this assumption is poor). The integral in Equation (3) can then be approximated by

$$J_{\lambda 0} = k G F_{\lambda} R_{\lambda} \Delta\lambda, \quad (4)$$

where F and R are evaluated at the center of the filter detector pass-band, λ , and $\Delta\lambda$ is the full width at half intensity of R . By taking the ratio of the solar calibration photometer response to the standard lamp response,

$$\frac{J_{\lambda 0}^{\text{sun}}}{J_{\lambda 0}^{\text{std}}} = \frac{k \cdot F_{\lambda}^{\text{sun}} \cdot R_{\lambda} \cdot G_{\lambda}^{\text{sun}}}{k \cdot F_{\lambda}^{\text{std}} \cdot R_{\lambda} \cdot G_{\lambda}^{\text{std}}}, \quad (5)$$

one finds that the filter/detector response cancels. Thus the sunphotometer calibration is given by

$$J_{\lambda 0}^{\text{sun}} = J_{\lambda 0}^{\text{std}} \frac{F_{\lambda}^{\text{sun}} G_{\lambda}^{\text{sun}}}{F_{\lambda}^{\text{std}} G_{\lambda}^{\text{std}}}. \quad (6)$$

The sunphotometer can also be calibrated by viewing a Lambertian screen that is directly illuminated by a standard lamp. This procedure is more appropriate if the photometer is used to measure diffuse radiation.

Once the calibration $J_{0\lambda}$ is established for a given photometer, any long term drifts caused by aging which may occur can be assessed by periodic comparison to the standard lamps.

CALIBRATION RESULTS

Two six-wavelength sunphotometers have been calibrated using the procedures described in the previous section. The nominal spectral bands of

each photometer are listed in Table 1. Photometer A employs a RCA 1P38 vacuum photodiode with S4 response (longwavelength cutoff around 650 nm) for light detection. Photometer B uses a UDT PIN6DP silicon photodiode operated in a photo voltaic mode; it has a long wavelength cutoff at about 1.08 μm . Photometer A has been tested and used extensively in the field (76 days in 1975 and 14 days in 1974); photometer B is relatively new and has been used in the field only 5 days. Hence most of the results discussed in this section will be confined to photometer A.

Because of the large number of observations taken with photometer A during 1975, it was possible to select

10 reasonably "clean" days for which the solar calibration method could be used. The selection process for these 12 days was somewhat subjective, since aerosol variations were apparent even for the "cleanest" days. Quantitatively, only those days for which the least squares solution to Equation (2) resulted in a calibration constant $J_{0\lambda}$ with less than 2% calculated uncertainty were retained. This set of days also had calculated optical depths τ_λ approaching the clean air values. The calibration constants for photometer A determined by this method are summarized in Table 2. Photometer B has not yet been calibrated by this method.

TABLE 1. Sunphotometer Spectral Bands--Central Effective Wavelength and Bandwidth

Filter No.	Photometer A		Photometer B	
	Wavelength (nm)	Bandwidth (nm)	Wavelength (nm)	Bandwidth (nm)
1	448.2	8.6	690.0	12.1
2	500.0	~60	640.1	6.0
3	500.7	6.6	435.8	7.4
4	557.8	8.5	500.0	7.4
5	618.9	7.8	550.0	9.2
6	598.4	~40	600.0	10.1

TABLE 2. Sunphotometer Calibration Constants

Filter No.	A Calibration		
	Gain	Solar Method	Standard Method
1	1	161.8 $\pm$ 2.7	159 $\pm$ 10
2(a)	2	238.3 $\pm$ 2.6	223 $\pm$ 27
3	10	203.9 $\pm$ 2.8	200 $\pm$ 12
4	5	251.6 $\pm$ 3.2	231 $\pm$ 14
5	100	199.0 $\pm$ 3.6	180 $\pm$ 27
6*	2	110.2 $\pm$ 2.2	100 $\pm$ 12

a. Wide band filter.

The standard lamp procedure was used to calibrate photometer A. Direct viewing of the standard lamp at several distances as well as viewing a Lambertian screen were carried out in the calibration procedure. The results for these tests are also shown in Table 2. A high intensity 1000-W tungsten halogen lamp (Optronic Laboratories, Inc., Model 100C, Serial No. HTS-83, calibration traceable to NBS) was used as the standard lamp. This lamp was chosen because of its stability and high output performance; it produces a one-solar constant irradiance standard at 40 cm. Because of the finite size of the lamp (5 x 7 cm) and the narrow field of view of the photometer (2° aperture), it was necessary to place the photometer farther than ~2.4 m away to insure that the photometer viewed the entire lamp. The actual distance from the standard was 2.45 m. The irradiance measured at 2.45 m was referred to the standard irradiance at 40 cm using the inverse square law. Because of the distance, the photometer was operated at

its highest gain sensitivity; meter response was at best only one fourth of full scale, leading to higher reading errors than normally encountered under actual field operation. The uncertainty in the spectral lamp irradiance is quoted by the manufacturer to be ~5% in the visible. The uncertainties in the photometer calibration by the standard method given in Table 2 reflect this 5% uncertainty. For wide band filters 2 and 6, the approximation of Equation (4) is poor and the uncertainty is appropriately larger--at least until a more exact calculation is carried out. The large uncertainty for filter 5 is mostly due to a large reading error, as the measured intensity was very low, ~2% of full scale. In general, however, the standard lamp calibration values are in fair agreement with those obtained by the solar method. Additional refinements of the standard method are still required, since at this point the solar method appears to produce smaller uncertainties.

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ATMOSPHERIC TURBIDITY OVER HANFORD DURING 1975

N. S. Laulainen

Continued solar radiation measurements at Hanford during 1975 using a multiwavelength sunphotometer are discussed. Seasonal variations in aerosol loading are apparent in the data. Changes in aerosol size distribution and/or index of refraction can be identified in the multiwavelength turbidities.

INTRODUCTION

The presence of aerosol in the atmosphere has significant effects on the incident solar radiation. In cases of high aerosol loading, insolation is severely reduced at the surface; this clearly has important implications for the utilization of solar energy. The climatic effects of aerosol on global and regional scales, through changes of the apparent albedo of the earth, are not yet fully appreciated because the critical aerosol optical properties are difficult to measure. Consequently, too few reliable data on these properties are at present available. Aerosol optical depth measurements, combined with other simultaneous optical measurements will be useful in assessing aerosol effects on insolation and albedo.

Multiwavelength aerosol optical depth (turbidity) measurements using sunphotometry techniques were initiated at the Hanford Meteorological Station (HMS) in July 1974. Frequent observations were continued during 1975. Results obtained during the last half of 1974 were summarized in an earlier report.¹ Here we summarize turbidity data collected on 63 days, including 2 blowing dust episodes on March 25 and September 11, during 1975 over the Hanford area.

The aerosol optical depth is derived from the extinction equation

$$J_{\lambda} = J_{\lambda 0} \exp (-\tau_{\lambda} m)/F, \quad (1)$$

where

J_{λ} is the actual photometer reading for wavelength band λ ,

$J_{\lambda 0}$ is the outside-the-atmosphere photometer reading or calibration constant for wavelength band λ ,

F is a correction factor depending on solar distance,

τ_{λ} is the total atmospheric optical depth through the zenith at wavelength band λ , and

m is the relative airmass thickness in units of the atmospheric thickness through the zenith.

The total optical depth, τ_{λ} , is the sum of attenuation contributions due to molecular (Rayleigh) scattering, molecular (usually ozone) absorption and aerosol scattering and absorption; explicitly it is

$$\tau_{\lambda} = \tau_{\text{Rayleigh}} + \tau_{\text{ozone}} + \tau_{\text{aerosol}}. \quad (2)$$

The Rayleigh contribution is relatively constant and predictable. The ozone contribution is mainly from stratospheric ozone and varies with time of year and latitude by as much as 30%. Both these contributions are assumed to be the standard atmosphere values tabulated by Elterman² and are subtracted from the total optical depth to obtain the aerosol optical depth. Simultaneous and independent measurements of ozone are recommended to determine aerosol extinction accurately.

RESULTS AND DISCUSSION

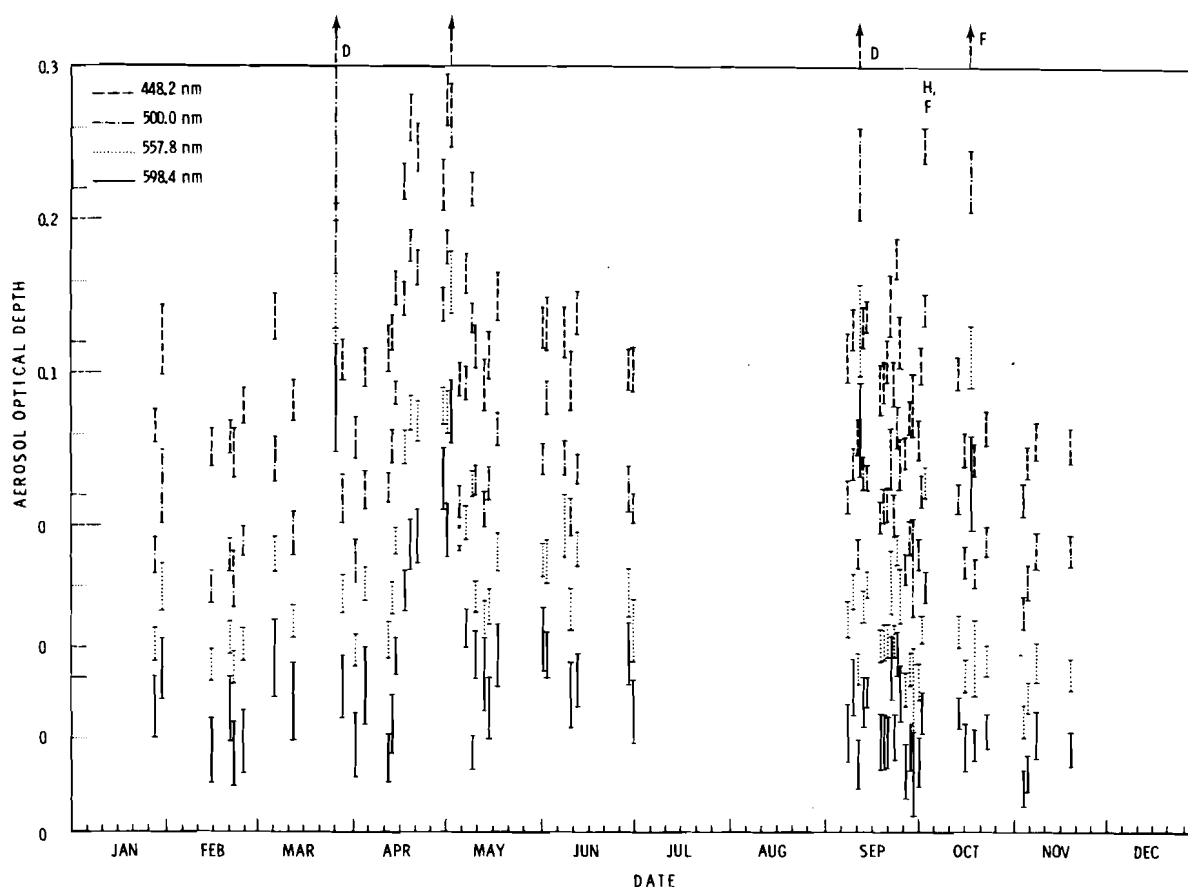
Average total optical depths were determined for 6 colors from solar radiation measurements on 63 days during 1975, using Eq. (1) and the

calibration constants $J_{\lambda 0}$ for each wavelength band (calibration of the sunphotometer is discussed elsewhere³). Aerosol optical depths (turbidities) were obtained by subtracting the Rayleigh and ozone contributions and are shown in Figure 1 for four colors. Three striking results are immediately apparent from an inspection of Figure 1:

1. Spring and fall show the largest optical depths while the winter and summer months show minimum values. The summer minimum is somewhat larger than in winter.
2. All colors exhibit similar day-to-day optical depth variations, with the magnitudes of the variations roughly comparable in each color.

3. Even during the seasonal maxima, occasional low values ("clean" air) in turbidity are observed.

The significance of the spring and fall maxima are related to agricultural activity, desert vegetation and meteorological conditions. During the spring months, considerable newly planted land is exposed to the dry, seasonal wind storms resulting in increased loading of particulate material. Smoke from smudge pot burning for frost protection was also apparent during these months. Late spring and early summer rains as well as crop growth reduce the amount of exposed soil material which can be resuspended. Because the desert vegetation (sages, etc.) is becoming more active for the new growing season,



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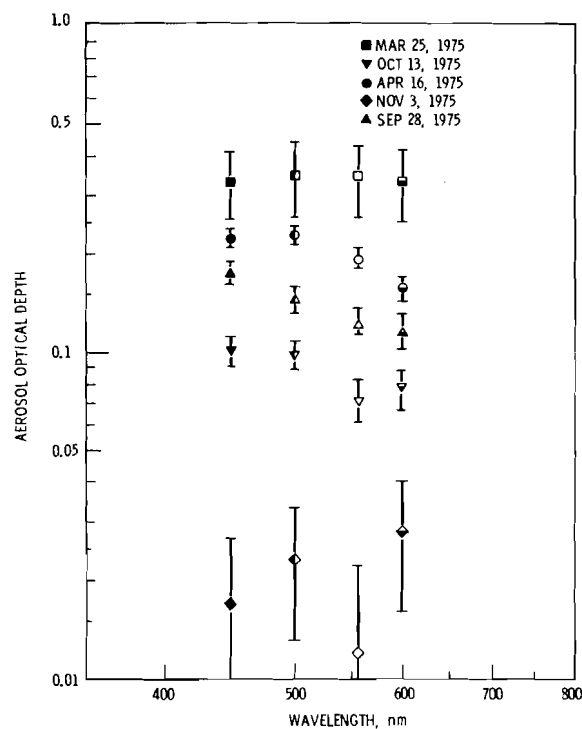
FIGURE 1. Aerosol Optical Depth in Four Colors (448.2, 500, 557.8 and 598.4 nm) over Hanford Meteorological Station During 1975. Note that the origins of the optical depth axes for each color have been displaced for better data separation.

the reduction of suspended material is not as great as in the winter when the vegetation is essentially dormant. Thus the summer minimum should be higher than in winter. (Unfortunately, no data were available during July and August of 1975; however data from these same months in 1974¹ do indicate a relative minimum for this time of year.) By late summer and early fall, desert vegetation activity is at a maximum; crop harvesting is also well underway. Occasional windy periods also occur during this season, as well as prolonged inversion conditions, which tend to trap dust and other particulate material in the lower atmosphere. Consequently the occurrence of a second turbidity maximum in the fall is not surprising.

Because the turbidity is an indicator of particulate loading, it is not surprising that all colors exhibit similar day-to-day variations due to changes in the amount of particles suspended in the atmosphere. Closer inspection of Figure 1 reveals that the variations in each color are not always the same, indicating that changes in aerosol size distribution and/or changes in aerosol refractive index (chemical composition variation) can also occur. These differences are more clearly indicated in Figure 2, where mean turbidities for 5 days are plotted against wavelength. Data for November 3 (a "clean" day) and March 25 (a very "dusty" day) roughly represent lower and upper bounds to the turbidity. Both of these days, despite the large differences in aerosol loading, indicate that the aerosols are roughly neutral with respect to color. This implies a predominance of particles larger than about 1 μm . The other days (April 16, September 28 and October 13) show varying degrees of wavelength dependence, with wavelength exponents of $\alpha = 1.7, 1.5$ and 1.2 , respectively. The parameter α is defined by the power law relation

$$\tau_{\text{aerosol}} \propto \lambda^{-\alpha} \quad (3)$$

The usual range of α is 0.5-2.5 with occasional values in the -1 to 0 range at background sites.⁴ Rayleigh scat-



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FIGURE 2. Aerosol Extinction Color Diagram Showing the Wavelength Dependence of Aerosol Optical Depth for Different Particulate Mass Loadings and Size Distributions

tering always has a wavelength dependence,

$$\tau_{\text{Rayleigh}} \propto \lambda^{-4} \quad (4)$$

(In an earlier report,¹ a red "hump" present in the color diagram may have been due partly to the use of incorrect calibration constants, as no dramatic red "lump" is present in the 1975 data.)

The occurrence of minimum turbidity values during seasonal maxima indicates the natural cleansing action of the atmosphere by washout, settling and ventilation such as the passage of a clean air mass through the region. More careful considerations of prevailing meteorological conditions will be useful in assessing atmospheric aerosol variations.

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COMMENTS ON THE DETERMINATION OF AEROSOL OPTICAL PROPERTIES

N. S. Laulainen

In considering the effects of aerosols on the radiative balance of the earth-atmosphere system, the two aerosol parameters which appear to have important climatological implications are the hemispheric backscatter coefficient and the absorption coefficient. Various procedures are usually employed to calculate these parameters, many of which suffer ambiguities from specifying physical and chemical aerosol characteristics, or from approximations used in the calculations. This paper suggests that since the important radiative properties are optical in nature, approaching radiative transfer from a new optical measurement techniques standpoint is more appropriate.

INTRODUCTION

Recent years have seen considerable effort to determine the physical and chemical properties of atmospheric aerosols, both of natural and anthropogenic origin. The reasons for this effort are clear; atmospheric aerosols profoundly affect precipitation development and can significantly modify the climate by altering the solar radiation field. The fundamental parameters which are required from measurements of aerosol systems

appear to be the size distribution, complex index of refraction, particle solubility and relative humidity, and some as-yet-unspecified parameter describing particle shape.

Many optical systems have been developed to measure aerosol size distributions. These systems take advantage of light-scattering properties of aerosols as a function of size. Most of their shortcomings occur because the light scattering is also

sensitively dependent on particle refractive index and shape, such that the size measurements can only be referenced to equivalent spheres of known index and size used in calibrating these measurement systems. When inferring the effects of particles on precipitation development, such ambiguities in specifying the true size distribution may be unimportant. However, when specifying the effects of aerosols on solar radiation, such ambiguities must be resolved. This paper discusses the problem of interpreting optical measurements in terms of those parameters which appear to have important climatological implications.

CRITICAL RADIATIVE PROPERTIES OF AEROSOLS

To a first approximation the problem of direct effects of aerosols on the radiation balance of the earth comes down to a specification of how much radiation particles in the atmosphere absorb and how much they reflect back into space. The absorption of energy leads to heating while back scattering leads to cooling. The critical value (ρ_c) of the ratio (ρ) of aerosol layer absorption to hemispheric back scattering has been shown to depend on the effective albedo (A) through the expression¹

$$\rho_c = \frac{(1 - A)^2}{2A} \quad (1)$$

An aerosol layer for which $\rho > \rho_c$ implies a decrease in system albedo (heating) whereas if $\rho < \rho_c$ an increase in system albedo occurs (cooling). Since the actual energy change may include dynamic and thermal (indirect) effects, the crossover between heating and cooling is not well defined.

In principle, simultaneous measurement of hemispheric backscatter extinction and absorption extinction coefficients for a wide range of aerosol layers found in the atmosphere, under a variety of meteorological conditions, is all that would be required to determine this ratio. Suitable instrumentation exists for the measurement of aerosol hemispheric backscatter in situ.² However, few real time methods exist for the measurement of aerosol absorption,^{3,4} in situ or oth-

erwise. Consequently various procedures have been developed for deducing this quantity from other measurements (some optical) of aerosol properties.

PROCEDURES FOR CALCULATING RADIATIVE EFFECTS OF AEROSOLS

Numerous calculations have been carried out to estimate the effects of aerosol layers on solar radiation, either through a complete numerical solution to the radiative transfer equation⁵⁻⁷ or through simplified two-stream approximations to this equation.^{8,9} The usual procedure for incorporating aerosols into these models is to make a number of assumptions about the size distribution, shape and refractive index, and to apply Mie theory to obtain, for example, the angular scattering phase function of the aerosols and optical depth of the aerosol layer. The calculations assume:

1. The particles are spherical in shape.
2. The particle size distribution is accurate and/or can be represented by a parameterized aerosol size distribution function such as a power law (Junge) distribution or a modified gamma (Deirmendjian) function.
3. All particles have the same complex refractive index (homogeneous aerosol) and the index is uniform within each particle. (Sometimes several calculations are carried out for a range of particle refractive indices.)

Most of these assumptions are poorly justified, since in general the particles are of various chemical compositions (heterogeneous) and each particle itself can be a mixture of substances or a single substance. The application of Mie theory to the particle size distribution to obtain the optical property of interest (usually an integral property) can be represented symbolically by an operator M applied to the aerosol distribution $n(r, m = n_{re} + in_{im})$

$$P_{opt} = M n(r, m), \quad (2)$$

where n represents the number of particles of size r with complex index

of refraction m . Clearly the calculated values of aerosol optical properties will contain errors, caused by an inaccurate description of the aerosol particles and the fact that the Mie operator can only be applied (at present) to spheres.

Other approaches which suffer from similar defects involve the inversion of optical measurements of aerosols to obtain particle size distribution and refractive index.¹⁰⁻¹⁴ In this case the optical measurements (e.g., turbidity, sky polarization, etc.) are not directly or easily related to the hemispheric backscatter extinction coefficient and absorption extinction coefficient, and yet may contain considerable information about the nature of the aerosol if one is clever enough to extract it. Thus it may seem reasonable to analyze the data in a manner schematically represented by:

$$p_{\text{opt}}^{\text{measured}} \xrightarrow{M^{-1}} n(r, m) \xrightarrow{M} p_{\text{opt}}^{\text{desired}} ; \quad (3)$$

i.e., a particle size distribution is derived from some suitable inversion of the measured optical data, $p_{\text{opt}}^{\text{measured}}$, and the derived size distribution is then used to calculate the desired optical property, $p_{\text{opt}}^{\text{desired}}$. The three assumptions mentioned previously still must be made, causing the inaccuracies already mentioned. More seriously, however, the inversion procedure M^{-1} usually does not lead to a unique solution for the size distribution and complex refractive index. Part of the reason for this nonuniqueness is that the optical measurements are only sensitive to particles in the 0.1-1 μm size range. Thus even more uncertainties are introduced to the value of the desired quantity. (In some sense, particle size distribution measurements using optical particle counters of one variety or another suffer from the same defects described above, since they are calibrated against spherical particles of a given refractive index. Thus even here ambiguities in the size distributions derived from such measurements are surely present and need to be correctly accounted for.)

Various improvements to the measurement and computational procedures outlined above are presently underway.^{15,16} These improvements involve simultaneous measurement of several aerosol properties such as aerosol size distribution and vertical distribution, aerosol optical depth (turbidity), and light scattering extinction coefficients (hemispheric backscattering and/or total scattering) or perhaps other combinations including those just mentioned, lidar backscattering, sky polarization and sky brightness. Simultaneous measurements allow more valid comparison between the various observed quantities than was previously possible, e.g., there is no need to assume a special size distribution or vertical profile since these quantities are also measured in equivalent optical units. The advantages of equivalent optical units are clear, although the procedure schematically represented by Eq. (3) is still preferred by many authors.^{1,15} Basically, the use of these equivalent optical units avoids vagaries of applying Mie theory to nonspherical particles, nonhomogeneous aerosol components and assumed particle size distributions. If one is clever enough, it would be ideal to manipulate some set of simultaneously measured optical properties, $\{p_{\text{opt}}^{\text{measured}}\}$, in order to derive another set of aerosol optical properties, $\{p_{\text{opt}}^{\text{radiative}}\}$, which explicitly contain the important radiative effects necessary for planetary heat balance calculations. This manipulation can be symbolically expressed as

$$\{p_{\text{opt}}^{\text{radiative}}\} = O \{p_{\text{opt}}^{\text{measured}}\}. \quad (4)$$

Since the radiative properties themselves are optical in nature, it makes sense to approach the radiative transfer problem from an optical measurement standpoint. In addition since most optical techniques can be applied to the in situ aerosol (remote or local sampling), the aerosol is measured in the same chemical and physical state as it exists in the ambient atmosphere.

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PLUME DIMENSIONING BY LONG EXPOSURE PHOTOGRAPHY

J. M. Thorp

Quantitative measurements of power plant cooling tower plumes have been made utilizing an established procedure, which permits three-dimensional plume description from a two-dimensional photograph. Photographs of the plume were made each hour from two camera sites located about 75° from each other. Double theodolite sightings were taken from the cooling tower site concurrent with the hourly photographs. Exposure times were 300 sec on 35 mm film to portray an integrated plume pattern. Time lapse movies at 2 frames/min were taken during all experimental periods.

INTRODUCTION

A Battelle research team conducted a series of experiments at the Rancho Seco Nuclear Generating Station near Sacramento, California, in February 1975, to define the characteristics of natural draft cooling tower plumes. This report deals solely with the procedures to obtain the plume's physical dimensions; the internal characteristics of the plume are discussed elsewhere in this publication.

The approach employs long time-exposure photography as described by Culkowski,¹ and a geometrical technique by Niemann, et al.,² for analyzing the photographs of plumes which are at an angle to the film plane. The procedures are presented in sufficient detail to serve as a guide for subsequent photograph analysis.

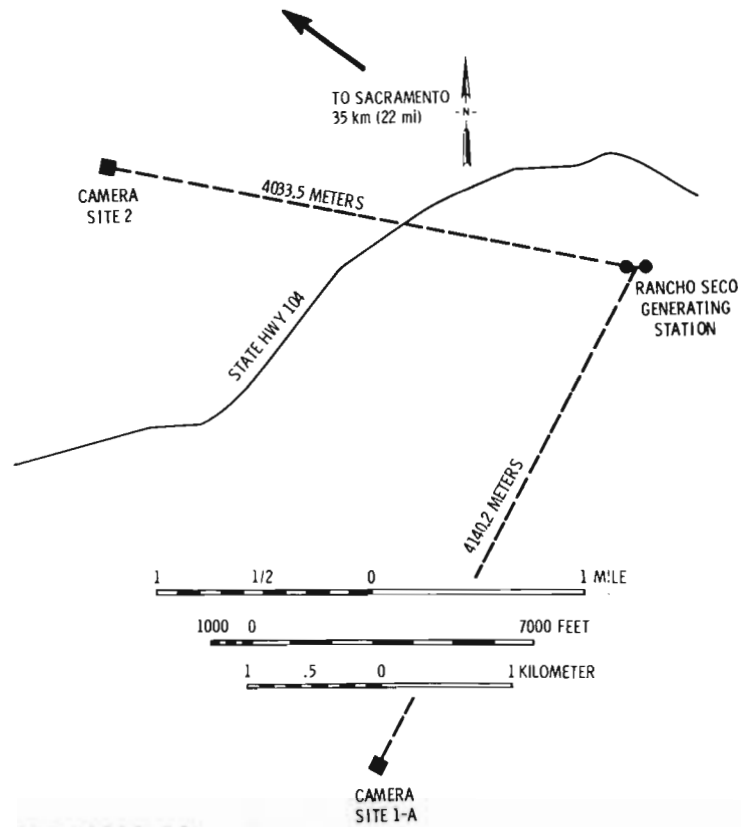
FIELD OPERATIONS

The experimental design called for photographing the plumes from two camera stations situated about 4 km from the cooling towers, at an angle of approximately 74° from each other. Figure 1 shows the location of the camera sites, cooling towers and general topographic features. Two cameras were positioned at each site (Figure 2); a 35-mm Topcon mounted on a modified transit platform which permitted precision leveling and orientation, and a 16-mm Bolex movie camera

enhanced with an automatic interval timer and automatic lens system.

The Bolex movie cameras were set at the beginning of each day's experiment in a time lapse mode of 1 frame every 30 sec and except for an infrequent film change required no further attention during the day. A clock was mounted in such a way that its face was readable in each movie frame. The movie lens used was a Vario-Switar f 1.9 with an automatic diaphragm and zoom capability from 16 mm to 100 mm; at 2 frames/min, a 100-ft film roll with a capacity of approximately 4000 frames permits more than 30 hr filming without a film change. Kodachrome II color movie film was used for this part of the photographic record.

The 35-mm cameras were manually operated once per hour to take a 300-sec time exposure. The procedure for long time exposures described by Culkowski¹ proved quite satisfactory. This method takes into account the so-called "reciprocity failure" of photographic films, which occurs when film is exposed much longer than designed to be. A "slow" film, Panatomic X, with an ASA rating of 25, was chosen for its fine grain and availability. To permit such long exposures with normal photographic lenses, a neutral density filter was placed in front of the lens. More than one neutral density or colored filter may be used in



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FIGURE 1. Photographic Stations at Rancho Seco, CA, Nuclear Generation Site



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FIGURE 2. Topcon 35 mm Camera on Transit, and 16 mm Bolex Movie Camera in Normal Operating Mode

this way to greatly reduce the amount of light reaching the film. We used an N.D. 3 filter, which reduces light transmission by a factor of 1000. Time exposures were made each hour on the hour synchronized at the two sites by stopwatch and radio.

The time-exposed photos of the plume result in a time-averaged integrated picture that can be analyzed to permit dimensioning of the plume boundaries or centerline. Since the real plume is rarely in a plane parallel to the photographic film plane, a method must be devised to allow reconstruction of plume bounds in three dimensions from a two-dimensional photograph. Niemann, et al.,² describes in considerable detail a method of analyzing plume photographs when the plume is at an angle to the camera film plane. With two camera stations at right angles to each other, the angle between the plume and one of the film planes is never greater than 45° . It is essential that an accurate record be kept of the exact orientation of the camera lens axis for each photographic period. Aligning the camera lens axis with the axis of the lens system of its transit mount permits precise orientation within tenths of a degree.

Crucial to each analysis is a measurement of the mean wind direction during the time of the photographic exposure. Experiments were normally conducted between 0900 PST and 1500 PST. Double theodolite pibals with 30-sec readings of azimuth and elevation angles were taken at 1000, 1200, 1300 and 1500 PST. At 1100 and 1400 PST rawinsondes were taken to provide temperature and relative humidity profiles in addition to wind information.

ANALYSIS

Analysis was performed on the photograph from the site whose view was more nearly normal to the plane of the plume as determined by the measured wind direction.

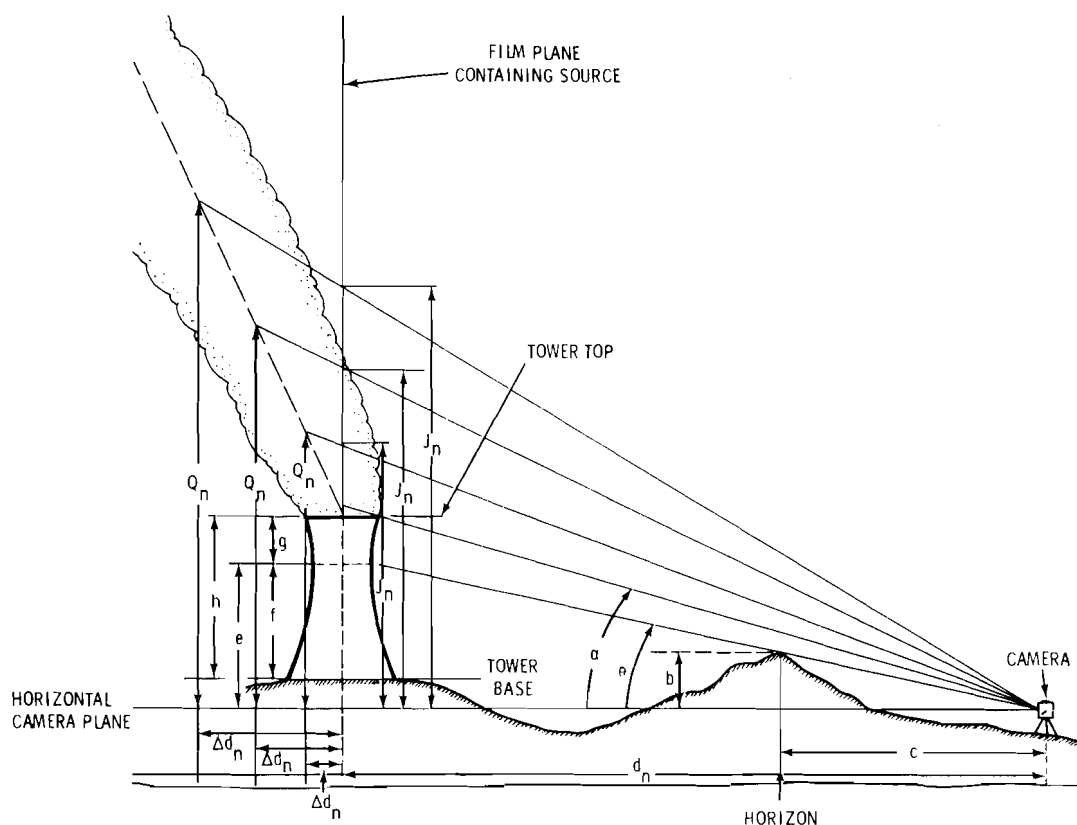
The distance from the camera to the center of the tower must be determined from the map of the area or from a survey. Depending on the strength of the wind, the camera was oriented with its lens axis either through the cooling tower's centerline or some mea-

sured angular distance downwind. These two situations require different sets of equations for reconstructing the location of the plume above the horizontal plane through the camera. At the Rancho Seco site, there are two cooling towers, their centers 141.7 m (465 ft) apart on an east-west line. For simplicity in dimensioning the plumes, they were treated as a single plume emitted from an imaginary tower located midway between the actual towers. In the plume dimensioning procedures, this imaginary tower is considered to be the source of a single plume, and all calculations are referenced to its location.

In order to identify portions of the plume for measurement, a grid is prepared on transparent or semitransparent overlay on the photographic print. The grid is calibrated in degrees of elevation vertically and in linear distance horizontally. The vertical calibration geometry is shown in Figure 3. Table 1 summarizes the necessary input data required for establishing the grid.

Using the horizontal camera plane as 0° elevation, the vertical grid was drawn on the transparent overlay at a spacing of mm/vertical degree with an assumed linearity within each degree of elevation. Parallax at the top edge of the photo has not been considered. Photographs made with the 50 mm and 35 mm lenses on the cameras used in these experiments indicated very little distortion in either the vertical or horizontal.

The horizontal calibration geometry is illustrated in Figure 4. This diagram applies only when the camera lens axis passes through some point downwind of the cooling towers. On days with very light winds, the camera was oriented with its axis through the cooling towers with resultant horizontal geometry as in Figure 5. The location of the vertical centerline of the camera field of view is located on the photoprint. Then, the horizontal increments, O , are measured to the right and left of this line in the general case when the camera axis points downwind of the source; and to the downwind side only, when the camera axis passes through the source. The distance from camera axis to cooling tower in the plane through the source and parallel to the film plane

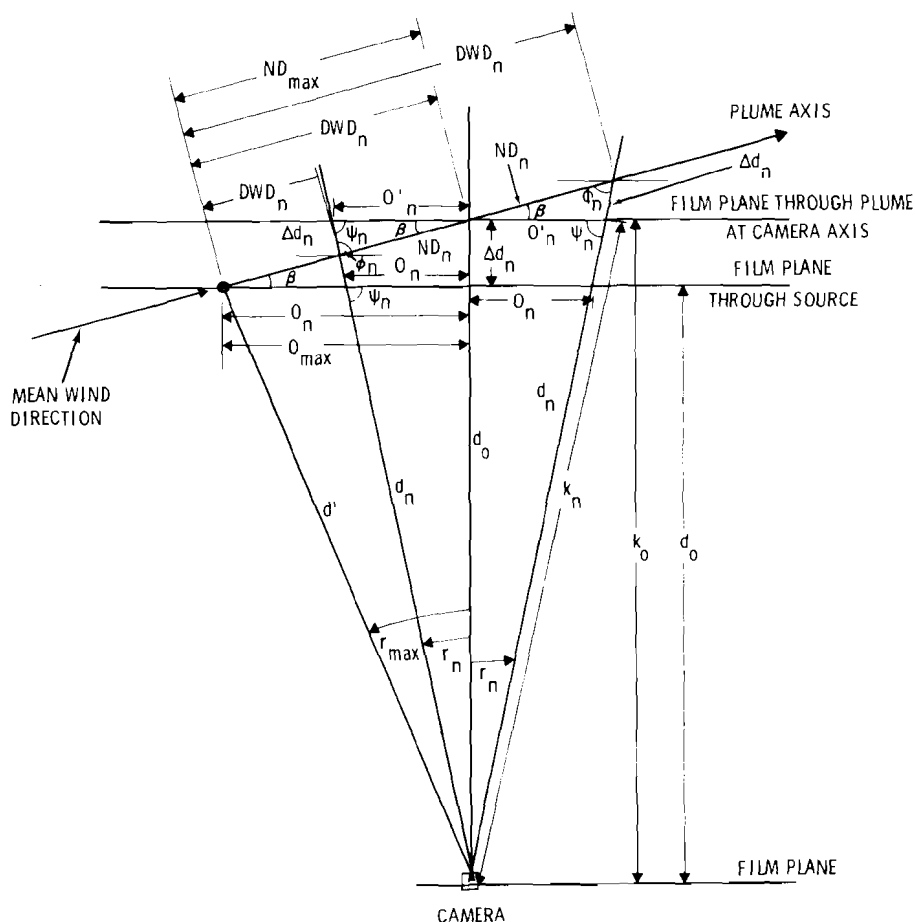


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FIGURE 3. Plume Elevation Geometry

TABLE 1. Data Needed to Establish Location of Plume

	CAMERA SITE 1-A	CAMERA SITE 2
Cooling Tower Height	129.5 m (425 ft)	129.5 m (425 ft)
Camera Elevation (interpolated from 5 ft contours, U.S.G.S. 1:24,000 topo map)	52.7 m (173 ft) MSL	33.5 m (110 ft) MSL
Height of Horizon (determined from U.S.G.S. 1:24,000 topo map)	61 m (200 ft) MSL	47.2 m (155 ft) MSL
Tower Base Elevation (estimated from U.S.G.S. 1:24,000 topo map)	55.5 m (182 ft) MSL	55.5 m (182 ft) MSL
Distance, Camera to Tower (measured from U.S.G.S. 1:24,000 topo map)	4140.2 m (13583 ft)	4033.5 m (13233 ft)
Visible Tower Height	101 m (332 ft)	110.6 m (363 ft)
Angular Elevation of Tower Top	1.83°	2.15°

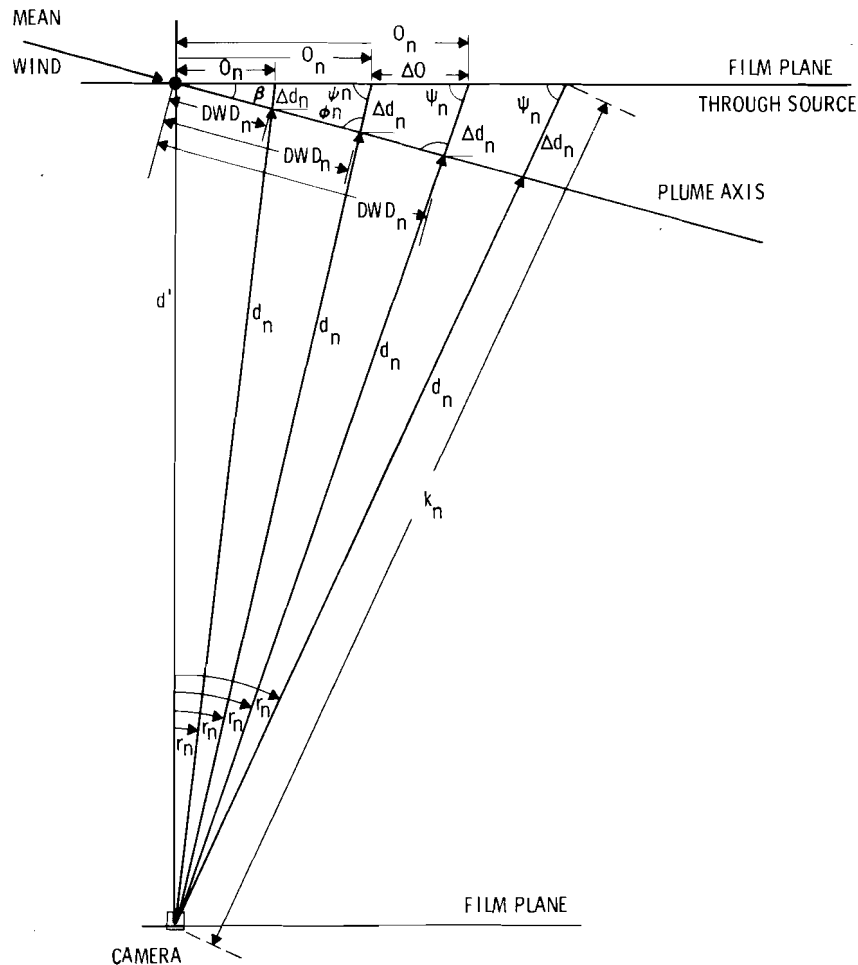


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FIGURE 4. Horizontal Geometry for Camera Axis Downwind from Source (After Niemann, et al., 1970)

is divided into convenient increments; we have used 61 m (200 ft). Figure 6 shows a typical photograph resulting from the 300-sec exposure and the gridded overlay with horizontal lines spaced 1° of elevation apart and the vertical lines spaced 61 scale meters apart. The outline of the plume is traced onto the overlay. Depending on the area or areas of interest, one can now identify any portion of the plume by a vertical angle J_n and a horizontal distance from the camera axis O_n .

To measure plume dimensions at this stage is only possible in the rare situation where the mean wind direction for the time period encompassed by the photograph is exactly parallel to the film plane. This situation never occurred during this series of experiments. Dual theodolite pibals with 30-sec readings were released when the 300-sec photographic exposure was begun and those pibal winds were taken as the mean wind direction for the photo period. There is certainly



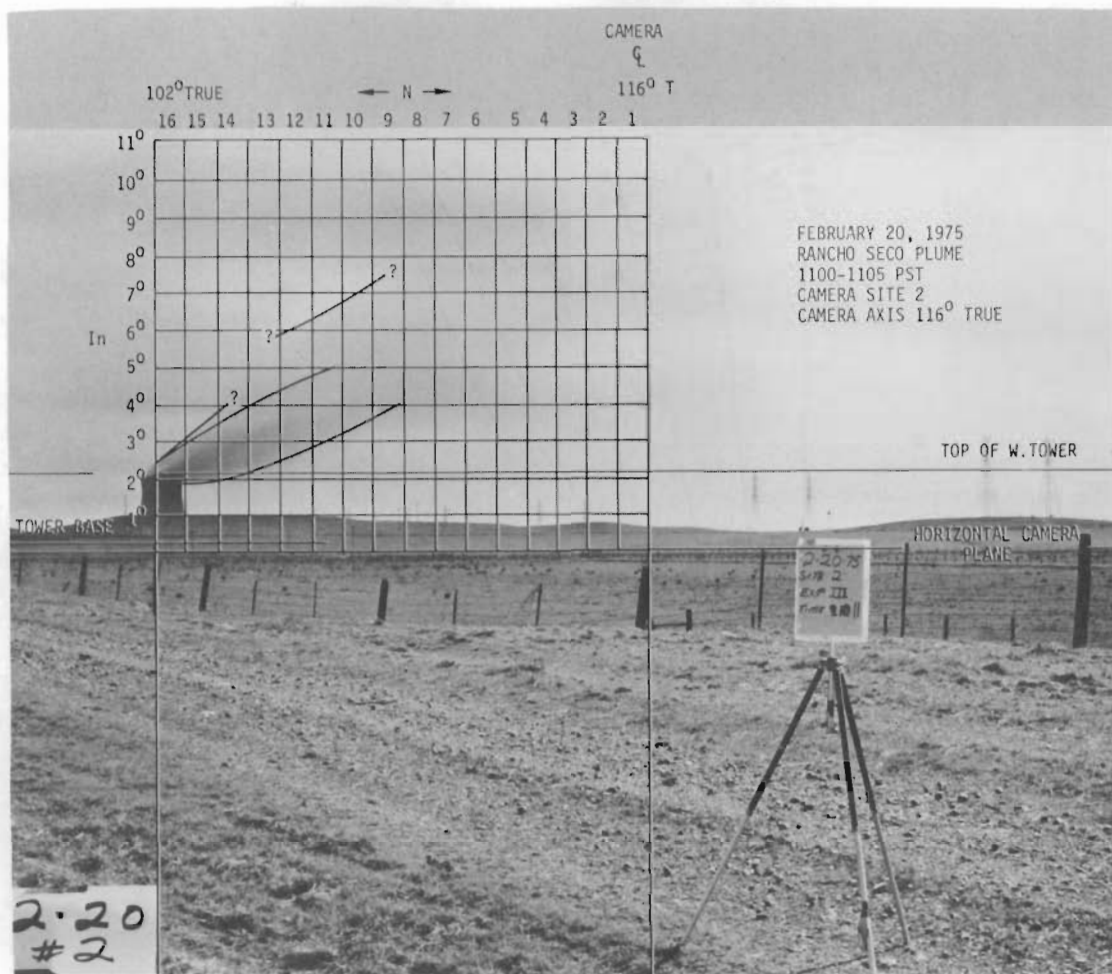
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FIGURE 5. Horizontal Geometry for Camera Axis Through Source

a possible error here, especially if atmospheric conditions are in a transition stage (breakup of an inversion, frontal passage, strong wind shear, etc.). In this series of experiments, wind shear was the most frequent problem and in such cases the plume has not been analyzed above the level of wind shear.

The angle between the film plane and the path of the plume is called β (refer to Figures 4 and 5). It can be seen that the plume calculation geometry is slightly different when the

camera axis passes through the source (Figure 5) compared with the case when it passes downwind of the source (Figure 4). In the latter situation, there are also some variations in calculations of the plume incremental distances upwind of the camera axis versus downwind of the camera axis. In addition, these variations are reversed in the cases when the plume is coming toward the camera from that when the plume is moving away from the camera. For simplicity, we have followed Niemann's nomenclature in the geometric calculations to determine:



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FIGURE 6. Cooling Tower Plume Photography with Scaled Grid Overlay

r_n The angle from camera axis to the increments O_n, O'_n or ND_n O_n The specified horizontal distance increment in the film plane through the source O'_n The projection of O_n on the film plane through the plume DWD_n The incremental downwind distances of the plume from the source ND_n The incremental plume length swept out by angle r_n ND_{max} The true plume length between source and camera axis	O_{max} Horizontal distance from camera axis to source in the film plane through source d_n Distance from film plane to parallel plane through source k_n Distance from film plane to parallel plane through plume When the camera axis is through the tower, the definitions of d and k are not changed. For the case where the camera axis is downwind of the source, the following calculations were made: β, r_{max}, O_{max}, d_0, Δd_0, k_0, ψ_n and ϕ_n are calculated from basic information of mean wind direction, map

measurements and photographic notes
(in the case of $r_{\max}$).

$$O'_{\max} = k_o \tan r_n \quad (1)$$

$$ND_{\max} = (\Delta d_o^2 + O_{\max}^2)^{1/2} \quad (2)$$

$$\psi_n = \tan^{-1} \frac{d_o}{O_n} \quad (3)$$

$$r_n = \tan^{-1} \frac{O_n}{d_o} \quad (4)$$

$$O'_n = k_o \tan r_n \quad (5)$$

$$\phi_n = 180^\circ - (\psi_n + \beta) \text{ [upwind from the vertical nadir plane]} \quad (6)$$

$$\phi_n = \psi_n - \beta \text{ [downwind from the vertical nadir plane]} \quad (7)$$

Equations (6) and (7) apply when the mean wind is carrying the plume away from the camera; when it brings the plume toward the camera (6) and (7) are to be reversed, i.e., (7) applies to upwind portion of plume and (6) to downwind.

For the upwind portion of the plume, from the source to the camera axis, true incremental downwind distances are given by

$$DWD_n = ND_{\max} - ND_n, \quad (8)$$

and the true incremental distances downwind from the camera axis are determined by

$$DWD_n = ND_{\max} + ND_n. \quad (9)$$

From the above, calculate

$$k_n = O'_n / \sin r_n \quad (10)$$

$$\Delta d_n = O'_n \sin \beta / \sin \phi_n \quad (11)$$

$$d_n = k_n \mp \Delta d_n \text{ [upwind-,downwind+]} \quad (12)$$

From the photo, grid values of J_n for the height at the top of the plume, Q_{top} , and bottom of the plume, Q_{bottom} , are read directly and inserted into

$$Q_n = d_n \tan J_n \quad (13)$$

to obtain heights of the top and bottom of the plume at each of the horizontal increments O_n (Figure 6). The intervals of O_n and J_n could be reduced for finer resolution of the plume dimensions.

When camera axis was oriented on the source, the following calculations were used: (Refer to Figure 5).

O_n is specified 61 m (200 ft) here, β is determined as previously

$$r_n = \tan^{-1} \frac{O_n}{d'} \quad \left[\begin{array}{l} d' = \text{distance} \\ \text{to tower} \end{array} \right] \quad (14)$$

$$\psi_n = \tan^{-1} \frac{d'}{O_n} \quad (15)$$

$$DWD_n = \frac{O_n \sin (180^\circ - \psi_n)}{\sin \phi_n} \text{ [plume away]} \quad (16)$$

$$DWD_n = \frac{O_n \sin \psi_n}{\sin \phi_n} \text{ [plume toward]} \quad (17)$$

$$\Delta d_n = \frac{O_n \sin \beta}{\sin \phi_n} \quad (18)$$

$$k_n = \frac{O_n}{\sin r_n} \quad (19)$$

$$d_n = k_n \mp \Delta d_n \text{ [toward-,away+]} \quad (20)$$

Again the plume dimensions are calculated by

$$Q_n = d_n \tan J_n$$

CONCLUSIONS

With careful attention to determination of camera location and orientation, and calculation of mean wind direction, it is possible to rather accurately estimate dimensions of the plume from time-exposed photographs which show the plume at an angle of 45° or less to the film plane. This series of experiments used 2 cameras

with their axes $\sim 75^\circ$ from each other (the ideal would be 90°), which were mounted on transit heads to permit precise orientation. Aircraft observations will provide some verification of these purely geometrical measurements of the plume dimensions. The time lapse movies with a frame exposed every 30 sec will give information on major fluctuations in the plume behavior during the 300-sec photographic period. Further experiments of this type should include time exposures more frequent than 1/hr; 2 time-exposed photographs with accompanying pibals made every half hour would enhance significantly the probability of good photographic coverage. During one of our experiments, a brief foggy period coincided with a photographic period and an hour later the plume had decreased noticeably in size from evaporation. A photograph made on

the half hour would have been very valuable here.

It may eventually be possible to incorporate the information in both hourly photographs as long as the plume is not transported directly towards or away from one camera. The major difficulty at present seems to stem from the fact that in a time-integrated photograph there are no readily identifiable plume features to allow established stereo photogram-metric techniques to be applied.

ACKNOWLEDGEMENTS

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NATURAL DRAFT COOLING TOWER PLUME CHARACTERISTICS
DETERMINED WITH AIRBORNE INSTRUMENTATION

M. A. Wolf

A field investigation of natural draft cooling tower plumes was conducted at the Rancho Seco Nuclear Generating Station near Sacramento, California, during February 1974. Measurements during penetrations of the plumes with an instrumented aircraft on 3 days are reported. It is concluded that the internal characteristics of cooling tower plumes can be defined with a properly instrumented aircraft.

INTRODUCTION

It is increasingly probable that future power production will concentrate 10-25 power units in nuclear energy centers which will release up to 100,000 MW of waste energy to the atmosphere from cooling towers. The possible implications to the environment of this release--principally as latent heat--have been reviewed by Hanna and Gifford¹ who note that current knowledge is inadequate to resolve many uncertainties.

We sought to acquire experimental data, providing increased definition of cooling tower plume characteristics so that the environmental impact may be specified more precisely. The initial phase was performed at the Rancho Seco Nuclear Generating Station near Sacramento, California, in February 1975. Rancho Seco, operated by the Sacramento Municipal Utility District, is a 913 MWe facility which dissipates its waste heat from two 130 m natural draft cooling towers. Measurements within the plumes and the adjacent ambient air were obtained with an instrumented Cessna 411 aircraft. The results presented here were derived from analysis of flights on 3 days of the 5-day field operation.

FIELD OPERATIONS

The aircraft penetrated the plume on each experimental day, measuring droplet spectra, relative humidity,

temperature and Aitken nuclei concentration at a variety of altitudes and distances. Penetrations ranged from less than 1 sec to about 10 sec; generally 2-4 sec in duration. Fast instrument response, therefore, is needed to delineate internal characteristics of the plume. The instruments used do not all have the same response time. Perhaps the least plume definition is provided by the Aitken nuclei counter, which has a 1-2 sec sampling cycle. Additional work is required to establish response times for the thermistor-bead temperature sensor and the carbon-strip humidity sensor in their housing. However, the response of both was judged to be satisfactory. There was evidence of depressed temperatures after plume passage, which suggests a wet-bulb effect that would cause lower indicated temperatures in an unsaturated plume as well. Similarly, the indicated humidities may be too low due to an apparent reduced response at high humidities. It is somewhat disconcerting that the maximum relative humidity observed was 83%. The sampling rate of the droplet size spectrometer, which previously had been noted as inadequate,² has been increased by a factor of 16 by substituting an incremental magnetic tape recorder for the printer originally used.

One objective of the field study was to observe differences in the condensation droplet spectra under

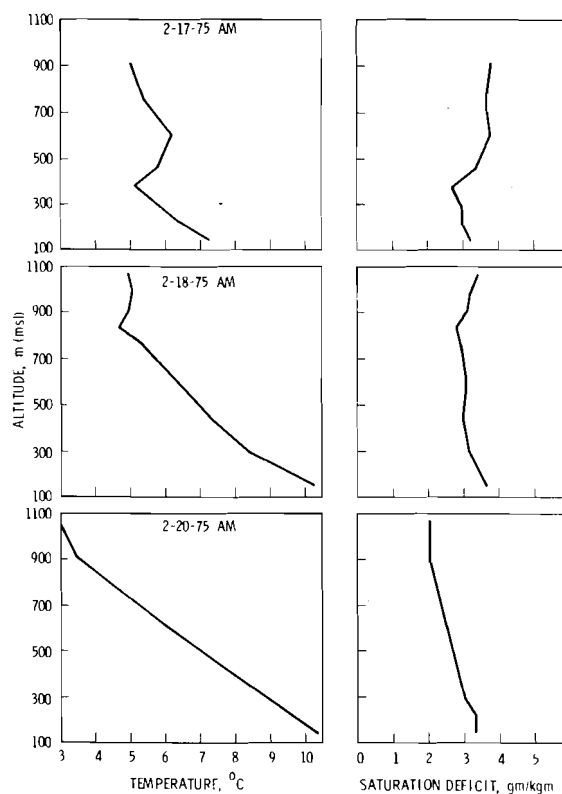
various atmospheric and plant operating conditions and at several heights above and distances from the cooling tower exits. However, there was not a wide range of plant or atmospheric conditions during the experimental period.

On the 3 days considered here--February 17, 18 and 20, 1974--the plant operated at 635-850 MWe range. The vertical profiles of temperature and saturation deficit taken during aircraft soundings on each day are shown in Figure 1. Perhaps the wind demonstrated the greatest variability. At cooling tower exit height, the wind speeds determined from pilot balloon tracking were 4.0, 2.5 and 8.7 m sec⁻¹ on February 17, 18 and 20, respectively. Wind directions below 915 m (3,000 ft) msl on the 18th were southerly whereas they were northerly above that height and at all heights on the other two days. Plume persistence on these days was such that the intercepts were all below 1220 m (4,000 ft) msl and within about 1 km from the tower. Minimum operating altitude over the plant of 305 m (1,000 ft) msl prevented plume sampling closer than 122 m (400 ft) from the exits.

RESULTS

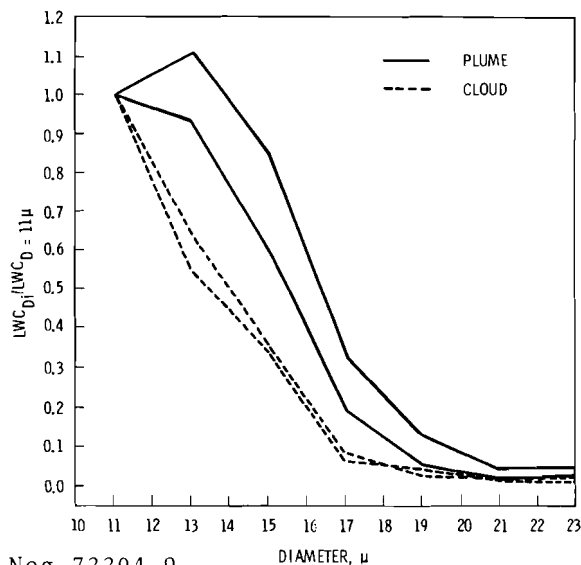
As shown in Figure 1, the highest humidity occurred on February 20; there were low-level fair weather cumulus on that day. Figure 2 shows liquid water content (LWC) distributions from two intercepts of the natural clouds and two of the cooling tower plume at the same height. The LWC is determined by summing the products of droplet concentration and their volume. The cloud droplet spectrometer was operated at 2-30 μ for those intercepts. Due to instrumentation misadjustment, counts in the first 4 channels (2-8 μ) were lost and only the large-drop tail is available. Counts of adjacent channels have been averaged for smoothing. The LWC in each interval has been normalized by the content in the 10-12 μ interval. It is observed that the cooling tower plume contains a relatively higher proportion of water in the larger drop diameters than does the natural cloud.

The left graph of Figure 3 shows the LWC distributions at a number of



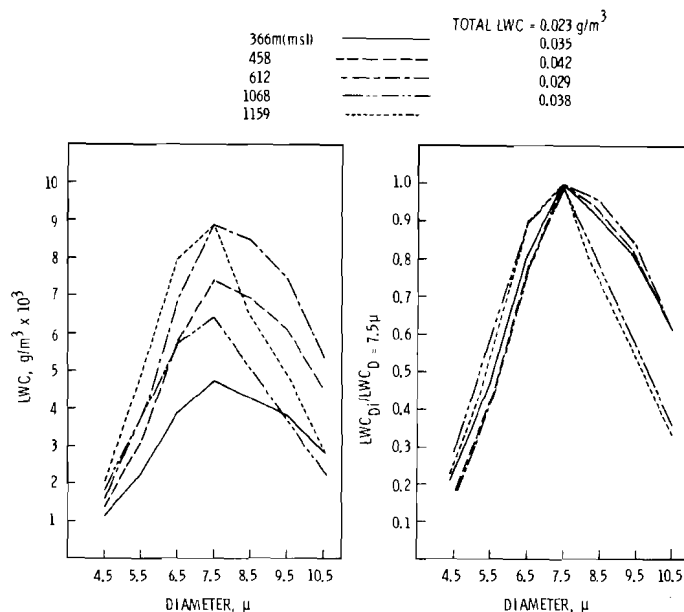
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FIGURE 1. Vertical Profiles of Temperature and Saturation Deficit



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FIGURE 2. Liquid Water Distributions in Plumes and Clouds; 610 m (msl), 2-20-75 am

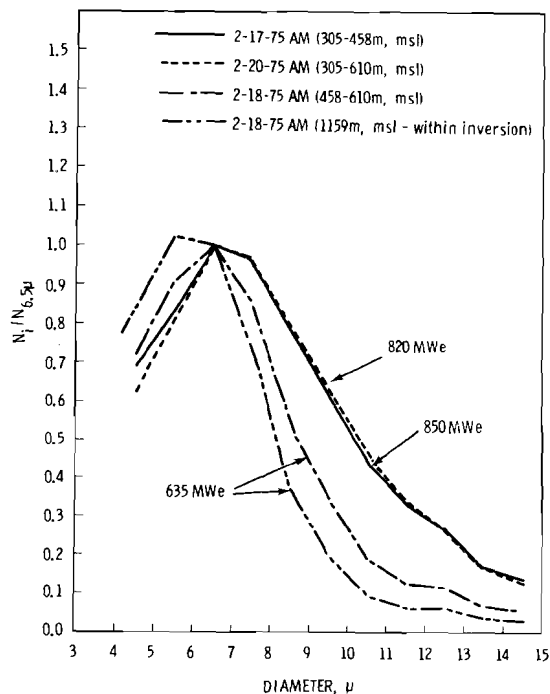


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FIGURE 3. Liquid Water Distributions, 2-18-75 am.

heights above the tower exit on February 18. Heights above cooling tower exit are determined by subtracting 180 m from the values shown. It is not apparent from the values of total LWC shown in the 4-11 μ diameter range that LWC is related to height. However, the plume centerlines may not have been penetrated in each intercept and, therefore, the LWC values may not be comparable. On the right of Figure 3, the LWC of each interval is normalized by the content of the peak interval. This shows that the liquid water content is shifted to smaller diameters at the uppermost levels; Figure 1 shows these above the inversion base in a region of increased saturation deficit. Apparently evaporation is occurring at these levels.

Figure 4 compares the droplet spectra observed on the 3 days. Individual spectra at heights below 610 m (2,000 ft) msl were examined on each day and no dependence on height was observed. Thus, the daily spectra were averaged and normalized by referring the number of droplets in each interval to the interval which generally contained the greatest



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FIGURE 4. Plume Droplet Spectra Versus Reactor Power Level

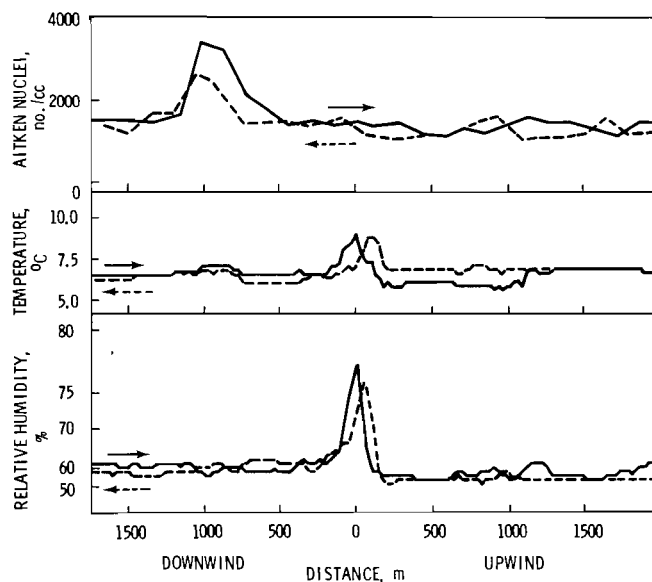
number--6-7 μ . The spectra for February 17 and 20 are nearly identical, but that for February 18 contains a substantially smaller proportion of droplets in the larger diameter intervals. The previously noted additional decrease in large droplets at greater heights on February 18, attributed to evaporation, is also shown here.

A probably cause for the smaller droplet diameters is the reduced plant power output on the 18th. February 17 and 20 outputs were 850 and 820 MWe, respectively, whereas the February 18 output was only 635 MWe. The proportionate reduction in waste heat release reduced the water vapor flux from the towers by 25% and that, in turn, could account for the impaired droplet growth.

Figures 5-8 are profiles of relative humidity, temperature and Aitken nuclei concentration for plume traverses at 458, 610, 915 and 1068 m (1500, 2000, 3000 and 3500 ft), msl, respectively on February 18. The abscissa shows upwind and downwind distances relative to one of the plume intercepts. At three levels, traverses were made in both directions and the paired profiles shown

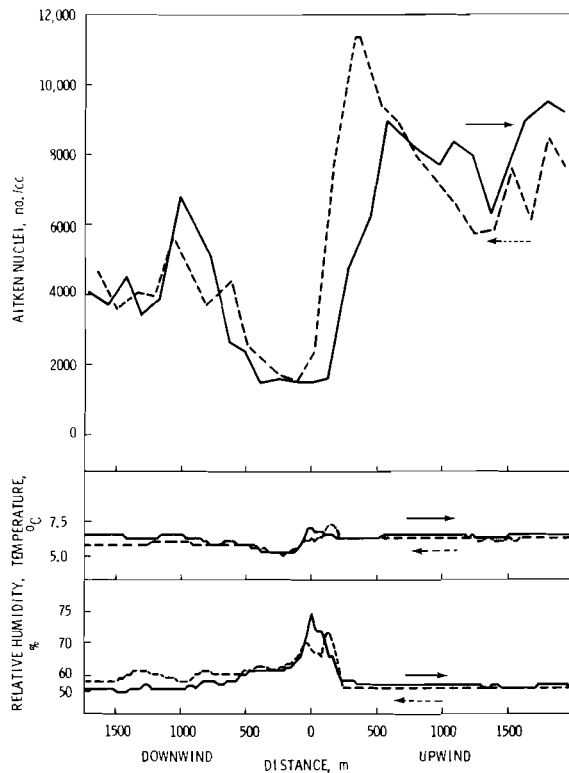
substantial agreement. Figure 9 shows the vertical distribution of Aitken nuclei determined on all 3 days during the sounding portion of each flight. The profile on the 18th differs from the others; it has the lowest concentration of the three near the ground and the highest of the three above 458 m. The sharp decrease in nuclei concentration above 915 m coincides with the inversion base.

At 458 m, the plume intercept is clearly shown in Figure 5 by the increased temperature and relative humidity. Here and elsewhere it is observed that the plume, though visible, was not saturated. Provided there was no major instrumental error, it must be assumed that the plume was well-mixed with ambient air and in the process of evaporation at all altitudes. The plume location is not apparent in the nuclei concentration indicating that concentrations are equal in the plume and in the ambient air. The downwind peaks suggest a vertical circulation that has transported the higher concentration from above. The relative humidity profiles both show higher humidities downwind than upwind in the clear



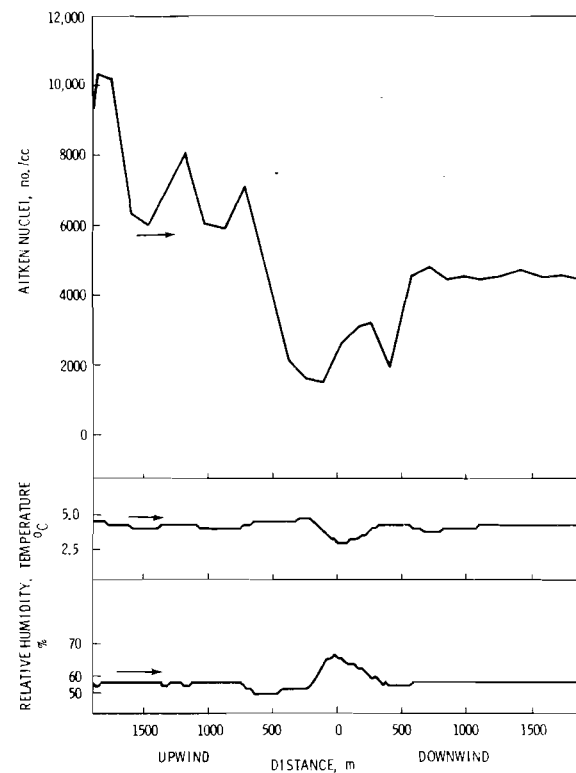
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FIGURE 5. Temperature, Humidity and Nuclei Concentration Profiles; 458 m (msl), 2-18-75 am



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FIGURE 6. Temperature, Humidity and Nuclei Concentration Profiles; 610 m (msl), 2-18-75 am



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FIGURE 7. Temperature, Humidity and Nuclei Concentration Profiles; 915 m (msl), 2-18-75 am

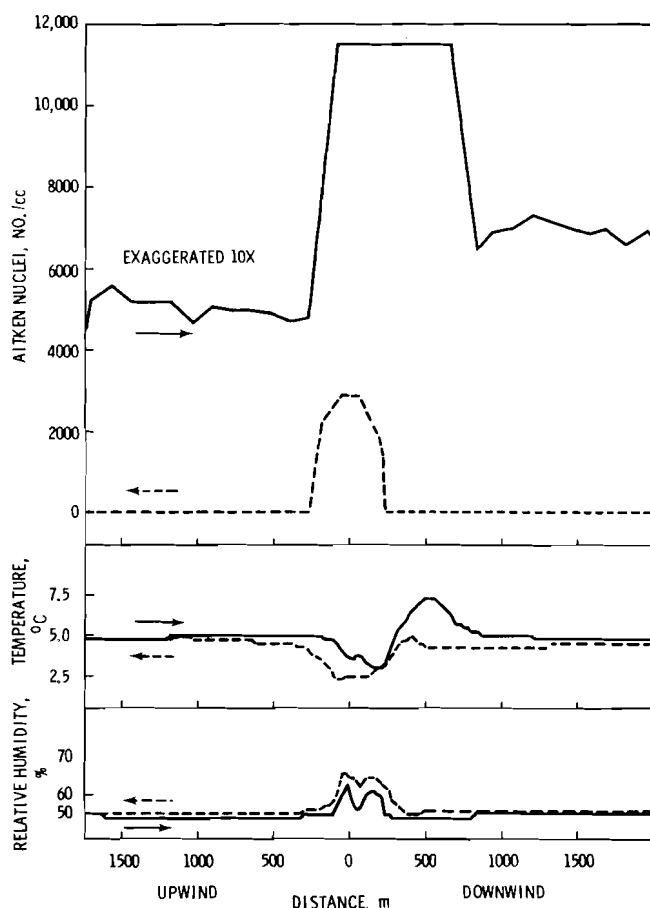
air, presumably due to moisture contributed by the plume. The temperature profiles, however, show decreases after plume penetration in both directions which suggest wetting of the sensor and a lowering because of the wet-bulb effect.

Figures 6 and 7 show higher nuclei concentrations in the ambient air than was observed at 458 m. At both 610 and 915 m, downwind concentrations were significantly lower than those upwind of the plume. Perhaps this is due to mixing of the lower elevation air having low nuclei concentration with the higher concentrations at these altitudes. The entrainment of air from below can be observed by the increased concentrations in the plume at the various heights. At 610 m, the in-plume concentration of 1500 cc^{-1} coincides with observed concentrations at 458 m. At 915 m the in-plume con-

centrations increase to between 1500 and 3000 cc^{-1} , reflecting the entrainment of higher concentrations in the ambient air below that height.

The relative humidity and temperature profiles at 610 m, shown by Figure 6, indicate increased humidity and decreased temperature downwind of the plume which suggest that plume evaporation is influencing downwind conditions. At 915 and 1068 m, evaporation is apparent within the plume by the obvious temperature decrease. The sharp increase in temperature downwind of one plume penetration is unexplained though it coincides with the broad peak of Aitken nuclei concentration.

The nuclei concentrations in the plumes at 1068 m are well above ambient air concentrations so that downwind values are increased relative to the upwind values. This



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FIGURE 8. Temperature, Humidity and Nuclei Concentration Profiles; 1068 m (msl), 2-18-75 am

downwind increase is the reverse of the effect observed at 610 and 915 m but clearly shows again that cooling tower plumes are effective in vertical transport of pollutants. In this case the plume penetrated the inversion and introduced pollutant concentrations higher than ambient by a factor of 6.

Although, as noted previously, the in-plume temperatures and relative humidities may be somewhat low due to sensor response, the derived mixing ratios are considered reasonably representative. Consequently, the change of mixing ratio with height was used to indicate the dilution of the plume. Figure 10 presents the dilution as a function of height for the 3 days. The dilution is defined as the ratio of mixing ratio difference between plume and

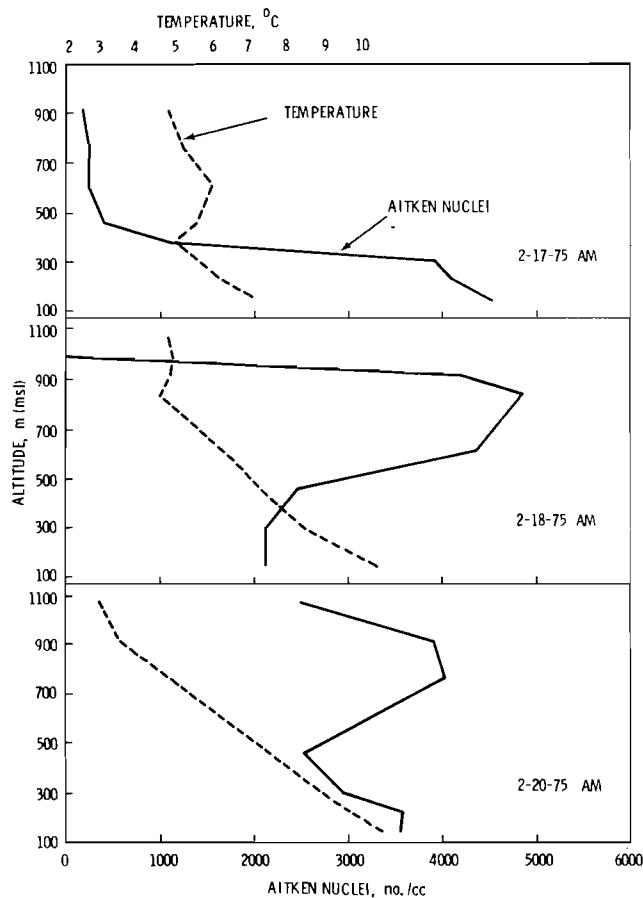
ambient air at the cooling tower exit to the difference at various normalized heights. Normalized height is the product of the number of cooling tower diameters above the exit and the ratio of the mean wind speed at the exit to the efflux velocity. The least-square fit to the data was determined as

$$D = 2.05H^{1.22}$$

where D is dilution and H is the normalized height.

CONCLUSIONS

The internal characteristics of cooling tower plumes can be defined with a properly instrumented aircraft. For this application rapid response



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FIGURE 9. Vertical Profiles of Aitken Nuclei Concentration

instrumentation are essential. Furthermore, instrument calibration must be performed under similar exposure conditions--rapid, large changes in values--to assure reliable measurements.

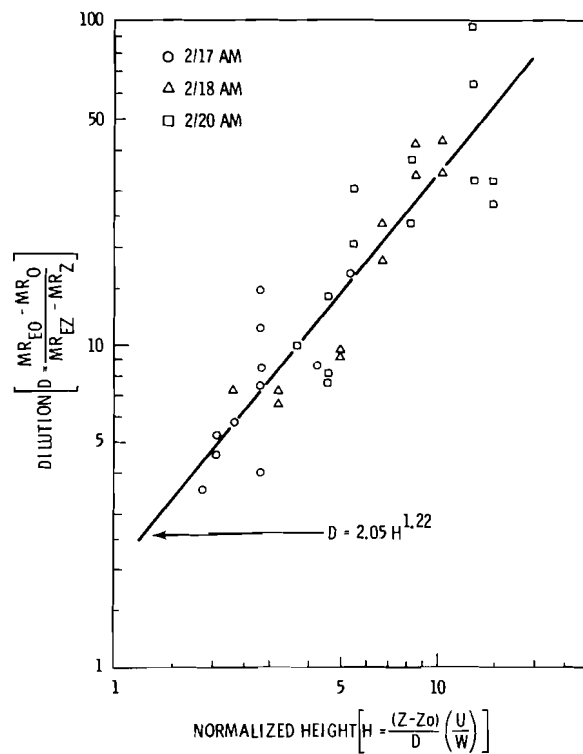
This initial phase of the study has shown:

1. Penetration of an inversion layer by the plume with accompanying transport of pollutant from lower levels.
2. Above the inversion, plume temperatures were noticeably cooler than the ambient air, presumably due to evaporation which also was apparent by the shift of droplet spectra to smaller diameters.

3. The proportion of larger droplet diameters increased with the level of heat rejection from the towers.
4. The relationships of plume dilution by ambient air to the height above the exit was dependent on the ratio of wind speed and exit velocity.

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FIGURE 10. Plume Dilution with Height

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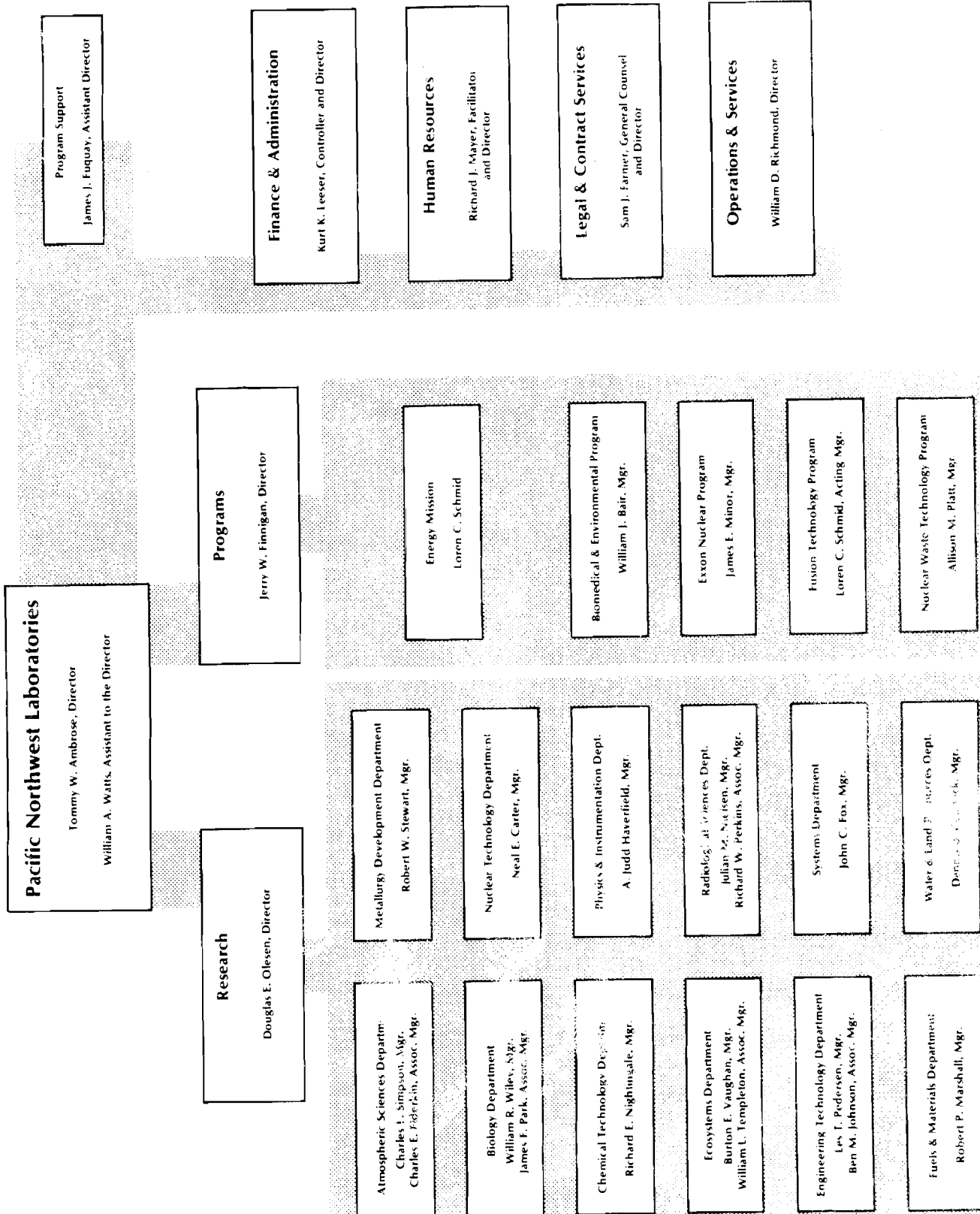
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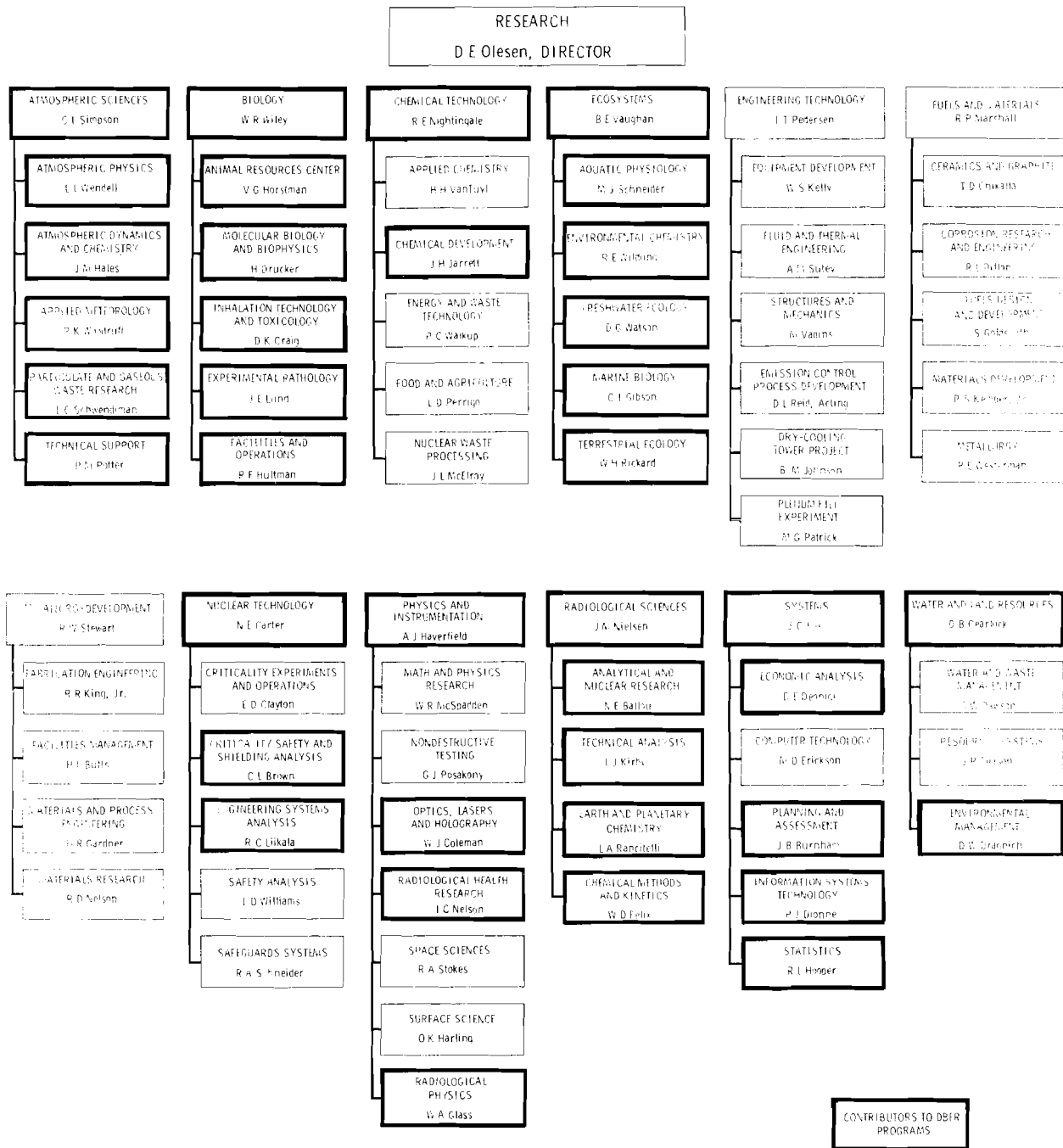
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