Pacific Northwest Laboratory
Annual Report for 1977
to the DOE Assistant Secretary
for Environment

February 1978

Part 3

Atmospheric Sciences

Prepared for the
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Battelle
Pacific Northwest Laboratories

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**Pacific Northwest Laboratory
Annual Report for 1977
to the
DOE Assistant Secretary for
Environment
Part 3 Atmospheric Sciences**

**by
C. L. Simpson and Staff Members
of Pacific Northwest Laboratory**

February 1978

**Battelle
Pacific Northwest Laboratories
Richland, Washington 99352**

PREFACE

The 1977 Annual Report from Pacific Northwest Laboratory (PNL) to the DOE Assistant Secretary for Environment introduces a new cover. The earth-green color used on past annual reports has been replaced by the "environmental colors," blue and green. The cover's abstract design is not intended to represent anything specific, but we would not be unhappy if it suggests something environmental or biological to the reader. The blue and green color pattern on the cover is different for each part of this report to help distinguish the five parts.

The five parts of the Report are oriented to particular segments of our program. Parts 1-4 report research performed for the DOE Office of Biomedical and Environmental Research. Part 5 reports progress on all other research performed for the Assistant Secretary for Environment, including the Office of Environmental Control Technology, Office of Technology Impact, and Office of Operational and Environmental Safety.

Each part consists of project reports authored by scientists from several PNL research departments, reflecting the interdisciplinary nature of the research effort. Parts 1-4 are organized primarily by energy technology, although it is recognized that much of the research performed at PNL is applicable to more than one energy technology.

The parts of the 1977 Annual Report are:

Part 1: Biomedical Sciences

Program Manager - W. R. Wiley	R. C. Thompson, Report Coordinator D. L. Felton, Editor
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Part 2: Ecological Sciences

Program Manager - B. E. Vaughan	B. E. Vaughan, Report Coordinator J. L. Helbling, Editor
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Part 3: Atmospheric Sciences

Program Manager - C. L. Simpson	R. L. Drake, Report Coordinator C. M. Gilchrist, Editor
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Part 4: Physical Sciences

Program Manager - J. M. Nielsen	J. M. Nielsen, Report Coordinator G. M. Garnant/L. Carson, Editors
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Part 5: Control Technology, Overview, Health, Safety,
and Policy Analysis

Program Managers - N. E. Carter
D. B. Cearlock
J. C. Fox
D. L. Hessel
H. V. Larson
S. Marks
W. J. Bair, Report Coordinator
R. W. Baalman, Editor

Activities of the scientists whose work is described in this Annual Report are broader in scope than the articles indicate. Knowledge and experience obtained by PNL staff in carrying out research in the Environment, Health, and Safety Research program have contributed to many other DOE interests. These include assistance in the preparation of several Environmental Development Plans for the Assistant Secretary for Environment, preparation of environmental statements for which the Laboratory is responsible, key membership in several national and international organizations, and numerous responses to the media on research projects of public interest.

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

Previous Reports in this Series:

Annual Report for

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1954	HW-30306, HW-33128, HW-35905, HW-35917
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1963	HW-80500, HW-81746
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1965	BNWL-280, BNWL-235, Vol. 1-4, BNWL-361
1966	BNWL-480, Vol. 1, BNWL-481, Vol. 2, Pt 1-4
1967	BNWL-714, Vol. 1, BNWL-715, Vol. 2, Pt 1-4
1968	BNWL-1050, Vol. 1, Pt. 1-2, BNWL-1051, Vol. 2, Pt. 1-3
1969	BNWL-1306, Vol. 1, Pt. 1-2, BNWL-1307, Vol. 2, Pt. 1-3
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1971	BNWL-1650, Vol. 1, Pt. 1-2, BNWL-1651, Vol. 2, Pt. 1-2
1972	BNWL-1750, Vol. 1, Pt. 1-2, BNWL-1751, Vol. 2, Pt. 1-2
1973	BNWL-1850, Pt. 1-4
1974	BNWL-1950, Pt. 1-4
1975	BNWL-2000, Pt. 1-4
1976	BNWL-2100, Pt. 1-5

FOREWORD

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

COAL, GAS AND OIL COMBUSTION

The behavior of air pollution resulting from fossil-fuel power plants is being explained. Involved in making this explanation are these factors: the type of pollutants emitted, their transport and diffusion in the air, their physical and chemical transformations during transport, their removal by wet and dry scavenging processes, and their impacts on climate, bodies of water and living species. Since the result of the current and projected coal utilization is the release of large quantities of sulfur dioxide to the atmosphere, current atmospheric research is being conducted primarily in the Multi-State Atmospheric Power Production Pollution Studies (MAP3S).

FISSION AND FUSION

Concern about long-lived particulates (i.e., plutonium and other radionuclides) released to the environment from fission and fusion plants dictates that the deposition and resuspension of these substances must be studied. For example, because the primary hazard from plutonium is inhalation, its residence in the atmosphere must be clearly defined. Current research, therefore, is evaluating the removal of particulates from the atmosphere by deposition (which limits initial exposure) and any future resuspension from the surface (which continues the potential for inhalation). In addition, the local and regional climatic impacts of heat and moisture releases from large energy centers (several power plants in close proximity) are being assessed.

SOLAR

The potential for solar energy development depends on the adequate assessment of ground-based insolation and the determination of the reduction of solar radiation due to atmospheric turbidity. Modest research activities are being conducted in these two areas.

OIL SHALE

The mountainous oil shale regions of Colorado, Utah and Wyoming present a particularly difficult air pollution problem because air may be trapped in the valleys of these regions for extended periods of time under certain meteorological conditions. Especially stringent siting requirements, therefore, must be fulfilled to meet State and Federal air quality standards. Adequate models and field measurements of the complex air flow and dispersion conditions in this complex area are not available; they must be developed to assure acceptable siting of oil shale facilities. PNL has undertaken the assessment of the requirements for the proper modeling activities and field measurement techniques that will be utilized in this very important area of research.

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

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

This report describes the progress in FY-1977 for each of these technologies. A divider page summarizes the goals of each area and lists project 189 titles that fund research in the technology as bulleted items.

R. L. Drake
Program Coordinator

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1.0

Coal,

Gas and Oil

COAL, GAS AND OIL

- **Aerosol and Trace Gas Transformations**
- **Precipitation Scavenging in MAP3S**
- **MAP3S Modeling Studies**
- **Atmospheric Boundary Layer Studies**
- **Particle Resuspension and Translocation**
- **Urban Pollutant Characterization, Transport and Deposition**
- **Coal Conversion Pollutant Chemistry**
- **Regional Studies**
- **DOE/RL Special Studies**

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

The research activities at PNL are basically related to the MAP3S Program being conducted in the Northeast Quadrant of the U.S. They include the data analysis and flight operations based at Muskegon, Michigan; the precipitation chemistry network in the eastern mountains and coastal areas; the incloud scavenging studies over Lake Michigan; the laboratory analysis of sulfur and nitrogen compounds and trace metals at Hanford; and the trajectory modeling studies being formulated at Hanford. In addition to this work, PNL is conducting extensive field and modeling studies concerned with the diffusion, transformation and deposition of pollutants. The field activities are being conducted in the St. Louis, Milwaukee and Chicago areas, as well as in the Northwest and other areas in the Midwest. The results from the MAP3S and related activities will contribute greatly to our ability to assess the impacts of the country's upcoming energy plan.

The deposition-resuspension work includes several field studies of the resuspension of plutonium and other respirable and nonrespirable particles, and dry deposition field studies leading to an improved predictive dry deposition velocity correlation.

Long-range transport studies were conducted through modeling and field measurements in the AFCT/TFCT Program, in the hemispheric radiation pollution studies, and in the analysis of the short-lived debris from the Chinese Nuclear test of September 17, 1977.

A series of theoretical laboratory and field programs were conducted during 1977 to assess the effects of the release of large quantities of waste heat and water vapor from energy centers. These studies included the postulated weather modification effects of large energy releases, the analogies between the heat release from fires and the precipitation enhancement due to topography and those corresponding effects from energy centers, and the rainfall enhancement due to cooling tower plumes.

PNL MUSKEGON OPERATIONS SUMMARY FOR FY-1977

J. M. Hales

During FY-1977 a contingent of Pacific Northwest Laboratory (PNL) personnel and the PNL DC-3 aircraft relocated to a field office in Muskegon, MI. This relocation brought the aircraft capabilities to a place in the northeastern quadrant of the United States where they could be used to greater advantage in the Multi-State Atmospheric Power Production Pollution Study (MAP3S). This article summarizes activities associated with this move during the past year.

Because of logistical, quality control, and cost considerations, it has been recognized for some time that a semipermanent relocation of the PNL aircraft and a sizeable scientific staff to the northeastern quadrant of the United States would be highly beneficial to the MAP3S program. Initial moving costs discouraged this decision, however, until late in the fall of 1976, when a contract awarded to PNL by the Electric Power Research Institute (EPRI) for further plume study research in this geographical area provided the critical additional funding necessary to complete the relocation.

The location chosen for the new field office was Muskegon, MI, which lies on the eastern shore of Lake Michigan. This location was chosen because of its strategic position in the northwest corner of the MAP3S region, because of its proximity to two major PNL-MAP3S field experiments, and because of the superior aircraft landing and maintenance facilities at the Muskegon airport.

The move from Hanford to Muskegon began in March 1977, when the DC-3 aircraft was transported to its new base and a MAP3S precipitation scavenging field experiment was conducted. In early April personnel were relocated permanently, and a flight series was conducted under the EPRI contract. During April and May, field offices, computer facilities and laboratories were set up. Two summertime flight series followed, the first involving the MAP3S study of urban plume transformations over Lake Michigan and the

second addressing MAP3S/Sulfate Regional Experiment (SURE) interactive characterization flights. The MAP3S/SURE flights were divided basically into three components: 1) an intercalibration series with three other aircraft at the Lewisberg, VA SURE station, 2) a northeast-southwest boundary-layer sounding series (Argonne cooperative) in southern Ohio and Indiana, and 3) a quasi-triangular characterization series in Michigan, Ohio, and Indiana.

During August the DC-3 instrumentation was modified to perform tracer characterization tests for the MAP3S scavenging program. This involved installing the acetone generator system on board the aircraft, locating a sample inlet probe near the tail, and mounting appropriate aerosol characterization equipment on board for analyzing generator output. Also during August the PNL Cessna 411 aircraft was brought to Muskegon for cooperative flights with Argonne National Laboratory to investigate O^{16}/O^{18} isotopic ratios in O_2 , water and SO_x .

After the tracer characterization tests were completed, the DC-3 aircraft was again modified for flights on the MAP3S Ambiens box-budget study, which was conducted in central Indiana in October. Table 1.1 summarizes Muskegon aircraft activities during FY-1977 and makes an initial value judgment of the success of the specific missions.

The physical facilities at the Muskegon field office consist of office space for personnel, a laboratory for chemical analysis

TABLE 1.1. Summary of Muskegon Flight Operations, FY-1977

Date	Activity	Scientific Flight Personnel ^(a)	Location	Preliminary Assessment of Scientific Value
March 1977	MAP3S Scavenging Flights	Scott, Busness, Laulainen, Thorp, Lee	Muskegon	Moderate
May-June 1977	EPRI Plume Studies	Busness, Easter, Hales, Lee	Muskegon Greenville, MS Terre Haute, IN	Moderate
July 1977	MAP3S Urban Plume Studies	Alkezweeny, Busness, Easter, Hales, Lee	Muskegon	Moderate - High
July 1977	MAP3S/SURE Intercomparison	Alkezweeny, Hales, Lee	Lewisburg, WV	Low (owing to bad weather)
August 1977	MAP3S/SURE/Argonne Characterization	Hales	Michigan, Ohio, Indiana	Low (owing to bad weather)
August 1977	MAP3S Tracer Characterization Tests	Scott, Hales, Young	Muskegon	High
August 1977	O ¹⁶ /O ¹⁸ Ratio Tests (Argonne Support, C-411)	Easter	Muskegon	Low (owing to initial nature of experiment and to poor aircraft performance)
October 1977	AMBIENS ^(b) Box-Budget Study	Alkezweeny, Hales, Lee	Columbus, IN	High

^(a)Pilots, R. F. Edwards and R. L. McIlvain present for all flight missions.

^(b)Atmospheric Mass Balance, Industrially Emitted and Natural Sulfur

and calibrations, hangar storage for the DC-3 aircraft, and a laboratory for computer systems. Present computer systems include a NOVA 1220 processing unit which is used for translating data tapes from a similar NOVA 1220 located on the aircraft and a CRT display terminal which permits dial-up access to the Battelle CDC 6400 computer in Columbus, OH. Current plans are underway to upgrade the local system to handle a major portion of the data processing load.

Personnel on permanent assignment to the Muskegon office are:

A. J. Alkezweeny
K. M. Busness
R. C. Easter
R. F. Edwards^(a)

J. M. Hales
R. N. Lee
R. L. McIlvain^(a)
P. A. Swanson^(b)

In addition to these personnel, others have been at the field site at specified times during the past year on an itinerant basis:

N. S. Laulainen
B. C. Scott
J. M. Thorp

The Muskegon field office will remain on-site throughout the duration of the MAP3S program. After the program is completed, equipment and personnel will return to Hanford.

^(a) Pilot.

^(b) Secretary.

THE MAP3S PRECIPITATION CHEMISTRY NETWORK

M. Terry Dana, D. R. Drewes, D. W. Glover and J. E. Rothert

Precipitation event samples have been collected at the four initial sites of the Network for approximately 1 year. These have been analyzed at Hanford for 11 pollutant species, and data reports are being issued monthly and periodically in summary form. Concentrations of the major species in samples collected concurrently with two different types of collectors compared favorably. Special handling of aliquots of samples for dissolved SO_2 analysis began during the summer of 1977. Three additional sites have been selected for further measurements and will be in operation by late 1977.

Precipitation event samples have been collected at four sites for approximately 1 year with a wet deposition-only collector developed by Pacific Northwest Laboratory (PNL)¹ and a standard wet-dry collector (Health and Safety Laboratory [HASL] collector). The initial sites, shown on Figure 1.1, were located at Whiteface Mountain (Site 1) and Ithaca (Site 2), NY; Pennsylvania State University (Site 3); and Charlottesville, VA (Site 4). Concentration data collected at these sites between 22 September 1976, and 1 July 1977, are listed in the first

summary report, along with a detailed description of the first year's operations.²

The samples have been chemically analyzed at PNL for 11 pollutant species -- pH, conductivity, SO_2 , SO_4^{2-} , NH_4^+ , NO_2^- , NO_3^- , Cl^- , PO_4^{3-} , Na^+ , and K^+ -- using ion chromatography, automated wet chemistry, flame emission spectrophotometry, and other standard techniques. When the procedures are available, the following species will also be analyzed: Mg^{++} , Al^{3+} , Ca^{++} (atomic absorption spectrophotometry), and acidity/alkalinity (titration).

Neg. 770733-1

FIGURE 1.1. MAP3S Precipitation Chemistry Network

The two precipitation collectors were compared by examining the concentrations of several species in concurrently collected samples. The results of a simple statistical analysis are shown in Table 1.2, where the number of samples, the number of samples where the percent difference in concentration exceeded 10, and the average percent difference are listed (positive $\Delta\%$ indicates that the HASL sample concentration was higher). There appears to be no statistically significant difference in the two collectors for sampling the species shown.

The funnel of the PNL collector was rinsed in the field with deionized water; analyses of these rinses indicated that they were effective for removing residual pollutants. The mass of remaining pollutants of interest were generally not significant for samples whose volume exceeded about 100 mL. At the

Virginia site, an additional PNL collector was operated; its funnel was returned to the local laboratory for HCl washing after each precipitation event. No significant differences in concentrations of the major pollutants between these two collectors were detected.

The need for special handling of samples for detection of dissolved SO_2 in precipitation was recognized through laboratory experimentation; special refrigeration equipment was developed for use in keeping collected samples cool during the summertime, and a procedure in which sample aliquots were treated to chemically fix SO_2 was instituted during the summer of 1977.

Three additional sites, shown in Figure 1.1 were selected and scheduled to begin operation late in 1977. An eighth site should be selected and placed in operation during 1978.

TABLE 1.2. Comparison of Concentration Differences Between HASL and Battelle Collectors

Site	Month(s)	H^+			$\text{SO}_4^{=}$			NH_4^+			NO_3^-		
		N ^(a)	N ₁₀ ^(b)	$\Delta\%$ ^(c)	N	N ₁₀	$\Delta\%$	N	N ₁₀	$\Delta\%$	N	N ₁₀	$\Delta\%$
1	February 1977	5	2	+8.7	6	1	0	0	-	-	6	1	+5.6
	March 1977	11	8	-3.3	11	2	+1.2	6	1	-5.7	11	1	-6.0
	April 1977	7	3	-3.3	7	1	+1.5	7	1	-7.7	7	1	-2.2
	May 1977	5	2	+1.2	5	1	0	5	3	+0.9	5	1	0
	June 1977	9	2	+1.7	9	1	+4.8	9	2	+9.0	9	1	+1.7
	Feb-June 1977	37	17	+0.7	38	6	+2.1	27	7	-0.5	38	5	-1.6
3	February 1977	4	2	+9.4	4	2	+15.	4	3	+10.	4	0	+5.6
	March 1977	6	2	+3.5	6	2	+1.6	6	1	+2.7	6	0	+0.4
	April 1977	4	0	+4.8	4	3	+13.	4	3	+16.	4	1	-4.7
	June 1977	3	0	-1.8	3	0	+1.3	3	1	-12.	3	2	+16.
	Feb-June 1977	17	4	-1.2	17	7	+7.0	17	8	+6.3	17	3	+1.9
1+3	Feb-June 1977	54	21	0	55	13	+3.7	44	15	+1.6	55	8	-0.5

(a) Number of sample pairs considered

(b) Number of sample pairs where the percent difference in concentration exceeded 10.

(c) $\Delta\% = \frac{\text{Concentration (HASL)} - \text{Concentration (PNL)}}{\text{Concentration (PNL)}} \times 100.$

TRACER SIZE DISTRIBUTION MEASUREMENTS

B. C. Scott and J. A. Young

An experiment has recently been conducted to determine the size distribution and microphysical characteristics of tracers used for in-cloud scavenging experiments. Observations were made of tracer size distribution and changes in cloud condensation nuclei (CCN) during tracer burns. Results show that the design of the Brad Patton aerosol generator currently in use is unsuitable in experiments examining the scavenging of submicron aerosol because the bulk of the aerosol mass generated is in giant particle sizes.

Whether tracer aerosols attach to cloud water depends strongly on the size distribution and solubility of the aerosol. For example, a soluble aerosol, such as ammonium sulfate, can be activated as a condensation nuclei at typical cloud supersaturations if its dimensions exceed some critical value (generally 0.01 to 0.1 μm). Less soluble tracers can also be effectively incorporated into the cloud drops through capture mechanisms, such as Brownian and turbulent diffusion, phoretic transport, and impaction. In contrast to the nucleation events where attachment rates to cloud water are essentially infinite, the attachment rates for less soluble particles are highly dependent on size and range from 0.01% h^{-1} for phoretic attachment to about 100% h^{-1} for inertial impaction. Thus, for an in-cloud tracer release experiment to provide information about attachment and removal of natural pollutants, it is absolutely essential that the size distribution and microphysical characteristics of the tracer are known. A recent experiment was designed to characterize tracers thought to be well suited for in-cloud scavenging experiments.

The experiment was performed by igniting tracer in a Brad Patton aerosol generator suspended below the main part of the aircraft fuselage and sampling the tracer material through a probe located in the aircraft's tail. The aircraft was equipped with a Whitby electrical aerosol analyzer and a Royco optical particle counter for measuring aerosol size distribution, an integrating nephelometer for indicating particles greater than 0.1 μm in diameter, and a device for measuring CCN. Filter samples were also obtained to verify that the aerosol measured was indeed the tracer released by the aircraft. The tracers examined were tantalum and europium, insoluble materials thought to be ineffective condensation nuclei in their pure form. Also burned was CaCl_2 , thought to produce a soluble aerosol.

As can be seen in Figure 1.2, the current generator configuration produces a bimodal

size distribution of tracer material with peaks in tracer concentration at diameters of $\sim 1.0 \mu\text{m}$ and $< 0.01 \mu\text{m}$. Figure 1.3 illustrates that 80-90% of the mass is contained in tracer particles greater than 1.0 μm in diameter. During the tracer burns, CCN concentrations at 1% supersaturation increased by a factor of 10 to 20 above background values, suggesting that a significant fraction of tracer was behaving as CCN. However, the concentration of CCN was relatively insensitive to the type of tracer material burned and to the absolute concentration of aerosol material generated. Thus, we were unable to conclude that the aerosol acting as CCN was composed of tracer material.

FIGURE 1.2. Typical Size Distribution of Tantalum Tracer

FIGURE 1.3. Typical Volume Distribution of Tantalum Tracer

This experiment demonstrated that the current design of the Brad Patton generator results in the production of significant numbers of giant particles. Thus, these generators are unsuitable for aerosol produc-

tion in experiments investigating the removal of submicron particles. Generators, modified to eliminate this giant particle mode, will be flight tested in January 1978.

ON THE CONCENTRATION OF SULFATE IN PRECIPITATION^(a)

B. C. Scott and N. S. Laulainen

Sulfate concentrations in precipitation, measured in field experiments conducted during winter, 1976-77, have been found to depend strongly on cloud supersaturation and liquid water content. The data suggested that most of the sulfate concentration may result from sulfate being incorporated into cloud water at nucleation, rather than from the transformation of SO_2 to sulfate.

The overall objectives of the precipitation scavenging program are to determine those precipitation mechanisms which are effective in removing SO_2 and $\text{SO}_4^{=}$ from the atmosphere and to provide the necessary field data for computing transformation rates of SO_2 to $\text{SO}_4^{=}$. Once effective removal and transformation mechanisms have been established, rates

of removing atmospheric pollutants and locations of removal can be predicted on an event-to-event basis.

The field program conducted during winter, 1976-77, was designed primarily to determine if a Lake Michigan site was adequate for accomplishing the overall objectives of the

(a) This article summarizes a publication submitted to the Journal of Applied Meteorology in October 1977.

scavenging program. The field site was accessible for aircraft flights and ground crews, and the storms were sufficiently predictable to enable frequent and periodic probing. Background concentrations of SO_2 , ammonia, and sulfate were readily detectable.

The influence of storm intensity and cloud type on the removal of sulfur was revealed in some detail during sampling of two widely varying synoptic situations. In the first storm, substantial liquid water content and high subcloud humidity were observed, while no liquid water was encountered during the second storm, and the humidity was considerably lower below cloud base. The sulfate ion was found to account for a considerable fraction of the total aerosol mass in each case, ranging from 70-100% for the first storm to 30-60% for the second one. The precipitation (and cloud water) for the first were quite acidic ($3.7 < \text{pH} < 4.2$) and contained large amounts of sulfate (11-33 mg/l); the precipitation for the second contained much less sulfate (~ 2 mg/l) and was much less acidic ($\text{pH} \sim 6.4$). Also of interest for the first case were data suggesting that acidic cloud water was neutralized by ammonia during the hydrometeors' fall to the ground.

When the observed aerosol spectra were related to the observed droplet spectra, it was possible to determine that a large fraction of the aerosol for the first case displayed microphysical characteristics similar to H_2SO_4 . In addition, $0.3 \mu\text{m}$ was established as the boundary diameter between those aerosol able to grow to sizes capable of being accreted by ice particles and those aerosol not removed by precipitation. Since most of these "activated" aerosol were sulfate, cloud-water concentrations of sulfate

could readily be computed. These in-cloud values are presented in Table 1.3 and compare favorably with the observed sulfate concentrations. The high sulfate concentrations of 33 mg/l near the cloud base appear to have been diluted to 12 mg/l as more water condensed in the cloud updraft. Falling snowflakes then accreted the cloud droplets to obtain a surface sulfate concentration of between 9.5 and 12.8 mg/l. The actual surface concentration produced by these mechanisms is dependent upon dilution by relatively pure snowflakes and by the vertically integrated accretion of cloud droplets; the greatest accretion rate occurs in regions of highest cloud water content (or lowest sulfate concentrations).

In contrast, the surface precipitation of the second case was considerably less acidic and contained about one-tenth the sulfate concentration. For this storm the strongest relationship between meteorological parameters and sulfate concentrations appeared to be the correspondence between enhanced sulfate concentrations and the passage of bands of precipitation. Although no cloud water content was observed during cloud penetrations, these cloud bands were not directly sampled. High intensity band echoes observed by the radar suggest significant cloud liquid water content. Thus, it is probable that the nucleation-riming mechanism, evident for the first case, was also the predominate mechanism leading to sulfate removal for the second case. As in the first case, sufficient sulfate aerosol flowed into the storm to account for all the sulfate in precipitation. In neither storm was it necessary to invoke an oxidation of airborne SO_2 to account for the sulfate observed at the ground.

TABLE 1.3. Comparison Between Computed and Observed pH and Sulfate Concentrations

	Computed SO_4 Concentrations, mg/l	Observed SO_4 Concentrations, mg/l	Computed pH Assuming H_2SO_4	Observed pH
Cloud Water Content				
(a) observed at 670m		32.9		4.0
0.10 g/m ³ (average)	32.0		3.18	
0.16 g/m ³ (maximum)	20.0		3.38	
(b) observed at 1525m		12.0		3.7
0.16 g/m ³ (average)	20.0		3.38	
0.32 g/m ³ (maximum)	10.0		3.68	

The data from these two case studies suggest that sulfate aerosol acted as cloud condensation nuclei in clouds supersaturated with respect to water and grew to cloud droplet size through the condensation process. The predominate removal mechanism then appeared to be accretion by large falling snowflakes. The concentration of sulfate in precipitation thus appears to depend strongly upon the cloud supersaturation and the liquid

water content. Both are related to the updraft velocity of the cloud, available cloud condensation nuclei, and ice content. The data suggest that the majority of the sulfate concentration in precipitation may result from the mass of sulfate incorporated into cloud water at nucleation, rather than from the transformation of SO_2 to sulfate during the time between nucleation and deposition on the ground.

PARAMETERIZATION OF SULFATE REMOVAL BY PRECIPITATION^(a)

B. C. Scott

In connection with studies to establish methods for predicting sulfate concentrations in the air and precipitation water, researchers at Pacific Northwest Laboratory have derived an expression to predict the sulfate washout ratio. This expression can be used to show that sulfate concentration in precipitation is directly proportional to the sulfate air concentration and inversely proportional to the cloud water content and that sulfate concentration in precipitation inversely depends on the precipitation rate. Calculations leading to the prediction of the ratio can also be used to predict the average oxidation rate of SO_2 over time intervals of about 4 days.

Sulfates are related to the structural, agricultural, and ecological damage associated with acid rains; they contribute to poor visibility because light is scattered by submicron particles; and they are suspected to be detrimental to human health. Because of this potential damage, accurate predictions of air and water quality are essential for responsible planning in the upcoming era of increased dependence upon fuels with high sulfur content. The work reported here is part of an effort to establish a capacity for predicting concentrations of sulfate in the air and in precipitation water.

Consideration of cloud microphysics and dynamics has been used to derive an expression for the sulfate washout ratio, ξ . Defined in terms of concentration per mass of air and water, the washout ratio can be predicted from

$$\xi = \frac{M_s(0)14000}{S_0 R^{0.88}} + \frac{1000 (1-4.41 \times 10^{-2} R^{-0.88})}{(1.56 + 0.44 \ln R)} \quad (1)$$

where S_0 is the airborne sulfate concentration below cloud base in units of g m^{-3} , R is the precipitation rate in units of mm h^{-1} , and $M_s(0)$ is equal to:

- (a) 0.0, when the Bergeron or ice growth process is necessary for precipitation initiation,
- (b) $0.1S_0$, when rain develops independently of any ice growth mechanism in the clouds,
- (c) $f(R)S_0$ (where $f(R)$ is a complicated function of R), when precipitation develops in intense convective storms or in clouds with tops warmer than 0°C .

As (1) illustrates, the removal of airborne sulfate is predicted to be strongly dependent upon the mechanisms of precipitation formation. If the precipitation is in the form of snow or originates as snow in the upper portions of the clouds, then the surface sulfate concentration is roughly proportional to $R^{-0.3}$. If precipitation-sized droplets develop independently of an ice stage, then the surface sulfate concentration becomes more strongly dependent upon

(a) This article summarizes a publication submitted to the Journal of Applied Meteorology in December 1977.

rainfall rate and the airborne sulfate concentration at the inflow levels of the storm. If the precipitation forms without the benefit of an initial ice growth stage, then for light precipitation rates ($R < 0.2 \text{ mm h}^{-1}$), the sulfate concentrations in precipitation water can increase by a factor of 70 or more over the ice-dependent predictions. At more moderate precipitation rates ($R \sim 1 \text{ mm h}^{-1}$), the precipitation originating as snow is predicted to have about one-third to one-half the sulfate concentration of that originating on water droplets. At heavy precipitation rates ($R > 7 \text{ mm h}^{-1}$), the sulfate concentration is nearly independent of the precipitation formation mechanisms. Limited surface observations give support to these predictions.

Equation (1) can be used to show that the sulfate concentration in precipitation is directly proportional to the sulfate air

concentration and inversely proportional to the cloud water content. Since the cloud water content is related to cloud supersaturation, which in turn depends upon the updraft velocity and ice content of the cloud, correlation between storm intensity, cloud temperature and sulfate concentration is to be expected. The parameterizations leading to (1) further illustrate an inverse dependence of the sulfate concentration in precipitation water on the precipitation rate.

Finally, the analyses leading to the prediction of the washout ratio can be applied to predict what the oxidation rate of SO_2 must be in order to replace the SO_4 removed from the atmosphere by each precipitation event. Taking typical values for precipitation rates and precipitation intervals in the eastern United States, SO_2 is forecast to be oxidized at an average rate of 0.2 to 2.0% per hour over 4-day intervals.

OZONE FORMATION RELATED TO POWER PLANT EMISSIONS^(a)

D. F. Miller^(b), A. J. Alkezweeny, J. M. Hales and R. N. Lee

Ozone concentrations in a power plant plume were investigated on July 20, 1977, at a plant near Lake Michigan. Measurements showed that ozone levels at first decreased and then increased significantly downwind from the stack, while the concentration of aerosol particles increased monotonically with distance from the stack. Outside the stack, sulfate aerosol and ozone concentrations increased with travel distance. To explain the chemical reaction in the plume, the authors simulated the reaction with a photokinetic model and found that the excess ozone could be explained by the excess nitrogen oxide in the plume.

The chemistry of plumes from fossil-fueled electric generating plants has become an important and controversial subject. One aspect of the controversy involves "ozone bulges" -- a phenomenon first reported by Davis, et al.³ According to the first reports, ozone concentrations in the domain of aged, yet well defined plumes sometimes exceeded the concentrations in the surrounding ambient air. Since ambient or background air makes up >99.99% of the gas volume of a dispersed plume, it seems unlikely to most skeptics that ozone concentrations in the plume path could be substantially different from those

of the ambient air, considering that the SO_2 , NO_x and particles are the only major pollutants directly emitted from power plants. Furthermore, investigators have shown that NO_x generally acts as a sink for ozone, and thus, all other air monitoring data have demonstrated the expected trend of ozone depletion of power plant plumes.

The production of ozone in a power plant plume was investigated on July 20, 1977, over Lake Michigan using the Pacific Northwest Laboratory's DC3 aircraft. The power plant is located just south of Milwaukee. The

(a) Summary of a paper submitted for publication in Science.

(b) Battelle-Columbus Laboratory, Columbus, Ohio.

plume was advected over the lake under the influence of a southwesterly wind. A substantial temperature differential between the lake surface and the air above it created a very stable atmospheric condition. Three north-south tracks, 300 m above the surface, were flown at the western, central, and eastern sites over the lake moving downwind with the advected air mass.

The plume was detected to a distance of about 120 km from the stack. The gas and particulate measurements indicate extremely low plume diffusion, a consequence of the stable atmospheric conditions. A pronounced ozone deficit was noted at the first downwind distance from the stack; however, further from the site, the ozone deficit was eliminated, and measurements showed a level significantly exceeding the ambient. On the

other hand, the concentration of aerosol particles increased monotonically with increasing distance from the stack. The chemical reaction in the plume was simulated by a photochemical kinetic model. The results of the simulation indicated that the measured excess of ozone could be explained by the excess nitrogen oxide in the plume, provided reasonable hydrocarbon levels existed in the background air.

Outside the plume, sulfate aerosol and ozone concentration increased with travel distance, from 34 to 38 $\mu\text{g}/\text{m}^3$ for the sulfate and from 95 to 102 ppb for ozone. These results agree reasonably well with earlier findings of Alkezweeny, et al.⁴ In that study, ozone production was found, but the sulfate formation was associated only with ozone level greater than 50 ppb.

TRANSPORT AND TRANSFORMATION OF POLLUTANT IN THE LAKE MICHIGAN AREA^(a)

A. J. Alkezweeny, J. A. Young, R. N. Lee, K. M. Busness and J. M. Hales

The transport and the transformation of pollutants emitted from the Milwaukee and Chicago areas were studied during August 1976, as part of the Multi-State Atmospheric Power Production Pollution Study (MAP3S). The experiments were conducted in approximately Lagrangian frames of reference with two instrumented aircraft and surface support units. The experiments showed that:

- Ozone levels build up downwind of the areas.
- Sulfate concentrations are higher in the urban plume than those upwind, and progressively increase with distance from the source.
- Significant concentrations of sulfate, probably associated with long-range transport of sulfur, are often detectable above the mixing layer.
- Vertical profiles of pollutants over the land and the lake show the effect of changing atmospheric stability on the transport of pollutants.

The objectives of Pacific Northwest Laboratory's Aerosol and Trace Gas Transformation Program are to measure and develop quantitative expressions to characterize pollutant transformation in plumes from urban areas and

power plant complexes. The geographical setting of interest is the energy intense northeastern United States, which is the focus of the MAP3S program. Of particular interest is SO_2 - SO_4 conversion. Field

(a) Presented at the 4th Joint Conference on Sensing of Environmental Pollutants, New Orleans, LA, 10 November 1977.

experiments conducted under this program are supported by diagnostic modeling and data interpretation with the ultimate goal of providing parameterized input for use by the MAP3S modeling community.

This report focuses on pollutant transformations in urban plumes. Other aspects of the experiment are discussed in a companion paper.⁵

EXPERIMENTAL DESCRIPTION

The experiments described in this report were conducted during August 1976, downwind of the Chicago-Milwaukee megalopolis. During the summer months, the urban plume is often transported within air masses that become stabilized when advected over Lake Michigan. This feature and the isolation from significant downwind sources create a situation where the effects of dry deposition and diffusion are dominant and can be easily included in the data interpretation process. The city of Milwaukee was assumed to be the principal source of pollutants in the study. Data were collected from two aircraft and mobile surface sampling units. An instrumented DC-3 aircraft conducted plume profile measurements at different downwind distances that were determined by wind speed and direction data recorded by on-board instrumentation. A light, twin engine aircraft flew a simultaneous mission collecting high-volume aerosol samples and SO₂ data upwind and outside the plume. Surface sampling was coordinated with aircraft activities within the urban plume utilizing lundgren impactors, high volume samplers, SO₂ bubblers, and CCl₃F collection bottles.

The parameters measured in real-time on board the DC-3 aircraft were: temperature, dew-point temperature, wind speed, wind direction, standard aircraft parameters (altitude, air speed, etc.), O₃, SO₂, NO, NO₂, light scattering, particle concentration, and aircraft position (longitude, latitude). The plume was further characterized using several integrated sampling techniques. Evacuated bottles opened during the course of the flight were used to obtain samples for CCl₃F analysis. Air samples were also captured in Tedlar bags and sent to the Environmental Protection Agency for hydrocarbon analysis. Three different high-volume samplers were used to characterize the aerosol component of the plume. The first one employed an IPC filter which was later analyzed by x-ray fluorescence and neutron activation techniques for trace metals. The second high-volume sampler, supplied by Brookhaven National Laboratory (BNL), provided

a measure of the principle atmospheric sulfur species. It consisted of an H₃PO₄-treated quartz filter in combination with base-impregnated backup filters for SO₂ capture. Analyses of the collected samples were conducted at BNL. The third sampler employed a 47 mm Fluoropore filter. The exposed filters were analyzed by infrared spectroscopy at Argonne National Laboratory or electron spectroscopy at Lawrence Berkeley Laboratory.

Results from the flights made during the 19-30 August test period are summarized in Figure 1.4. The location of each plume cross section is identified for each of the seven flights in this series. Each line represents a series of plume traverses at different altitudes.

EXPERIMENTAL RESULTS

Typically, during late August, the air and surrounding land masses are warmer than the lake surface. Under such circumstances, air advected over the lake becomes increasingly stable. This effect can be seen from the ozone data shown for 27 and 28 August (Figure 1.5). Each point in the figure is an average of the concentration measured at one altitude. At the first downwind distance from Milwaukee, the concentrations are nearly constant with altitude. Presumably, the air

Neg. 778921-1

FIGURE 1.4. Summary of DC-3 Sampling Flights

Neg. 773079-1

FIGURE 1.5. Vertical Profiles of Ozone at Different Downwind Distances

mass has not had enough time to adjust to the new surface temperature. However, the successive profiles show some vertical structure. The fifth profile, which was obtained a few miles inland on the opposite side of the lake, indicates a well-mixed atmosphere. In contrast to these over-water flights, the profile measurements over land display a rather uniform distribution of ozone throughout the mixing layer. All flights, however, indicate that ozone is being generated within the city plume and that the concentration is increasing with distance from the city. This is a feature which has been reported previously for other metropolitan areas.^{6,7,8}

The data generated during this test series are of particular significance to how SO_2 oxidizes in the atmosphere. Laboratory and modeling studies have indicated the features of potentially significant atmospheric reactions. Examination of our urban plume data in the light of these results suggests that homogeneous free-radical oxidation of SO_2 may be the dominant oxidation path during the events studied. This suggestion is supported both by the occurrence and the formation rates of

sulfate observed during these experiments. The occurrence of sulfate, for example, was observed to correlate more strongly with ozone (correlation coefficient of 0.56) than with trace metals, which typically exhibited correlation coefficients of much less than 0.4. Similar relationships were noted for the production rates of sulfate in the plumes, which were estimated from the data using the method of Alkezweeny and Powell.⁹ The data from 27 and 28 August (see Table 1.4), for example, were obtained under similar meteorological circumstances and SO_2 loadings, but exhibited marked variations in sulfate production rate. Large increases of aerosol in the light scattering range with distance downwind and the relatively large sulfate production rate of 6.8% per hour characterized 27 August. In contrast, 28 August exhibited essentially no sulfate production with no detectable increase in light scattering downwind. In these cases, ozone concentration again showed a rather strong correlation with sulfate production rate, while the concentrations of potential heterogeneous catalysts (e.g., iron, manganese) did not.

TABLE 1.4. Summary of Data Collected on 27 and 28 August

	27 August	28 August
Average SO ₄ (μg/m ³)	19.5	1.57
Average NH ₄ (μg/m ³)	3.34	0.42
Average Pb (ng/m ³)	225.	60.
Average Ca (ng/m ³)	1700.	1820.
Average Mn (ng/m ³)	34.	96.
Average Fe (ng/m ³)	1300.	4130.
Average Zn (ng/m ³)	77.	24.
Ozone (ppb) max.	108.	49.
Average Temperature, °C	28.	25.
Average R.H., %	58.	56.
k, % hour ⁻¹	6.8	0
Initial SO ₂ , μg/m ³	50.	64.
Light scattering	inc. with dist.	constant
Average acetaldehyde (ppb C)	52.	7.

The concurrent formation of sulfate and ozone noted in these experiments suggests that a rather strong link exists in the reaction change leading to these products. This is certainly in concordance with current concepts of oxidation by free radical chains involving species such as HO₂ and RO₂, which are generated in the systems containing NO_x and hydrocarbons and can react to form both sulfate and ozone as oxidation products.¹⁰ Hydrocarbon measurements obtained on these days give limited additional support for the free radical mechanism. Concentration of the radical precursor acetaldehyde on 27 August, for example, was 52 ppb, while on 28 August it had reduced to 7 ppb.

Current laboratory and theoretical evidence suggests that the peroxy radicals predominate in the oxidation process. The

recent experimental modeling study of Walter, et al., involving the trans-2-butene - SO₂ - ozone system, for example, indicates that the radicals HO₂ and RO₂ can account for most of the observed oxidation.¹¹

Another interesting result of the Lake Michigan experiment is the detection of sulfate layers above the mixing layer (Table 1.5). These layers were seen only when the wind was from the east or the northeast direction. For example on 23 August, a 7.4 μg/m³ sulfate concentration was found at 10,000 ft MSL. This value is higher than the 6.27 μg/m³ measured inside the Milwaukee plume. It is possible that SO₂ or sulfate was lifted to high altitudes from the northeast and then transported to this area.

TABLE 1.5. Sulfate and Trace Metals Upwind of Milwaukee

Date	Altitude in feet	Sulfate μg/m ³	Ca ng/m ³	Mn ng/m ³	Fe ng/m ³	Pb ng/m ³
23/8	1,500	1.6	239	6.3	159	7.1
	10,000	7.4	28	ND	17	4.4
24/8	1,500	5.4	3,140	25.6	614	46.
	6,000	9.0	88	3.4	73	24.
	8,000	ND	ND	ND	ND	ND
29/8 ^(a)	1,000 to 6,000	0.3	79	1.7	54.9	3.6
	5,000	0.9	535	17.3	539	ND

ND = Not Detected

^(a)Over Lake Michigan, wind from the north

THE PERFORMANCE OF A SULFUR ANALYZER UTILIZING A FLAME PHOTOMETRIC DETECTOR DURING SIMULATED AIRCRAFT OPERATION

R. N. Lee

The pressure dependence of an SO₂ analyzer operated from an aircraft platform during atmospheric monitoring programs was investigated with an altitude simulation chamber. Analyzer output to zero air and dilute SO₂ was found to drift at a rate equivalent to 1 ppb SO₂ per thousand foot elevation gain.

The Multi-State Atmospheric Power Production Pollution Study (MAP3S) encompasses the fate of atmospheric pollutants produced by the combustion of fossil fuels. Within this framework the transport and transformation of sulfur dioxide is of primary importance because of the magnitude of SO_2 emissions accompanying combustion. A realistic interpretation of concentration data acquired during field experiments conducted under the MAP3S program depends on the accurate characterization of analyzer performance under the experimental conditions encountered. Aircraft-mounted sulfur analyzers employing the flame photometric detector, while offering excellent sensitivity and response characteristics, are subject to a pronounced pressure dependence. Analyzer response recorded during aircraft soundings of the atmosphere must therefore be processed using corresponding elevation data and suitable correction factors to derive accurate concentration data.

The performance of a Meloy SA285 sulfur analyzer employed in MAP3S programs, such as the AMBIENS experiment, has been examined using the altitude simulation chamber at the University of Michigan. The stainless steel chamber, which measured 3 x 3 x 3 ft, could be sealed and pumped down at adjustable rates to pressures corresponding to elevations of more than 20,000 ft. A port at the front of the chamber permitted equipment operated within the chamber to be monitored during tests.

Sulfur analyzer performance was investigated over a pressure range equivalent to altitudes of 1000 to 7000 ft MSL. Sulfur dioxide concentrations ranging from 0 to 56 ppb (v/v) were examined to approximate

atmospheric concentrations in well-mixed air parcels similar to those encountered downwind of major area sources. Analyzer configuration (Figure 1.6) was identical to that employed in normal aircraft operation. Sample gas was introduced through 1/4-in. OD Teflon tubing. A Teflon tee permitted excess sample to vent to the chamber while a portion was withdrawn through a Teflon filter before it entered the detector. Although a filter is always employed during field operations to exclude sulfate aerosol, altitude simulation tests were conducted both with and without the filter in-line. During tests in which chamber air was sampled, the tee and upstream tubing were removed from the sample stream. Hydrogen gas to fuel the reducing flame of the FPD was supplied by a hydrogen generator. Analyzer output was measured during these tests using DVM and was hand recorded. Chamber pressure was determined with a mercury barometer.

Variation in analyzer output with respect to pressure was determined for the three sulfur dioxide concentrations cited in Table 1.6. Zero gas was introduced to the sample line from a compressed air cylinder via a Meloy Zero Air Filter Assembly to ensure the absence of sulfur gases. A sample, 56 ppb in sulfur dioxide, was generated when a measured flow of air was passed through a calibrator containing an SO_2 permeation device. A third intermediate concentration of sulfur dioxide was obtained by introducing gas from the calibrator to the open chamber. After a measurable concentration was achieved, the chamber was sealed and pumped down to simulate aircraft ascent. It was assumed that SO_2 absorption by chamber walls was insignificant during the course of the 10-15 minute experiment.

FIGURE 1.6. Sample Flow Schematic for Analyzer Operation at Reduced Pressures

TABLE 1.6. Variation in Analyzer Output with Decreasing Pressure (simulated elevation gain)

(SO ₂), ppb	ΔOutput/1000 ft	Range
0	-0.13 volts	100
0	-0.13	100
0	-0.14	100
16	-0.11	100
16	-0.12	100
56	-0.14	100

Data presented in Table 1.6 describe the variation in detector response to zero gas and dilute sulfur dioxide at pressures corresponding to normal aircraft operation.

Flame-out down to pressures equivalent to approximately 8500 ft MSL presented no difficulty. Detector response decreased uniformly with elevation at a rate independent of the presence of sulfur dioxide in the sample. This variation is equivalent to a decrease of 1 ppb SO₂ per 1000 ft elevation gain and is in good agreement with the zero drift measured by sampling ram air passed through the zero air filter assembly in flight.

The results of these tests are preliminary and will serve as a guide for the design of future experiments dealing with aircraft operation of the FPD sulfur analyzer. The valuable assistance of Prof. Bartman of the Department of Atmospheric and Oceanic Sciences, The University of Michigan is gratefully acknowledged.

A VARIABLE TRAJECTORY MODEL FOR MULTI-STATE
ASSESSMENT OF AIR POLLUTION FROM SULFUR OXIDES^(a)

D. C. Powell, D. J. McNaughton,
L. L. Wendell and R. L. Drake

A sulfur oxides atmospheric pollution model is described that calculates trajectories using single-layer historical wind data, as well as chemical transformation and deposition following discrete contaminant masses. Vertical diffusion under constraints is calculated, but horizontal diffusion is effectively a function of trajectory variation. The model also estimates pH of real-time precipitation based on the contribution to rainfall acidity associated with sulfate. The ground-level air concentrations, deposition and pH are calculated in a rectangular area comprising northeastern United States and southeastern Canada.

Calculations for a 29-day assessment period in April 1974 are presented along with a limited verification method. Results for studies were calculated using a source inventory comprising 61% of the anthropogenic SO₂ emissions. Using current model parameterization levels, the predicted concentration values are most sensitive to variations in dry deposition of SO₂, wet deposition of sulfate, and transformation of SO₂ to sulfate. Replacement of the variable mixed-layer depth and variable stability features of the model with constant definition of each results in increased ground-level concentration predictions for SO₂ and markedly for sulfate.

(a) Abstract of a paper submitted to Atmospheric Environment.

PROGRESS REPORT ON THE MAP3S-PNL REGIONAL SCALE MODELING

D. C. Powell and D. J. McNaughton

The principal advance made by Pacific Northwest Laboratory (PNL) in its regional modeling work for the Multi-State Atmospheric Power Production Pollution Study (MAP3S) during FY-1977 was the calculation of pH based on rainfall acidity derived from sulfate-and-carbon-dioxide-associated ionization. This development and the initial results are described in another report in this volume.^(a)

Improving the concept and coding the vertical sophistication described in last year's annual report involved considerable time.¹² These steps were necessary to eliminate the uncharacteristic physical behavior in calculations detected when subsequent results were scrutinized. The principal conceptual

change was the adoption of a G stability growth rate rather than an E stability for new plume elements released at a height above the temporary mixed-layer depth, as shown in Figure 1.7. In the figure, L_d is the temporary lid height for the mixed layer, L_{dm} is the maximum daily lid height for the mixed layer, L_t is the tropopause height, and stability (t_h) is the stability of the hour.

Other changes were as follows:

1. After any hour at which a puff is considered evenly distributed within the mixed layer, the puff continues to be evenly distributed in the growing layer until the layer loses its definition. Dissolution of the mixed layer is modeled to occur daily at 1800 hr, and after that time, the vertical dimension increases as a function of time, as Powell has explained.¹²
2. The diurnal stability vector of hourly values was reapportioned to reflect the proper percentages of stable, neutral, and unstable hours over northeastern United States in the spring. The percentages were determined by averaging the percentage of occurrence values for each stability given on maps by Doty, Wallace, and Holzworth.¹³ The new diurnal vector of hourly values determined is

555666 444443 234444 44445.

The leading 5 means that E stability is presumed to prevail in the growing nocturnal boundary layer from 0000 to 0100; the 2 means that B stability is presumed to apply in the growing daytime mixed layer from 1200 to 1300, etc.

3. The model now calculates assessments involving any number of sources taken in groups of ten for each traverse of the wind and precipitation tapes. The average concentration and deposition contributions from each set of sources can be stored individually or additively.
4. Model parameters for pollutant removal were reassessed to reflect more realistic values as shown by experimental programs.¹⁴
5. Model options were developed to allow for modification of transformation rate during precipitation periods and to provide

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FIGURE 1.7. Daytime Illustration of Stability Definition and Vertical Constraints

(a) D. C. Powell and D. J. McNaughton, "Calculation of Rainfall pH on a Multistate Scale," p. 1.23.

for initial sulfate emissions from each source. These options were incorporated to reflect observations made in field programs showing a dependence of transformation rate on relative humidity¹⁵ and initially higher transformation rates expected in urban areas.¹⁶

Significant model testing also took place in 1977. Sensitivity tests on the following model parameters were performed:

- wet and dry removal parameters for SO₂ and sulfate
- transformation rates for SO₂ to sulfate
- variations in stability
- fixed versus variable mixing depth
- time-averaged versus sequential precipitation
- variations in maximum mixing depth
- stabilities above the mixing depth

- transformation during precipitation periods.

Sensitivity tests are underway studying:

- emission height variations
- urban transformation rates
- emission source grouping or spacing to reduce computations.

Emissions input data have been updated to provide a 63-source, U.S.-Canadian SO₂ emissions inventory for 1973-1974 to better represent emissions during the April 1974 test case. The inventory is comprised of the 4 largest utility and 22 nonurban sources on the calculational grid.

In the area of model verification, preliminary verification on the April test case was performed using experimental data from recent regional sulfate experimental programs and a statistical package designed to interface with the model results. Data for further verification studies have been identified.

IDENTIFICATION OF DATA SOURCES FOR MAP3S MODEL VERIFICATION

D. J. McNaughton

Interactions within the component parts of the Multi-State Atmospheric Power Production Pollution Study (MAP3S) and between the MAP3S Program and the Electric Power Research Institute (EPRI) Sulfate Regional Experiment (SURE) are providing additional data for regional model verification. Data that will be used in the PNL-MAP3S modeling studies have been identified.

Regional modeling activities at Pacific Northwest Laboratory (PNL) have focused on the prediction of the long-term fate of sulfur pollutants originating as emissions from utility and nonutility sources. Preliminary model verification efforts have been hampered by uncertainties in the ambient air quality data used in this work. Three versions of the regional model are being developed to better describe the regional transport, transformation, and removal of sulfur contaminants, each of which will be able to use some of the newly available data for verification. The model versions are:

- the existing long-term, source-oriented forward trajectory model
- an episodic (1- to 4-day periods), source-oriented forward trajectory model
- an episodic, receptor-oriented back trajectory model.

Forward trajectory models can study the effects of emissions sources on a computational area, and back trajectory models can study and tag the emission sources which influence a particular receptor, such as an

ambient air quality monitoring station. The model currently predicts sulfur dioxide and sulfate concentration and deposition, and precipitation acidity through pH estimates.

Data sources available for verification are the EPRI SURE I and II Programs, Environmental Research and Technology, Inc. (ERT) episode data sets, and the MAP3S precipitation chemistry network data. U.S. Environmental Protection Agency National Aerometric Sampling Network (NASN) data are also available.

EPRI SURE I DATA

Current model verification efforts make use of a limited set of data from the SURE I Program.¹⁷ In this program, daily meteorological data and ambient total suspended particulate (TSP), SO₂, and sulfate concentration data were collected at 12 regional monitoring sites in the northeast United States from April 1974 through March 1975. The data sites were selected to be regionally representative, that is, representative of regional sulfate patterns at sites not influenced by local emission sources or local meteorological phenomena such as valley winds. Data capture was generally good except early in the period when start-up problems caused some difficulty. In conversation with the author, E. Y. Tong reported that one site has recently been removed from the data set due to the influence of a local emission source.

EPRI SURE II DATA

An extension of SURE I design phase work is being carried out in the SURE II Program. This work involves regional air sampling in the Northeast for a 19-month period which started in August 1977. Two sets of monitoring sites are active. An intensive set of nine stations will sample continuously for SO₂, O₃, NO and NO₂, daily for TSP and sul-

fate, and every three hours for TSP and sulfate in two particle size ranges during the central month of each season. A second set of 49 stations will monitor TSP and sulfate concentrations daily and SO₂ data continuously over the entire project term. All sites selected were at locations intended to be regionally representative.

ERT EPISODE DATA

ERT, the prime contractor for the SURE studies, with the cooperation of Boston University, compiled a collection of air quality and concurrent meteorological data for 27 episode days during the period from April 1974 through May 1975.¹⁸ These episodes are periods of either high or low concentrations during times when the Northeast was dominated by a single air mass. Data sources included in the analysis are the EPA NASN data, the EPRI SURE data, and data from the Tennessee Valley Authority.

MAP3S PRECIPITATION CHEMISTRY NETWORK DATA

A portion of the MAP3S experimental activities includes the operation of a precipitation chemistry network in the Northeast to sample for expected ionic pollutant species in precipitation samples.¹⁹ Currently, data are collected with a minimum sampling period of 24 hours at 4 locations in New York, Pennsylvania, and Virginia.

MODEL VERIFICATION

Verification of long-term model predictions will continue to make use of SURE data sets. The episodic forward trajectory model verification will make use of the EPRI SURE II data and possibly the ERT episode data sets. The receptor-oriented back trajectory version of the model will be used to analyze source regions for the precipitation chemistry network data and data from other field programs where possible.

SENSITIVITY TESTING OF THE
MAP3S-PNL REGIONAL MODEL

D. C. Powell and D. J. McNaughton

Sensitivity tests of a regional model developed by Pacific Northwest Laboratory (PNL) as part of the Multi-State Atmospheric Power Production Pollution Study (MAP3S) have recently been conducted. The removal and transformation parameters used in the model were varied within the range of uncertainty, and the variable mixed-layer depth and stability treatments in the model were replaced with constant definition of either or both. The results of these tests show that 1) conservative concentration estimates result from assuming constant stability, and 2) sensitivity to dry deposition of SO_2 , to wet deposition of sulfate, and to transformation is greater than is sensitivity to wet deposition of SO_2 or dry deposition of sulfate.

Recently a number of sensitivity tests have been conducted with the MAP3S-PNL model 1) by varying within the range of uncertainty the removal and transformation parameters used in the model and 2) by replacing the variable mixed-layer depth and stability treatments in the model^{20, (a)} with constant definition of either or both. All these tests were made using real-time wind and precipitation data for a 29-day period in April 1974 and 10 or more sources of SO_2 emissions.

The model's sensitivity to changes in parameter magnitudes was tested using a set of linear differential equations for the time-rate of change of mass in a plume element as described by Wendell, et al.²¹ The processes of interest are dry deposition of SO_2 , wet deposition of SO_2 , dry deposition of sulfate, wet deposition of sulfate, and transformation of SO_2 to sulfate. It was found that the sensitivity of the average concentration of either SO_2 or sulfate to dry or wet removal or to transformation is very similar to the sensitivity of plume element mass to the same processes as prescribed directly in the differential equations relating removal and transformation to change of plume mass.²¹ A hypothetical time-rate of mass change equation for a plume element illustrates:

$$\frac{dQ}{dt} = p_1 + p_2 \quad (1)$$

where Q is a plume-element mass and where p_1 and p_2 are two relevant processes. If the numerical magnitude of p_1 is an order of magnitude greater than the numerical magnitude of p_2 , the calculated change of mass will be that much more sensitive to p_1 than to p_2 .

Extending this concept to the model differential equations, with current best estimates of the range over which deposition velocities and scavenging coefficients (length^{-1}) vary, we would predict that the change of mass is much more sensitive to dry deposition of SO_2 , wet deposition of sulfate, and transformation than to wet deposition of SO_2 or dry deposition of sulfate. These results are illustrated in Table 1.7. The numbers in the first column are ratios between the altered parameter values and the base-case parameter values. The base case parameter values were:

Maximum mixing depth	1500 m
Dry deposition velocity for SO_2	1 cm sec ⁻¹
Scavenging coefficient for SO_2	5 m ⁻¹
Dry deposition velocity for sulfate	0.1 cm sec ⁻¹
Scavenging coefficient for sulfate	250 m ⁻¹
Transformation rate	0.02 hr ⁻¹

The value of 0.67 opposite maximum mixing depth means that the altered value used for the sensitivity test was 0.67 times the base value, 1000 m as opposed to 1500 m. The source inventory consisted of 40 utility sources and 20 urban sources.

The fully developed model contains specifications for varied mixing depth and varied stability, with both variations being in accordance with fixed diurnal cycles.²⁰ When

(a) D. C. Powell and D. J. McNaughton, "Progress Report on the MAP3S Regional Scale Modeling," p. 1.16.

TABLE 1.7. Percent Change of Average Values from Base Case

Case	Parameter Ratios	Percent change in average values from the base case							
		SO ₂ Concentration		SO ₂ Deposition		Sulfate Concentration		Sulfate Deposition	
		All Sources	Utility	All Sources	Utility	All Sources	Utility	All Sources	Utility
Maximum Mixing Depth	0.67	10.7	12.9	10.3	12.3	20.3	21.7	-0.2	-0.1
Dry Deposition Velocity for SO ₂	2.00	-30.5	-26.4	41.8	47.4	-21.7	-18.5	-21.1	-18.7
	0.50	25.3	19.8	-37.3	-39.3	17.5	13.7	16.7	13.4
Wet Removal Coefficient for SO ₂	10.00	-4.7	-5.4	9.2	11.6	-2.9	-3.1	-6.9	-7.0
Dry Deposition Velocity for Sulfate	2.00	0.0	0.0	0.0	0.0	-4.1	-3.7	14.3	13.6
Wet Removal Coefficient for Sulfate	0.10	0.0	0.0	0.0	0.0	14.8	15.4	-59.4	-60.2
	0.60	0.0	0.0	0.0	0.0	3.8	4.0	-16.1	-16.3
SO ₂ to Sulfate Transformation	0.50	14.5	17.5	13.7	16.8	-43.6	-43.1	-43.4	-43.2
	2.00	-20.7	-24.5	-19.8	-23.7	63.0	60.5	62.1	61.1
SO ₂ to Sulfate Transformation Rate	3.50	-6.6	-7.8	-6.5	-7.9	11.1	10.7	43.6	43.0
During Precipitation	0.50	1.7	2.1	1.7	1.7	-3.3	-2.8	-11.4	-11.2

the model uses the varied mixed-layer depth option, the vertical expansion rate of newer releases increases when the mixed-layer depth changes from below the release height to above it; this version also allows older releases to expand beyond the maximum diurnal mixed-layer depth up to the tropopause limit.

In assessment runs using elevated releases only (utility sources, as opposed to urban sources, which are modeled as ground-level releases), a base case model version using full vertical sophistication was compared to the following cases:

- 1) constant mixed-layer depth of 1500 m, variable stability
- 2) variable mixed-layer depth, constant stability (neutral)
- 3) constant mixed-layer depth of 1500 m, constant stability (neutral).

In almost all cases, the predicted ground-level air concentrations and the predicted deposition rates increased when computations from one of the simplified model versions were compared to computations from the full model. The percentages of increase using each of the three simplified versions of the model as compared to the full model are given in Table 1.8.

Three observations are made from the numbers in this table. First, the model predic-

TABLE 1.8. Percentage Increases of Concentration and Deposition Predictions when Simplified Models are Compared to Full Model

Simplification	SO ₂		Sulfate	
	Conc.	Dep.	Conc.	Dep.
Constant lid	3	3	19	3
Constant stability	28	28	66	2
Both lid and stability constant	21	21	43	1

tions based on the assumption of constant stability are significantly higher than those based on the diurnally varying stability. Second, while predicted SO₂ concentrations and deposition are increased by the same ratio, predicted sulfate concentrations and deposition differ markedly from each other. Under the present parameterization, the SO₂ deposition is dominated by the dry process, while the deposition of sulfate, using the real-time precipitation, is dominated by the wet process. Since dry deposition is proportional to ground-level concentration, the two SO₂ budgets increase similarly. Since wet deposition is presently modeled as proportional to vertically integrated concentrations, the two sulfate budgets increase differently. If the deposition of sulfate were modeled entirely wet (i.e., zero deposition

velocity for sulfate), the percentage increases in the table would be negative since there is less sulfate in the air in each case of simplified modeling, particularly when constant stability is used. The small positive percentages are due to the particular mix of dry and wet deposition modeled for sulfate, with wet deposition dominating.

Third, the predicted sensitivity of sulfate concentrations is significantly greater than that of SO_2 concentrations. This sensitivity is due to the modeling of unstable conditions, since the vertical distribution quickly fills out the mixed-layer depth under the unstable

the unstable conditions and reduces subsequent ground-level concentrations abruptly in a way that does not occur when constant stability is modeled. Since plume elements are released hourly and since the transformation rate is only 2% per hour, most plume elements make significant contributions to ground-level SO_2 concentrations before the sudden expansion due to afternoon instability occurs. This is not the case for sulfate. A more quantitative explanation and demonstration will be given if this feature of sensitivity remains in the model after more refinement has been incorporated.

VERIFICATION OF PREDICTIONS MADE USING THE MAP3S TRAJECTORY MODEL

D. J. McNaughton and D. C. Powell

Verification studies for long-term predictions using the Pacific Northwest Laboratory (PNL) regional trajectory model are in progress as part of the Multi-State Atmospheric Power Production Pollution Study (MAP3S). Data available for the preliminary verification period (April 1974) were limited so that only results for sulfate concentrations are presented. The example analysis in this report is qualitative since subgrid scale contributions to observed sulfate data could not be separated from the contributions due to regional transport. Improved results are expected with improved data sets now becoming available.

Verification efforts by other investigators for regional scale atmospheric pollutant transport and removal models have been limited in large part to predictions over episode periods of one to four days rather than the longer periods over which the PNL regional trajectory model is being run for the MAP3S program. Episode data are generally more available from limited term field programs, and verification of regional models appears to be successful based on high correlations of observed-to-predicted concentrations. Contrary to expectations in model verification, the episode predictions of sulfate concentrations appear to be more accurate than long-term predictions. This seems to result since episodes present periods of peak concentrations when simplified transport schemes perform best, while long-term predictions include both peaks and a large number of values near the noise level of observed data. Long-term average predictions, since they consider all time periods, may also include the effects of highly variable

meteorological conditions, such as frontal passages, where simplified transport schemes may or may not be appropriate.

PNL work on a MAP3S regional scale model is currently directed toward improving long-term predictive capabilities for sulfur dioxide and sulfate concentrations and deposition, as well as for rainfall pH, although particular emphasis on verification is directed toward ambient sulfate concentrations. The sulfate data are most readily available, and variations are largely regional in nature since sulfate is a secondary pollutant formed by transformation from sulfur dioxide. Work on episode predictions is now in progress.

The purpose of this report is to show preliminary verification results from techniques currently being employed to relate predicted and observed sulfate concentrations. The analysis will be extended to sulfur dioxide concentrations as more data become available.

PREDICTED SULFATE VERIFICATION

A statistical verification program has been written to provide analysis of observed versus predicted concentrations at specified sites by providing information on predictive errors, linear correlation and regression, maximum and average concentrations, and the frequency distribution of predicted concentrations within specified multiples of observed data. In addition, qualitative verification can be performed by examining the spatial patterns in observed and predicted concentration fields.

Statistics for a base case simulation for April 1974 appear in Table 1.9. The base case prediction was made using a 60-source SO₂ emissions inventory representing approximately 60% of total emissions in the computational area and best estimates for model parameters for pollutant removal and transformation. Three verification cases are listed in the table showing verification on an unedited set of all available observing data, all rural data, which should be more representative of regional patterns, and all available stations excluding one point to show the sensitivity of verification to observed data. Due to the small number of rural observing sites, statistics for this case may not be as significant as those in the larger data sets.

TABLE 1.9. Verification Statistics

	Monitoring Sites		
	Urban and Rural	Rural	
	107	106	15
Mean Observed Concentration ($\mu\text{g m}^{-3}$)	9.8	9.8	8.4
Mean Predicted Concentration ($\mu\text{g m}^{-3}$)	5.1	5.1	5.0
Correlation Coefficient	0.43	0.49	0.37
Regression Line:			
Slope	0.26	0.29	0.26
Intercept	2.65	2.27	2.78
Fraction (%) of Concentrations Within a Factor of:			
2	57	57	79
3	87	87	94
Average Absolute Error ($\mu\text{g m}^{-3}$)	4.7	4.7	3.4

Mean observed sulfate concentrations are higher than predicted as would be expected since a limited emissions inventory was used. The smaller difference between observed and predicted data in the rural data implies that observed data in urban areas include components from sources not resolved by the model.

The correlation coefficient along with a subjective analysis of observed and predicted concentration plots can provide information on the prediction of spatial variability in concentrations. Results show a weak positive correlation for the base case with a correlation coefficient of 0.43. This result may be in part attributable to the quality of the data set used for verification. A correlation of 0.49 is obtained if one questionable data point is removed. Possible problems may be arising due to use of a data set which contains data points near the calculational grid boundaries and at points in urban areas where subgrid scale effects influence concentrations.

The regression between observed and predicted concentrations is immediately useful in determining if the model is overpredicting and if the model does not allow for contributions by different effects to the observed data. In this example, the model generally underpredicts, as is shown by the regression line slope of 0.2 to 0.3, except at low concentrations shown by an intercept 2.7 $\mu\text{g m}^{-3}$. The overprediction may be a result of the source inventory configuration, the verification data, or the model transport scheme.

Both the concentration frequency distribution and average error show improvements in verification at rural monitors over the urban-rural mix. This result again suggests difficulties with verification data with subgrid scale contributions. Later assessments with better rural data will allow a quantitative assessment of the predictive capabilities of the model.

OBSERVATIONS

Preliminary verification results show improvement is required in the prediction of low concentrations and concentrations near the computational boundaries. The statistics presented can only be qualitatively assessed because of questions on the verification data set. Future activities will include more rural data which are more comparable to the regional scale predictions made by the model.

A MULTISOURCE COMPARISON OF THE EFFECTS OF REAL-TIME VERSUS
TIME-AVERAGED PRECIPITATION DATA ON SO₂ AND SULFATE PARTICULATE
REMOVAL IN A REGIONAL AIR QUALITY MODEL (a)

L. L. Wendell, D. C. Powell and D. J. McNaughton

A regional-scale air quality model incorporating transport dispersion, chemical transformation, dry deposition, and precipitation scavenging was used to test the sensitivity of such models to different treatments of wet removal. Average ground-level air concentration and deposition patterns of SO₂ and sulfate particulate over a 29-day period were produced for the emission estimates of 30 power plant sources in the northeast

United States. More than twice the wet removal of SO₂ was observed for the time-averaged precipitation than for the real-time precipitation. The SO₂ not removed in the real-time precipitation case was available for transformation into sulfate particulate and resulted in higher ground-level air concentrations of sulfate particulate by about a factor of two, as well as more deposited sulfate and significantly more sulfate escaping the computational area.

CALCULATION OF RAINFALL pH ON A MULTISTATE SCALE

D. C. Powell

An approximation method for calculating rainfall pH has been developed. Through a derivation procedure, it predicts the pH of the total rainfall that falls at a given location over long time periods. Favorable comparisons of predicted pH values and the average observed pH values encourage further development of the method.

The growing concern over the ecological effects of acid rainfall has motivated the development of an approximation method for calculating rainfall pH. In developing this method, the following assumptions have been employed:

1. The hydrogen ion concentration in the rain comes from two sources: 100% dissociation of H₂SO₄ and other contributions that may be parameterized as a background.
2. The wet removal rate for the sulfate over a given area is directly proportional to the precipitation rate and to the vertically integrated sulfate concentration. The proportionality is assumed to be constant.
3. Although each individual calculation that goes into the final pH calculation depends explicitly on sampling grid square area,

the total time T has enough sampling intervals Δt that the final calculation does not depend on grid square area. Physically this amounts to assuming that horizontal diffusion is a function of synoptic scale wind variation rather than turbulent diffusion.

The final result of the derivation predicts the pH of the total rainfall that falls at a given location over time T. The equation is

$$\text{pH}_{ij} = -\log \left[\text{Base} + 2.08 \times 10^{-11} \lambda \frac{\sum_t Q(t) P_{ij}(t)}{A_{ij} \sum_t P_{ij}(t)} \right] \quad (1)$$

The pH is an average value for an entire sampling grid square area A_{ij} in square meters.

(a) Abstract of paper printed in Proceedings of the AMS Air Pollution Symposium, Salt Lake City, UT, Nov.-Dec. 1977.

The $P_{ij}(t)$ are gridded precipitation rates averaged over gridded square ij at each sampling time t . The $Q(t)$ are the masses of sulfate (micrograms) in all the puffs that happened to be over grid square ij at various sampling times t . The λ is an inverse length parameter in meters⁻¹. This parameterizes the wet removal rate W of a puff mass Q according to

$$W = Q \lambda P \quad (2)$$

where P is again the precipitation rate. The base is the assumed hydrogen ion concentration from atmospheric CO_2 . According to Likens,²² a reasonable figure is $10^{-5.6}$ moles per liter. The 2.08×10^{-11} is the necessary conversion factor to render the second term in the same units as the base.

From (1) we see that according to the assumptions employed, the pH depends only on the vertically integrated sulfate concentration and on the scavenging parameter λ . The precipitation rate enters only in the role of weighting factor.

The value of λ used was taken from the range of possible values indicated by Dana and Hales.²³ Unfortunately this range extends over three orders of magnitude when considering particle washout by frontal storms only. One must assume a mean geometric particle radius and a mean geometric standard deviation of particle radii about this mean to come up with a value for λ . On the basis of personal communication with the authors, a decision was made to use a relatively high value

$$\lambda = 250 \text{ m}^{-1}.$$

A pH map resulting from a case analysis for April 1974 is shown in Figure 1.8. The distribution correlates fairly well with the 1972-73 map given by Likens.²² However, there is a considerable extension of low pH down the Ohio River area shown by Likens that does not appear on this prediction map.

Chiefly due to differences in the Ohio Valley, the average pH on Figure 1.8 is approximately 0.3 higher than the average pH

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FIGURE 1.8. Predicted Average pH of Rainfall

on the map by Likens. This is acceptable 1) since the sulfur compound emissions inventory used represents only 61% of the anthropogenic sulfur compound emissions, and 2) since sulfur compound emissions according to Likens constituted only 66% of the total contribution to ambient acidity, 30% being from nitrates. Therefore, the SO₂ emissions inventory used consists of not more than 40% of the total contribution to ambient acidity, and average predicted

pH value based on this inventory should be about 0.4 higher than the ambient pH.

A comparison between average predicted and average observed pH values is one potential means of assessing the highly uncertain value of λ^{-1} . But rather than making any inferences at this time, the wisest course seems to be to consider this result as an encouraging approximation indicating that the method is worthy of further development.

PLANETARY BOUNDARY LAYER PROFILE STUDIES UNDER INVERSION CONDITIONS

J. G. Droppo

The data from a 1976 cooperative field effort with Argonne National Laboratory have been analyzed. The results consist of Planetary Boundary Layer vertical profiles of temperature, mixing ratio, ozone, and condensation nuclei from heights as low as 20 m up to heights of 2000 m. A typical example of these profiles, demonstrating the structure of the profiles through nocturnal inversion layers, is given. The results of an interlaboratory comparison of results are discussed.

Regional scale atmospheric pollutant modeling efforts are limited by an incomplete understanding of the importance of various processes. This study is concerned with complex and poorly understood development of nocturnal inversion layers.

The existence of inversion layers implies limitation of vertical dispersion and uncoupling of flow between layers. The classical processes for inversion development are either by radiational cooling or by subsidence. The former is the process for the development of the surface nocturnal inversion, and the latter for elevated inversion layers associated with atmospheric circulations. Largely unexplained by these processes is the development of multiple nocturnal inversion layers in the planetary boundary layer (PBL). This phenomenon has important implications in the regional modeling of pollutant dispersion and needs better definition. The objective of this analysis is to provide a data base for study of nocturnal inversion processes to aid in the regional modeling of pollutant dispersion.

FIELD EFFORT

The cooperative field effort during July 1976 with Argonne National Laboratory (SAGAMON)

was briefly described in a previous report.²⁴ The effort of the combined laboratories resulted in a data base for the meteorological and pollutant PBL structure in the late afternoon and evening under inversion formation conditions.

The site in central Illinois was over agricultural surface covers (corn and soybeans) primarily. The study extended from the surface up to heights as high as 2000 m. Argonne National Laboratory obtained extensive surface flux, radiosonde, tetron and acoustic sounding data. Pacific Northwest Laboratory (PNL) recorded detailed vertical profiles of temperature, moisture, ozone and condensation nuclei from 20 m up to heights of 2000 m using the PNL Cessna 411 aircraft as a measurement platform.

DATA ANALYSIS

Considerable progress has been made in the reduction and analysis of data. The PNL profile data has been transferred from field cassettes to 9-track standard tapes. Two dates of data have been reduced to plotted profiles and analyzed in detail.

Figure 1.9 contains an example of the multiple inversion layer soundings of atmospheric

FIGURE 1.9. Vertical Profiles of Air Temperature, Mixing Ratio, Ozone and Particle Density

PBL physical and chemical properties that PNL obtained. This is one of a sequence of profiles obtained on this date that demonstrated the consistent progressive development of the inversion layers.

The vertical resolution for these profiles is between 3 and 10 m, depending on the sensor or pollutant monitor response times. The data recording rate provided a potential vertical resolution better than 1 m. For the elevated profiles presented in Figure 1.9, about a 20-m vertical averaging has been performed to smooth the profiles.

The strong correlations between pollutant and meteorological profiles under inversion conditions is demonstrated in Figure 1.9. The interactions of the vertical source-sink distribution of pollutants and the vertical dispersion rates may be inferred from these profiles, but are more clearly demonstrated by temporal sequences of inversion formation profiles obtained in this study.

The definition of the interrelationships of the pollutant and atmospheric structure illustrated in Figure 1.9 emphasizes the importance of these processes in modeling pollutant dispersion. The additional definition of the atmospheric flow structure by Argonne measurements will allow case studies of the processes involved in the formation of these profiles.

COMPARISON OF PNL AND ARGONNE RESULTS

An information exchange meeting was held with Argonne National Laboratory during the past year to compare data for one selected date and to plan for further data analysis.

The concurrent PNL and Argonne profiles of the same variable demonstrated excellent structural correspondence. The detailed structure in the lower layers where an overlap of profile definition occurred was essentially identical. In addition, there was a close calibration agreement between the lower portions of the PNL temperature and moisture profiles and Argonne surface observations.

DISCUSSION

The comparison of profiles indicated that the combination of the data sets will provide a consistent definition of the physical and chemical structure of the atmosphere under inversion conditions in much greater detail than has been previously available. As a result, it was decided to continue the data reduction of the July 1976 field data for additional dates. A standard format was devised to allow direct combination of the results in a consistent data set for analysis of the processes involved in multiple nocturnal inversion formation.

DRY DEPOSITION OF GASEOUS POLLUTANTS FROM COAL-FIRED POWER PLANTS

J. G. Droppo and J. C. Doran

Field tests of a system to determine deposition resistances of specific pollutants and the eddy diffusivity for pollutants in the surface layer have been conducted, and the data have been analyzed. The results add considerably to the data base available for study of pollutant dry removal processes.

Since the Northwest's coal deposits represent a significant part of national energy reserves, studies of the natural dry removal processes of the air pollutants associated with emissions from coal-fired fossil fuel power plants are being conducted in increasing numbers. This study is designed to provide quantitative information for air pollutant modeling needs through in situ measurements of the dry removal rates of pollutants. The current objective is to provide a data base both for modeling dry deposition of specific pollutants (SO_2 , O_3 , NO_x , NO) and atmospheric pollutant transport processes by defining deposition resistances of specific pollutants and the appropriate eddy diffusivity for pollutants in the surface layer.

FIELD TESTS

Ozone was selected for intensive study for two reasons: 1) it is of considerable interest as a secondary pollutant from fossil fuel plants, and 2) unlike the other pollutants, the flux of ozone in the surface layer can be measured by both the gradient and eddy flux methods using current instrumentation. The deposition of SO_2 and other gaseous pollutants will also be determined to allow generalization of the results.

As indicated by an earlier study,²⁵ a mobile probe, using a high-flow Teflon intake line, is the best method of measuring ozone gradient. An automated dual line profile system was assembled for this experiment, and is shown in Figure 1.10 as it operated for ozone. Pollutant concentrations are sampled at predetermined heights on the "Roving Intake Tower." Since the dual intake system is designed to use two pollutant monitors simultaneously, any differences in the sensitivity of the monitors may be compensated for by switching the intake line positions at regular intervals determined largely by the monitor response times. The roving intake instrumentation assembled for these studies is designed to allow simultaneous measure of the dry removal rates of O_3 , SO_2 , NO_x and NO . The system can be used with other pollutant monitors that have a sufficiently fast, real-time response.

The ambient meteorological conditions are recorded from sensors on the "Profile Tower." The ozone direct flux measurement system consists of a high-flow Teflon sample line with a fixed input at the Gill anemometer on the top of the Profile Tower. The ozone and wind components time series are recorded at this height.

FIELD TEST RESULTS

A field test of instrumentation systems to detect sulfur compounds and ozone has been completed, and the data have been analyzed. The site near Yelm, WA, an abandoned farm field with a grass cover 6-8 in. high, was in the downwind region of the anticipated plume of the coal-fired power plant at Centuria, WA.

Results from filter samples indicated measurable sulfur aerosols only marginally above normal filter background concentrations. The SO_2 concentrations were very low, below or at the threshold of the SO_2 monitor during the entire field deployment period. Ozone concentrations were generally at background levels and were readily measurable by the O_3 monitor.

Summaries of the ozone dry deposition results at the Yelm site in terms of deposition velocities defined using ozone concentration values at 30 ft are given in Table 1.10. The experiments are coded as direct eddy flux compilation (F) and gradient flux compilation (G). These results are given in terms of resistance values in Table 1.11.

DISCUSSION

The reasonable consistency of the gradient and eddy flux determinations of ozone fluxes supports the validity of the methods, although further analysis is necessary to define implications for pollutant flux mechanisms. These results represent a considerable addition to the data base available for study of pollutant dry removal processes. Ozone flux rates for the near background concentrations had deposition velocities in the range 0.2 to 0.6 cm/sec.

FIGURE 1.10. Pollutant Profiler Configuration for Ozone

TABLE 1.10. Summary of Ozone Dry Deposition Results at Yelm Site, F = Eddy Correlation Method, G = Gradient Method.

Test No.	Date	Time (min)	Method	V_d cm/s	V_m cm/s	u (15m) m/s	R_l (15m)
1	6/9/77	1245-1330(45)	F	0.27	2.3	6.3	-0.085
2	6/9/77	1330-1415(45)	F	0.52	1.5	6.1	-0.10
3	6/9/77	1415-1513(59)	F	0.64	2.1	5.7	-0.11
4	6/9/77	1715-1810(55)	G	0.35	1.8	5.5	-0.09
5	6/9/77	1811-1909(58)	G	0.16	1.3	5.1	0.15
6	6/9/77	1911-2011(60)	G	0.40	4.0	3.9	0.24

TABLE 1.11. Resistance Values for Ozone Dry Deposition Results at Yelm Site, R_T = Total Resistance, R_a = Aerodynamic Resistance and R_s is the Apparent Surface Resistance ($= R_T - R_a$).

Test No.	R_T s/cm	$R_a^{(a)}$ s/cm	R_s s/cm
1	3.7	0.43	3.3
2	1.92	0.66	1.3
3	1.56	0.48	1.1
4	2.85	0.56	2.3
5	6.25	0.77	5.4
6	2.50	0.25	2.3

(a) R_a is assumed to be inverse of V_m .

The "surface resistances" in Table 1.11 are only the difference between the total momentum flux and the pollutant flux resistances. These differences may be a true surface difference or they may variously reflect differences in atmospheric transport processes between ozone and momentum. The recommended application is to determine the dry deposition

flux rates of a number of gaseous pollutants along with ozone flux by both the eddy and gradient methods. This is to provide relative gaseous pollutant deposition rates with a direct determination of the eddy diffusivity for ozone. This will provide a data base for determination of the nature of the apparent surface resistances.

THE DEPENDENCE OF DEPOSITION VELOCITY ON DISTANCE DOWNWIND OF A POINT SOURCE OF ATMOSPHERIC CONTAMINATION

T. W. Horst

The surface depletion model was used to demonstrate that the deposition velocity for a reference height of 1 m is, for practical purposes, independent of downwind distance for an elevated ($h \geq 2$ m) point-source release of atmospheric contamination. This was not found to be the case for a surface source ($h=0$).

Dry deposition can cause a significant loss of material from a diffusing plume of atmospheric contamination. The downward flux of material due to deposition is commonly parameterized as a deposition velocity v_d multiplied by the air concentration χ measured at some reference height z_d ,

$$F_d = -v_d \chi(z_d).$$

Therefore, if a vertical gradient of contamination exists, as is usually the case for a diffusing substance, the deposition velocity will be a function of the reference height.²⁶

The deposition velocity parameterization is most useful if v_d depends only on the meteorological conditions, the properties of the contaminant, and the nature of the deposition surface and, thus, is independent of distance from the source. The vertical concentration gradient, however, reflects two processes: diffusion away from the source of contamination and diffusion towards the deposition surface. The deposition velocity will be independent of distance from the source only if the vertical contaminant profile between the surface and the deposition reference height is controlled solely by the deposition flux and does not depend on diffusion away from the source of contamination.

For a nondepositing material, there is no vertical flux at $z=0$, and hence the vertical gradient at the surface is equal to zero. This condition is also closely approximated within a finite layer whose depth

increases with downwind distance x . For a depositing material, therefore, deposition alone will determine the vertical gradient up to a height which increases with x . Equivalently, deposition will determine the vertical gradient below z_d only beyond a specific downwind distance.

A practical choice for a deposition reference height is 1 m. The surface depletion model²⁷ was consequently used to investigate the ratio $\chi(1m)/\chi(1cm)$ for a depositing material, assuming a reference height of 1 cm gave a deposition velocity independent of downwind distance. The distance $x_{1\%}$ was found, beyond which the ratio $\chi(1m)/\chi(1cm)$ is within 1% of its asymptotic value at large x . Beyond $x_{1\%}$ deposition is the dominant process in the lowest 1 m. As expected $x_{1\%}$ was also found to be the distance at which $\chi(1m)/\chi(1cm)$ without deposition was within 1% of unity.

More important, it was found that for an elevated release, $h \geq 2m$, essentially all of the deposition occurs beyond $x_{1\%}$. Hence, for practical purposes, the deposition velocity at a height of 1 m is independent of downwind distance for an elevated release.

This was not found to be the case for a surface release, however. In this case the ratio $\chi(1m)/\chi(1cm)$ is negative for a large distance downwind, and significant deposition occurs before the ratio reaches the positive asymptotic value indicating dominance of the deposition process in determining the vertical profile.

Small Table

2	10m	150	100
10	10	150	500
100	150	200	2000

BEHAVIOR OF LATERAL DIFFUSION PARAMETERS

J. C. Doran, T. W. Horst and P. W. Nickola

Data from a number of field experiments have been analyzed to study the behavior of the quantity $S = \sigma_y / x \sigma_\theta$. A previously proposed relation is shown to be valid only under rather restricted circumstances, and significant variations with sampling period, averaging time, and release height are shown.

The determination of the lateral dispersion parameters of a diffusing substance in terms of measurable meteorological variables has been the subject of considerable study. Pasquill^{28,29} and Draxler³⁰ have examined the quantity $S = \sigma_y / x \sigma_\theta$ as measured in a number of field experiments. Despite the considerable scatter in the data, both have found evidence of systematic behavior, either as a function of travel time T or downwind distance x . In particular, Pasquill³¹ has suggested that S may be reasonably represented by a universal function of x which is valid independent of terrain roughness, release height, and sampling duration up to one hour.

In the present study, results from nine different field programs were examined in an effort to determine the validity of this suggestion and to gain an understanding of possible deviations from this "universal" behavior.

The application of Taylor's theorem to the turbulent diffusion process may be shown to lead to the following expression for S :

$$S = \frac{[\sigma_y]_{\tau, T}}{x [\sigma_\theta]_{\tau, t}} = \left\{ \frac{\int_0^\infty F_L(n) \left(1 - \frac{\sin^2 \pi n \tau}{(\pi n \tau)^2} \right) \frac{\sin^2 \pi n T}{(\pi n T)^2} dn}{\int_0^\infty F_E(n) \left(1 - \frac{\sin^2 \pi n \tau}{(\pi n \tau)^2} \right) \frac{\sin^2 \pi n t}{(\pi n t)^2} dn} \right\}^{1/2} \quad (1)$$

where $[\sigma_y]_{\tau, T}$ is the lateral dispersion measured over a sampling time τ at a distance downwind x which is reached in time T , and $[\sigma_\theta]_{\tau, t}$ is the variance of the horizontal wind direction measured over a sampling time τ and averaged over a time t . $F_L(n)$ and $F_E(n)$ are the Lagrangian and Eulerian turbulent spectra, respectively, and n is the frequency.

While the precise forms for $F_L(n)$ and $F_E(n)$ are not known, it may also be shown

that the qualitative behavior described by (1) is similar for a wide variety of possible spectral shapes. In particular, S should increase as either τ or t increases.

Figure 1.11 shows the variation of S with x for various combinations of τ and t for ground-level releases, as determined from five different field programs. (The Green-glow and 30-Series have been combined.) As may be seen, the values suggested by Pasquill are reasonable for shorter averaging and sampling times, but do not represent the data well for larger values of τ and t .

The effect of release height on the variation of S with x was also found to be important. If the diffusing plume is confined to a plane, then it is not unreasonable that the value of σ_θ measured in that plane would play a role in the dispersal of the material. For elevated releases, however, the situation is considerably more complicated. As the plume descends toward the ground, it encounters turbulent fluctuations which vary with height.³² It is not at all evident, then, at what elevation σ_θ should be measured to provide useful predictions of diffusion at ground level.

Figure 1.2 shows the results obtained for three release heights, where σ_y and σ_θ are measured near the ground. As can be seen, the curves for the three release heights coincide only at some distance downstream from the source, and the distance to this point increases with increasing height of the release. Near the origin, the variation of σ_θ with height has a clear effect on the behavior of S . Farther downstream, the lateral dispersion is dominated by the crosswind fluctuations near the ground, and the value of σ_θ at the release height is not a governing factor.

This interpretation is borne out by the results shown in Figure 1.13, where S is again plotted as a function of x , but σ_θ has been measured at the release height rather than near the ground. No apparent order can be seen in these curves, indicating that the

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FIGURE 1.11. Variation of $S = \sigma_y / x \sigma_\theta$ with Distance as Determined from Several Field Programs

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FIGURE 1.12. Variation of $S = \sigma_y / x \sigma_\theta$ with Downwind Distance for Three Release Heights, Hanford-67 Series. σ_θ was measured at 1.5 m elevation.

value of σ_θ at elevated release points is not a good predictor of the ground-level dispersion. Moreover, the scatter of the data about the mean values is worse than for the analyses described by Figure 1.12, particularly for the 26 m releases.

The variation of S with x was found to be considerably more complicated than suggested

by the simple relationship given by Pasquill. While those values suffice for a rough description of diffusion from a ground-level source, they are not very satisfactory for longer sampling and averaging times and are not applicable to the case of elevated releases.

Neg. 77C771-3

FIGURE 1.13. Variation of $S = \sigma_y / x \sigma_\theta$ with Downwind Distance for Three Release Heights, Hanford-67 Series. σ_θ was measured at release heights.

ISOKINETIC AIR SAMPLER

G. A. Sehmel

An isokinetic air sampler was developed for sampling both respirable and nonrespirable particles as a function of wind speed for a selected wind direction. When the air sampler is in the field, the sampler inlet is closed except when opened by a solenoid.

Airborne concentrations of both respirable and nonrespirable particles must be measured in particle resuspension and deposition experiments in the field. In these studies, airborne particles are sampled for selected wind speed increments, temperature lapse rates, wind direction, and particle size. Ideally, this air sampling should be isokinetic, and there should not be any air impact flow through the sampler when it is not activated. The objective of this study was to design a new isokinetic air sampler to meet these specifications.

DESIGN

An isokinetic air sampler inlet was designed for attachment to a standard 20 x 25 cm (8 x 10-in.) high-volume air sampler operating at 1.13 m³/min (40 cfm). In Figure 1.14, a cut-away view of the assembled inlet is shown with the inlet attached to a filter holder. The inlet is 25 cm wide, and its height can be adjusted by matching isokinetic flow and the selected wind speed increment to be sampled. The inlet cross-sectional area is held constant for the first 7.5 cm towards the filter,

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FIGURE 1.14. Isokinetic Sampler — Inlet Open

after which the top of the 16 ga aluminum inlet is angled upward to fit over the filter holder. The total distance between inlet and filter is 25 cm.

A solenoid operated closure uncovers the inlet when the air sampler is activated. The cover is shown opened in a cut-away view of the sampler. Also shown schematically are the paths of motion for "large" and "small" particles. In this case, large refers to those particles which settled on the bottom of the inlet, and small refers to those particles collected on the filter.

SAMPLER EVALUATION

The isokinetic sampler has been partially evaluated for particle loss and for wind-caused impact pressure airflow through the filter, but only without the inlet closure. The effects of the closure on isokinetic sampling have yet to be determined.

Airflow through the filter caused by wind impact pressure was determined in a wind tunnel. Results are shown in Figure 1.15 as flow rate through the filter versus wind speed for a Gelman Type-A glass fiber filter. For each wind speed, the inlet depth was adjusted for isokinetic flow for a sampling rate of $1.13 \text{ m}^3/\text{min}$.

The impact pressure flow rate increased from 2×10^{-4} to $6 \times 10^{-3} \text{ m}^3/\text{min}$ as wind speed increased from 2 to 15 m/sec. These flow rates were less than the impact pres-

sure flows, also shown in Figure 1.15 when the isokinetic inlet was not attached to the filter holder. However, the impact pressure was always a small fraction of the sampling flow rate of $1.13 \text{ m}^3/\text{min}$. Figure 1.15 also shows the impact flow rates when wind direction caused impact pressure to occur at the back of the samplers. With the inlet cover closed, impact flows should decrease toward zero.

Particle losses within the sampler were determined in the wind tunnel for monodispersed particles of 6 and $3 \mu\text{m}$ diameter and a $0.25 \mu\text{m}$ polydispersed aerosol. Percent losses are shown in Figure 1.16 as the fraction of particles entering the inlet which were not collected on the filter. Particle losses for nonrespirable $6 \mu\text{m}$ diameter particles were less than 17% and decreased with increasing wind speed. For the smaller respirable particles, losses were an order of magnitude less. The maximum measured loss for $3 \mu\text{m}$ particles was only 4%.

CONCLUSIONS

The design of the isokinetic air sampler allows sampling of both large and small particles in near isokinetic conditions. The size fractionation of this sampler could be improved by building a horizontal elutriator within the inlet length (currently 7.5 cm). The existing isokinetic sampler or possible modifications including cylindrical geometry could have application for many air pollution sampling situations.

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FIGURE 1.15. Air Flows Through Isokinetic Air Sampler Caused Only by Air Impact Velocity

Neg. 7711495-11

FIGURE 1.16. Particle Losses Within Isokinetic Air Sampler

THE RATES OF CHANGE OF POLLUTANT CONCENTRATIONS
DOWNWIND OF ST. LOUIS

J. A. Young

The atmospheric concentrations of CCl_3F , CO , SO_2 , sulfates, and several trace elements were measured in a Lagrangian mode upwind, in and downwind of St. Louis during the afternoons of several days in July and August of 1973, 1974 and 1975, to determine: 1) the emission rates of these pollutants, 2) the rates at which deposition and dilution due to atmospheric mixing caused the concentrations to decrease downwind of St. Louis, and 3) the rate of conversion of SO_2 to sulfates. Beyond 10 to 20 miles downwind of St. Louis, dilution due to atmospheric mixing decreased the concentrations $6.6 \pm 2.27\% \text{ hr}^{-1}$ (2σ) in 1973 and $4.5 \pm 1.3\% \text{ hr}^{-1}$ in 1975. The concentrations of anthropogenic particulates such as lead, bromine and zinc decreased $13.3 \pm 3.5\% \text{ hr}^{-1}$ in 1973 and $10.0 \pm 2.4\% \text{ hr}^{-1}$ in 1975 as a result of deposition and mixing. The deposition velocity for these elements was calculated to be $2.2 \pm 1.4 \text{ cm sec}^{-1}$ in 1973 and $1.8 \pm 0.7 \text{ cm sec}^{-1}$ in 1975. Beyond 40 to 50 miles downwind of St. Louis, mixing, deposition and conversion to sulfates caused the SO_2 concentrations to decrease $25 \pm 10\% \text{ hr}^{-1}$ in 1973 and $22.8 \pm 5.5\% \text{ hr}^{-1}$ in 1975. This decrease was due primarily to deposition and the SO_2 deposition velocity and was calculated to be $3.8 \pm 1.7 \text{ cm sec}^{-1}$ in 1973 and $6.2 \pm 1.3 \text{ cm sec}^{-1}$ in 1975. The rate of conversion of SO_2 to sulfates was calculated to be $3.3 \pm 1.7\% \text{ hr}^{-1}$ in 1973 and $3.7 \pm 1.2\% \text{ hr}^{-1}$ in 1975.

An urban-industrial area releases a wide spectrum of pollutants into the atmosphere. These pollutants affect not only the urban area itself, but also regions downwind. Therefore, it has become increasingly clear in recent years that the problem of air quality is of regional concern, not just local. In order to determine the impact of pollutant sources on downwind regions, it is necessary to determine the rates of processes, such as atmospheric mixing, deposition on the earth's surface, and physical and chemical reactions which cause the pollutant concentrations to change with time and distance downwind of a source. Therefore, during the Metropolitan Meteorological Experiment (Metromex), the concentrations of CCl_3F , CO , SO_2 , sulfates, and a wide spectrum of trace metals were measured in a Lagrangian mode upwind, in and downwind of St. Louis to determine the rates of these processes and the emission rates of these pollutants by the St. Louis metropolitan area.³³

EXPERIMENTAL

The concentrations of trace elements, sulfates, SO_2 , CO and CCl_3F were measured in samples collected upwind, in, and up to

130 miles downwind of St. Louis on several days in July of 1973 and 1974 and in August of 1975. Aerosol samples were collected for sulfate and trace element analysis with HiVol pumps which drew air through 4.8 cm diameter IPC-1478 filter paper at a flow rate of $1.7 \text{ M}^3 \text{ min}^{-1}$ in 1973, and through 11 cm diameter Whatman-41 filter paper at a flow rate of $0.6 \text{ M}^3 \text{ min}^{-1}$ in 1974 and 1975. These filters were analyzed by x-ray fluorescence and neutron activation analysis.³⁴ Samples were collected for SO_2 analysis by bubbling air through sodium tetrachloromercurate solutions at a rate of one liter per minute. These samples were analyzed chemically. In 1973 samples were collected for CCl_3F and CO analysis by letting air into evacuated 460 cm^3 capacity gas cylinders. Each sample was collected in roughly one second. Probably because of the short sampling time, the CCl_3F and CO results were rather erratic; in 1974 and 1975, samplers which collect air samples by slowly drawing a piston up a cylinder were used instead. These air samples were analyzed for CCl_3F with an electron capture detector and for CO with a Beckman CO monitor.

On each sampling day at least one set of air samples was collected upwind of St. Louis;

sequential samples were collected on the roof of the 11-story Federal Building in downtown St. Louis; and five to seven people equipped with mobile air samplers collected samples at locations from 10 to 130 miles downwind of St. Louis. The sampling locations were chosen each morning on the basis of predicted wind speeds and directions. Sampling generally began around 11:00 a.m. and continued throughout the afternoon. On all of the sampling days the mixed layer was capped by an inversion.

The pattern of sampling locations was changed each year on the basis of the results obtained from previous years. In 1973 an arc of sampling stations was set up 20 miles downwind of St. Louis, and every person collected one set of one-hour samples at his sampling location. Each person then moved downwind several miles and collected a set of two-hour samples on a second arc, and finally moved downwind again to collect a set of two-hour samples on a third arc. The distance they moved downwind between samples was chosen so that the average rate they traveled was about the same as the wind speed.

In 1974 a different sampling procedure proved not as useful, so the 1974 results will not be discussed in this report. In 1975 a sampling procedure more like that in 1973 was used. However, instead of sampling on three arcs downwind of St. Louis, the sampling locations were chosen to form a more uniform grid. Three sampling stations were set up on an arc 10 miles downwind of St. Louis, and one set of air samples was collected at each site. Approximately one hour after sampling began 10 miles downwind, two other people began collecting one-hour air samples at two sites on an arc 20 miles downwind. When sampling was completed 10 miles downwind, the three people moved downwind a distance determined by the wind speed and collected two-hour air samples at three locations, and then moved downwind again to collect a third set of samples. The people who began sampling 20 miles downwind also moved downwind after sampling to collect a second set of samples, and then moved downwind again to collect a third set of samples.

On several days in 1975, the PNL Cessna-411 aircraft was used to collect air samples at three altitudes in the mixed layer above the downwind air sampling stations. Each sample was collected over a 45-minute period while the Cessna was flying back and forth across the plume perpendicular to the wind.

RESULTS

The measured atmospheric pollutant concentrations showed considerable variability, with samples collected at nearby locations or samples collected sequentially at one location showing considerably different concentrations. Presumably, this variability resulted because the atmosphere is non-homogeneous and because of the relatively short sampling times employed. Since the variability complicated the determination of the rates of change of pollutant concentrations with time and distance, groups of days with similar wind speed, direction, and pollutant concentrations were averaged to increase the number of measurements and thereby obtain better values for the average pollutant concentrations. This report considers average concentrations for two periods: 22, 27 and 28 July 1973, when the wind was from the west, and 3-6 August 1975, when the wind was from the northeast. The wind speed averaged about 9 mph during both these periods.

The average concentrations of CCl_3F , CO , Pb , Zn , SO_2 and sulfates in the St. Louis plume are plotted as a function of downwind distance for the two periods in Figures 1.17 and 1.18. Each point in the figures represents the average of all the measurements made within the St. Louis plume at approximately the same downwind distance. The error limits reported are the 1σ values for the reliabilities of the averages calculated from the scatter of the individual measurements.

Dilution Due to Atmospheric Mixing

The rates of decrease in the concentrations of the conservative gases, CCl_3F and CO , were used to indicate the rate at which dilution due to atmospheric mixing caused pollutant concentrations to decrease with distance downwind of St. Louis. Molina and Rowland³⁵ have estimated that the atmospheric residence time of the fluorocarbon, CCl_3F , released primarily by aerosol spray cans, is between 40 and 140 years. The atmospheric residence time of CO has been estimated to be one to three years.³⁶ The concentrations of CCl_3F and CO decreased very rapidly between the Federal Building and 10 miles downwind due to vertical mixing through the mixed layer. On 3-6 August the CCl_3F concentrations averaged 6 times the concentrations 12 miles downwind. On all days the concentrations decreased much more slowly beyond about 10 miles downwind. On 22, 27 and 28 July 1973, the average rate of decrease of the CCl_3F and CO concentrations with time and distance downwind of St. Louis

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FIGURE 1.17. Pollutant Concentrations as a Function of Distance on 22, 27 and 28 July 1973

Neg. 776571-1

FIGURE 1.18. Pollutant Concentrations as a Function of Distance on 3-6 August 1975

was $6.6 \pm 2.7\% \text{ hr}^{-1}$. On 3-6 August 1975, the average rate of decrease from the two gases was $4.5 \pm 1.3\% \text{ hr}^{-1}$. The error limits reported throughout this report are 2σ error limits. They were calculated from the deviation of the points from the lines in Figures 1.17 and 1.18. Since CCl_3F and CO are conservative gases, these rates of decrease should be approximately equal to the rates at which the concentrations of the other pollutants were decreased by atmospheric mixing.

Deposition Velocity of Particulate Materials

The concentrations of particulate materials emitted by St. Louis decreased downwind of St. Louis as a result of atmospheric mixing and deposition on the earth's surface. Therefore, the rates of decrease of the concentrations of anthropogenic elements such as lead, bromine and zinc downwind of St. Louis can be used to calculate the deposition velocities of particulate materials after using the conservative gas concentrations to correct for dilution due to mixing. Lead and bromine are emitted primarily in automobile exhaust, while zinc is emitted primarily by industrial processes. Since, except near highways, lead and bromine are present on particles too small to undergo appreciable gravitational settling, their dry deposition on the earth's surface results from turbulent diffusion. A Lundgren impactor sample collected on 17 July 1973, at the Federal Building showed about 70% of the lead and bromine on particles smaller than 0.3μ in diameter and only 2 or 3% on particles larger than 10μ . Zinc was present on larger particles, with 27% present on particles smaller than 0.3μ , and 25% present on particles larger than 10μ . However, the zinc concentrations decreased at essentially the same rate downwind of St. Louis as did the concentrations of lead and bromine. The concentrations of these elements first decreased fairly rapidly downwind as a result of vertical mixing, and then decreased much more slowly farther downwind. Beyond about 10 miles downwind the concentrations of these three elements decreased an average of $13.3 \pm 3.5\% \text{ hr}^{-1}$ in 1973 and $10.0 \pm 2.4\% \text{ hr}^{-1}$ in 1975. The decrease in the concentrations, therefore, appears to have been due about equally to deposition and mixing.

The deposition velocity is defined as the flux to the earth's surface divided by the atmospheric concentration above the surface. After correction for dilution due to atmospheric mixing, the flux to the surface is equal to the rate of decrease in the mass in the column of air above the surface. The masses of lead, bromine and zinc in the

mixed layer were calculated by multiplying the surface concentrations in gcm^{-3} times the depth of the mixed layer in cm. Vertical profiles indicated that beyond about 20 to 40 miles downwind of St. Louis the pollutant concentrations were fairly uniform throughout the mixed layer (Figure 1.19). The rate of change with downwind distance of the masses cm^{-2} of these elements was then corrected for dilution due to atmospheric mixing and divided by the surface concentrations to give the deposition velocities. The average deposition velocities for these three elements were calculated to be $2.2 \pm 1.4 \text{ cm sec}^{-1}$ in 1973 and $1.8 \pm 0.7 \text{ cm sec}^{-1}$ in 1975.

Rate of Conversion of SO_2 to Sulfates

Ground-level SO_2 concentrations reached a maximum about 20 miles downwind of St. Louis in 1973 and about 40 miles downwind in 1975, presumably because SO_2 from smokestacks passed over the sampling sites closer to St. Louis. Beyond these distances the SO_2 concentrations decreased rapidly due to atmospheric mixing, deposition on the earth's surface, and conversion to sulfates in the atmosphere. The SO_2 concentrations decreased $25 \pm 10\% \text{ hr}^{-1}$ in 1973 and $22.8 \pm 5.5\% \text{ hr}^{-1}$ in 1975.

The sulfate concentrations also reached a maximum downwind of St. Louis, but decreased

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FIGURE 1.19. Lead Concentration as a Function of Altitude Downwind of St. Louis

at a slower rate farther downwind than did the other pollutants because of the conversion of SO_2 to sulfates. At greater distances when the SO_2 concentrations become negligible, the rate of decrease in the sulfate concentrations should approach that of the lead and bromine concentrations, since the sulfate is present on the same size particles as the lead and bromine.

Since the sulfate concentrations would decrease at about the same rate as the lead, bromine and zinc concentrations if it were not for the conversion of SO_2 to sulfates, the amount of SO_2 which converted to sulfates can be calculated from the difference between the decrease in the sulfate concentrations and the decrease in the lead, bromine and zinc concentrations. When this was done, SO_2 to sulfate conversion rates of $3.3 \pm 1.7\% \text{ hr}^{-1}$ and $3.7 \pm 1.2\% \text{ hr}^{-1}$ were calculated for 1973 and 1975, respectively.

Deposition Velocity of SO_2

The processes which cause the SO_2 concentrations to decrease downwind of a source are mixing, deposition and conversion to sulfates. Since the total rate of decrease in the SO_2 concentrations and the rates of decrease due to mixing and conversion have been determined earlier, the rate of deposition can be determined by difference. When this was done, SO_2 deposition velocities of $3.8 \pm 1.7 \text{ cm sec}^{-1}$ for 1973 and $6.2 \pm 1.3 \text{ cm sec}^{-1}$ for 1975 were calculated with the method used earlier for lead, bromine and zinc.

It can be seen that beyond about 20-40 miles downwind of St. Louis the decrease in the SO_2 concentrations was due primarily to deposition. In 1975 SO_2 decreased $22.8\% \text{ hr}^{-1}$, but mixing caused only a $4.5\% \text{ hr}^{-1}$ decrease, and conversion only a $3.7\% \text{ hr}^{-1}$ decrease.

CHARACTERIZATION OF SUBSTANCES IN PRODUCTS, EFFLUENTS, AND WASTES FROM COAL CONVERSION PROCESSES

M. R. Petersen

Researchers at Pacific Northwest Laboratory (PNL) are investigating materials from synthetic fossil fuel processes. During the past year, samples have been collected from the Solvent Refining Coal Pilot Plant (SRC-I mode), Lignite Gasification Pilot Plant, Eyring Research Institute Gasifier, and Hanna III In Situ Coal Gasification Experiment. Inorganic and organic analyses have been performed, and comparisons of the data show some important differences in the potential emissions.

The objectives of this program are to identify and quantify the constituents occurring in products, effluents, and emissions from coal conversion processes that warrant consideration in the development of control measures. Sampling and analyses of emissions from four coal conversion processes during this year have shown the variety of constituents which are present in the processes. The following paragraphs detail some of our sampling methods and findings to date at these sites.

SOLVENT REFINING COAL PILOT PLANT

During the final stages of a long run to obtain several thousand tons of solid solvent-refined coal (SRC-I mode), samples were

collected at the Solvent Refining Coal Pilot Plant at Fort Lewis. This sampling trip included collecting the usual solid and liquid samples, but primary emphasis was placed on collecting gaseous and particulate samples. Since the mass balance for mercury indicated it was escaping in some volatile form, sampling during this field trip included collecting volatile forms of mercury in the process gas lines. No mercury species, however, were detected. The lines were also sampled for volatile arsenic species, but none were found. After the plant resumed operation in the SRC-II mode, several sets of water samples were collected and analyzed for compounds soluble in methylene chloride. Phenol, the cresols and xylenols were the most concentrated organic species at 20-100 ppm (see

the accompanying chromatogram -- Figure 1.20). Aromatic hydrocarbons were present up to 20 ppm for some individual compounds such as methylnaphthalenes or phenanthrene. The compounds present in the SRC-II water did not differ markedly in kind or concentration from the SRC-I process.

Chemical fractions (SRC-I) from the light oil, wash solvent, and process solvent were given early in the year to biologists at PNL for biotesting studies. Of all the fractions, the polynuclear aromatic hydrocarbons and the basic fractions of the process solvent were the most active in the Ames test, and further studies with these two fractions are underway.

LIGNITE GASIFICATION PILOT PLANT

Several sets of samples (including gaseous, particulate, and aqueous effluents) were collected at the Lignite Gasification Pilot Plant in Rapid City, South Dakota. Gaseous mercury was detected in the preheater vent gas and in the overhead gas from the gasifier, and some

arsenic was present in the same overhead gas. Water quenching of this overhead gas removed the mercury and most of the arsenic. Organic analyses of the various quench waters and preheater condensate showed that the concentration organic compounds were mostly of the sub-ppm level. Quench water of gases from the acceptor regenerator contained a variety of oxygenated species including aldehydes, ketones, alcohols, and phenols. Naphthalene was the single major compound in the gasifier quench water, but the water also contained a number of other polynuclear aromatic compounds in sub-ppm concentrations. Condensate from the preheater vent line contained many volatiles evolving directly from the coal during drying, including aliphatic hydrocarbons and a series of resin-acid related hydrocarbons, such as abietane ($C_{20}H_{36}$), abietatriene ($C_{20}H_{30}$), ferruginol ($C_{20}H_{30}O$), and retene ($C_{18}H_{18}$). A reconstructed chromatogram of an extract of that condensate is shown in Figure 1.21.

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FIGURE 1.20. Reconstructed Chromatogram of a Methylene Chloride Extract of Water from Surge Reservoir at Solvent Refining Coal Pilot Plant

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FIGURE 1.21. Reconstructed Chromatogram of a Methylene Chloride Extract of Condensate from Preheater Vent at Lignite Gasification Pilot Plant

EYRING RESEARCH INSTITUTE GASIFIER

Water, solid and gas samples were collected from this small bench scale (40 lb/hr) gasifier. The water and solid samples contained sub-ppm concentrations of phenols, alkanes, and polynuclear aromatic hydrocarbons. We suspect that these compounds found in the aqueous phase were adsorbed on the fine particulates suspended in the quench water.

HANNA IN SITU COAL GASIFICATION EXPERIMENT

The *in situ* coal gasification experiment was conducted over about one month, and special emphasis was focused on sampling volatile inorganic species present in the raw product gas. During each sampling period, about 15 ft³ of gas was filtered and passed through impinger solutions. Some selective absorption traps were also used for arsenic and mercury species. The following elements were below the detection limits of about 3-100 ppb: Fe, Pb, Br, Rb, Cd, and Se. Mercury was found to be present in the gas phase only as the volatile metal, and no detectable arsenic species were found.

TRACE ELEMENT CONCENTRATIONS DOWNWIND OF MILWAUKEE AND CHICAGO

J. A. Young and A. J. Alkezweeny

The atmospheric concentrations of several trace gases and trace elements were measured downwind of Chicago and Milwaukee in August 1976 to determine the rates at which atmospheric mixing, deposition, and chemical reactions cause the pollutant concentrations to change with time and distance downwind of these cities. The experiments were conducted in an approximately Lagrangian frame of reference with two instrumented aircraft and surface samplers. This report will discuss the measurements of trace elements and the conservative gases, CO and CCl₃F. Vertical profiles of trace element concentrations indicate that vertical mixing was less vigorous over Lake Michigan than over land. Trace element concentrations decreased about 6% per hour downwind of Milwaukee. Many trace elements appeared to be present on larger particles when they were measured on a boat on Lake Michigan than when they were measured on land. Trace element emission rates by Chicago were much higher than by Milwaukee.

An urban area releases a wide spectrum of pollutants, many of them toxic, into the atmosphere. The effect of these pollutants on downwind regions depends on the rates at which atmospheric mixing, deposition on the earth's surface, and chemical reactions cause the pollutant concentrations to change with time and distance downwind of the urban area. In order to measure the rates of these processes, a Lagrangian experiment was designed to measure pollutant concentrations as a function of distance downwind of Milwaukee over Lake Michigan, where there are no pollutant sources to complicate the determination.

A major goal of this experiment was to determine the rate of conversion of SO₂ to sulfates. However, as many other pollutants as possible were measured at the same time because 1) the rate of conversion of SO₂ to sulfates depends on the concentrations of species which either react with SO₂ or catalyze the conversion of SO₂ to sulfates; 2) certain pollutants can serve as tracers of the behavior of other pollutants; and 3) many of these pollutants also have harmful effects on the environment. The concentrations of SO₂, NO_x, O₃, CO, CCl₃F, hydrocarbons, sulfates, trace elements, and particulates were measured in this experiment. This report will discuss the measurements of trace elements CO and CCl₃F; another report by A. J. Alkezweeny and J. A. Young will discuss the other measurements. (a)

Most of the measurements of pollutant concentrations were carried out aboard the PNL DC-3. On each sampling day measurements of particle concentrations and lapse rate were used to determine the depth of the mixed layer. Pollutant concentrations were then measured at two or more altitudes in the mixed layer approximately 10 miles downwind of Milwaukee while the DC-3 was flying horizontally perpendicular to the wind across the Milwaukee plume. During sampling, the wind speed and direction were measured by the Global Navigation System on board the DC-3. The DC-3 was then moved downwind the distance necessary to sample the same air, and sampling traverses were again made across the plume at two or more altitudes. This procedure was repeated until either the air passed over a major pollution source or the DC-3 ran short of fuel after 7 to 8 hours. Pollutant concentrations were measured north or west of Milwaukee over land on three days, east of Milwaukee over Lake Michigan on two days, and northeast of Chicago over Lake Michigan on one day.

Air filter samples for trace element analysis were collected aboard the DC-3 with a Staplex pump which drew air through a 5 cm diameter IPC-1478 filter paper at 1.4 M³ min⁻¹. Samples were also collected on IPC filter paper aboard the PNL Cessna-411 in the mixed layer upwind of Milwaukee and above the mixed layer on days the Milwaukee plume was measured. Air samples for CO and CCl₃F analysis were

(a) A. J. Alkezweeny, et al., "Transport and Transformation of Pollutant in the Lake Michigan Area," p. 1.10.

collected aboard the DC-3 by allowing air to bleed slowly into one evacuated 460 cm³ cylinder during each traverse of the plume. Air samples were also collected at ground level for CO, CCl₃F, SO₂, sulfate and trace element analysis.

VERTICAL PROFILES

Vertical profiles of trace element concentrations indicated that there was considerably less vertical mixing over Lake Michigan than over land. On 24 August 1976, the Milwaukee plume traveled westward over land. Thirteen miles west of the eastern edge of Milwaukee, the trace element concentrations decreased considerably with altitude, except for the zinc and sulfate concentrations, which varied erratically with altitude (Figure 1.22). However, by 35 and 49 miles downwind,

the trace element concentrations had become fairly uniform throughout the mixed layer (Figure 1.23).

On 27 August the Milwaukee plume traveled eastward over Lake Michigan. Ten miles downwind of Milwaukee the trace element concentrations showed a slight decrease with altitude (Figure 1.24). However, 46 miles downwind, many of the elements showed large concentration maximums at 1000 ft. Apparently the layer containing these high concentrations had not been sampled 10 miles downwind. By 82 miles downwind these high concentrations had spread to ground level and 2000 ft, but not to 3000 ft.

TRACE ELEMENT FLUXES

Trace element fluxes downwind of Milwaukee and Chicago were calculated by multiplying the

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FIGURE 1.22. Trace Element Concentrations as a Function of Altitude 13 Miles West of Milwaukee on 24 August 1976

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FIGURE 1.23. Zinc Concentrations 13, 35 and 49 Miles Downwind (West) of the Eastern Edge of Milwaukee on 24 August 1976

FIGURE 1.24. Zinc Concentration Downwind (East) of Milwaukee on 27 August 1976

wind speed times the trace element concentrations and integrating over the depth of the layer sampled and over the length of the sampling traverse. Wind speeds and trace element concentrations were assumed to vary linearly between sampled altitudes. On all days but one the highest measured altitude was near the top of the mixed layer. Since the sampling traverses 10 miles downwind of Milwaukee extended beyond the edges of the Milwaukee plume, the calculated fluxes should be only slightly smaller than the true total fluxes in the Milwaukee plume. Farther downwind, however, the Milwaukee plume may have spread beyond the ends of the sampling traverses, causing the calculated fluxes to be a somewhat smaller fraction of the total fluxes.

The calculated trace element fluxes downwind of Milwaukee decreased about 6% per hour. No significant difference in the rate of decreases between the days the plume traveled at about 25 mph over water and the days the plume traveled at about 9 mph over land was detected. If the decrease in the calculated fluxes had been due entirely to deposition, then the rate of decrease in the flux would have equaled the rate of deposition. Dividing the rate of flux decrease by the average surface concentrations would give a deposition velocity of about 1.2 cm sec^{-1} . However, since at least some of the calculated flux decrease was due to mixing of pollutants above or beyond the ends of the sampling traverses, 1.2 cm sec^{-1} represents the upper limit for deposition velocity. The decrease in the calculated fluxes of the conservative gases CO and CCl_3F was to have been used to determine the fraction of the calculated flux decrease due to mixing so that true deposition velocities could be calculated; however, the CCl_3F concentrations

were near or below the detection limit and could not be used for this purpose. The CO concentrations are being measured now.

The trace element fluxes upwind of Milwaukee were calculated from the upwind concentration measurements and subtracted from the downwind fluxes to obtain the Milwaukee emission rates. The emission rates calculated for Milwaukee on 23 August are reported in Table 1.12. Emission rates calculated for St. Louis by this method during August of 1975^(a) are also reported for comparison. Since no upwind concentrations were measured on the day concentrations were measured downwind of Chicago, the calculated fluxes downwind of Chicago were reported as emission rates in Table 1.12. The upwind fluxes would have to be subtracted from these numbers to obtain the true emission rates. The calculated emission rates for Chicago and St. Louis were clearly much higher than those for Milwaukee, and the Chicago emission rates were generally higher than those for St. Louis.

TABLE 1.12. Trace Element Emission Rates by Milwaukee, Chicago and St. Louis

Element	Trace Element Emission Rates in g/sec		
	Milwaukee 23 August 1976	Chicago 19 August 1976	St. Louis August 1975
Ca	153	757	870
Mn	2.8	29.7	4.9
Fe	69	798	197
Zn	9.4	57.9	20
Pb	10.4	137	50
Br	3.8	25.8	11

(a) J. A. Young, "The Rates of Change of Pollutant Concentrations Downwind of St. Louis," p. 1.35.

PARTICLE SIZE DISTRIBUTIONS

The averages of three trace element particle size distributions measured aboard a fishing boat on Lake Michigan and the averages of three size distributions measured on land downwind of Milwaukee are reported in Table 1.13. Arsenic, lead, manganese, and zinc appeared to be present on larger particles when the size distributions were measured aboard the fishing boat than on days when the distributions were measured on land. The concentrations on the 0.3 to 1 μ stage increased at the expense of concentrations on the backup filter. There is no evidence that the larger particle sizes measured on Lake Michigan resulted from higher humidities. The relative humidities were 57% and 59% on two days the distributions were measured over water, and 56% and 73% on the two days the distributions were measured on land. Spray in the air may have either caused particle growth or affected

the operation of the impactor, although there was no evidence that spray got into the impactor.

Though sulfur dioxide is believed to react with particles containing metals such as manganese and iron and to deposit sulfate on these particles, the sulfate particle size distribution downwind of Milwaukee was considerably smaller than the size distributions of calcium, iron, manganese, zinc, and vanadium. The rate of deposition of sulfate on particles might be expected to be proportional to the surface area of the particles rather than the mass, however. Rough calculations indicate that the total surface area of the metals in each size range is approximately proportional to the mass of sulfate in the size range, indicating that the sulfate on the particles could have resulted from heterogeneous reactions with sulfur dioxide.

TABLE 1.13. Trace Element Particle Size Distributions Downwind of Milwaukee Measured on Land and on Lake Michigan Using a Lundgren Impactor

Stage	50% Cutoff Diameter Microns	Percent of Total Concentration															
		As		Ca		Fe		Pb		Mn		S		V		Zn	
		Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water	Land Water
1	10	1	1	12	20	12	11	1	1	5	6	2	2	10	8	9	6
2	3	3	4	30	32	40	29	6	6	10	11	2	1	16	8	16	21
3	1	2	4	16	12	16	21	7	5	10	19	3	5	11	12	10	16
4	0.3	12	26	9	4	5	9	6	34	12	26	6	44	9	16	12	38
Filter	0.3	81	65	33	32	29	30	80	53	67	37	88	48	54	56	50	21

AIR QUALITY IMPACTS OF PROJECTED COAL UTILIZATION IN THE WEST

D. S. Renné, D. L. Elliott and W. J. Eadie

Pacific Northwest Laboratory (PNL) has estimated the short-range and long-range impacts on air quality in the western United States from two coal development scenarios projected to the year 2020. Short-range impacts for the Pacific Northwest were computed using standard point source diffusion models which require meteorological data from the region in which the models are being applied. Long-range impacts were estimated using a regional transport, transformation, and removal model applied to the western United States. Results show that the most significant air quality constraint to future coal-fired power plant development in the western United States is the Regulations for the Prevention of Significant Deterioration as described in the Clean Air Act Amendments of 1977.

This report is abstracted from a part of the National Coal Utilization Assessment (NCUA) sponsored by the Assistant Administrator for Environment and Safety of the U.S. Energy Research and Development Administration. PNL is responsible for the analysis of coal utilization under various growth scenarios for the Pacific Northwest (Washington, Oregon, Idaho, and Alaska). The objective of NCUA is the construction of a national perspective on coal utilization through the aggregation of six regional studies, all made on the same basis. PNL analyzed the impact of four coal growth scenarios for Alaska, Washington, Oregon, and Idaho (Federal Region Ten) over the period 1975 to 2020. The two national scenarios most relevant to Federal Region Ten are the Recent Trends and the High Coal Electric. These result in compound growth rates of coal-fired generation of 5.7% and 7.7%, respectively, over the 45-year period. Air quality impacts for the two scenarios were studied by developing current and projected emission inventories for the Pacific Northwest (excluding Alaska). These data were incorporated into both short-range and long-range air quality diffusion models.

CURRENT EMISSIONS AND AIR QUALITY

Air quality impacts for the two development scenarios were estimated by developing a current and projected emission inventory for the Pacific Northwest and incorporating these emissions into air quality diffusion models. The emission inventory for existing sources is available through the National Emission Data System (NEDS) of the Environmental Protection Agency (EPA) and includes point and area source emission rates for all criteria pollutants (sulfur dioxide, particulates, oxides of nitrogen, carbon monoxide, hydrocarbons, and photochemical oxidants).

The inventory, which consists of data compiled by state and local agencies during the years 1972 through 1975, shows that major sulfur dioxide (SO₂) emissions occur from large point sources in the Puget Sound and Willamette Valley regions and in northern Idaho. Particulate emissions are high in the Seattle-Tacoma, Portland-Vancouver, and Spokane regions, and these areas as well as others in the Willamette Valley have been designated Air Quality Maintenance Areas (AQMA's), requiring special planning to assure that the National Ambient Air Quality Standards for particulates are not exceeded by the year 1990. The Portland-Vancouver AQMA, in addition, must plan for the control of SO₂, oxides of nitrogen (NO_x), and carbon monoxide (CO).

PROJECTED EMISSIONS

Emissions for a reference 3000 MW_e coal-fired power plant have been estimated assuming the emissions are equivalent to the Federal New Source Performance Standards (NSPS), and the plant operates at a 60% load factor. This information has been used to estimate additional emissions in the Pacific Northwest projected to the year 2020 for both the Recent Trends and the High Coal Electric energy development scenarios. Table 1.14 summarizes the emissions from a reference 3000 MW_e power plant and the total increase in emissions by state.

PROJECTED AIR QUALITY

Short-Range Impacts

Short-range annual average air quality impacts for various counties around the Pacific Northwest using emissions from a reference 3000 MW_e power plant were estimated from application of the Climatological Dispersion Model (CDM).³⁷ Data from various meteorological stations in the region were used as input to the model. In most cases data were obtained from National Weather Service stations where hourly or three-hourly wind and cloud cover data are gathered. These data are then converted into "STAR" Program data by the National Climatic Center in Asheville, NC, for direct use in the CDM. The "STAR" Program data consist of a joint wind speed, direction, and stability frequency function. In a

TABLE 1.14. Total Electrical Generation Capacity and SO₂ Emissions^(a) for the Year 2020 Scenarios

Reference: 3000 MW_e Power Plant Emissions

SO ₂ Emissions	2.45 x 10 ³ gm/sec(a)		
Particulate Emissions	2.03 x 10 ² gm/sec(a)		
NO _x Emissions	1.42 x 10 ³ gm/sec(a)		
Total Electrical Generation Capacity		SO ₂ Emissions for the Year 2020 Scenarios	
Recent Trends			
Washington	12,300 MW _e	349.3 x 10 ³ tons/yr	10.05 x 10 ³ gm/sec
Oregon	8,000 MW _e	227.2 x 10 ³ tons/yr	6.53 x 10 ³ gm/sec
Idaho	3,500 MW _e	99.4 x 10 ³ tons/yr	2.86 x 10 ³ gm/sec
Total	23,800 MW _e	675.9 x 10 ³ tons/yr	19.44 x 10 ³ gm/sec
High Coal Electric			
Washington	27,300 MW _e	777.3 x 10 ³ tons/yr	22.30 x 10 ³ gm/sec
Oregon	17,500 MW _e	497.0 x 10 ³ tons/yr	14.29 x 10 ³ gm/sec
Idaho	8,500 MW _e	241.4 x 10 ³ tons/yr	6.94 x 10 ³ gm/sec
Total	53,300 MW _e	1,513.7 x 10 ³ tons/yr	43.53 x 10 ³ gm/sec

(a) Based on Federal NSPS. Use of NSPS eliminates the emission variations due to coal supply.

few cases, tower meteorological data, collected for the purpose of obtaining permits to construct coal or nuclear power plants, were also made available to this study. These data utilize vertical temperature gradients instead of cloud-cover information to obtain the stability classes. The results of the assessment for SO₂ are shown in Table 1.15.

Short-term maximum concentrations were estimated primarily from the results of a GE study³⁸ and described in detail in an unpublished Argonne report, which discusses the results of the analysis as well as the methodology employed. The analysis shows that for a 60% load factor, the maximum 24-hour SO₂ concentrations are estimated to be 300 µg/m³ for a 5.0 m/sec wind speed and 250 µg/m³ for a 2.5 m/sec wind speed. These values are somewhat below the National Ambient Air Quality Standards (NAAQS) for this pollutant.

Long-Range Sulfur Transport

A significant impact of coal-fired power plant emissions is the long-range transport of sulfur dioxide and its transformation to sulfate aerosols before being removed from

the atmosphere by wet and dry deposition processes. These sulfate aerosols can have significant health effects, and over long periods of time they can build up in the soil profile and have important ecological and agricultural implications, even at large distances from the source.

A regional transport, transformation, and removal model, which utilizes actual upper air meteorological data to account for the temporal and spatial variability of the wind field, has been applied to the year 2020 coal-related energy development scenario for the entire western United States (West of the 100th meridian). An earlier version of this model has been applied to coal scenarios in the Northwest,³⁹ and the present version is applied to coal-fired power plant emissions in the northeast United States.⁴⁰ This section describes the application of this model to the western United States for the year 2020 scenario and the results of the model calculations for a one-month period of meteorological data.

Candidate siting areas for coal-fired power plants and coal-gasification plants in the western United States are based on data supplied by Argonne National Laboratory (North Dakota, South Dakota, Nebraska, and Kansas), Lawrence Berkeley Laboratory (California), Los Alamos Laboratory (Rocky Mountain states), and Oak Ridge National Laboratory (Texas and Oklahoma). Fifty-six candidate sites for new facilities to 2020 have been specified for the western region. Table 1.16 gives a breakdown of existing versus projected emissions used in the assessment for the western United States.

Ground-level air concentrations and surface depositions of SO₂ and sulfates from existing facilities and projected coal-related facilities were computed using October 1974 meteorological data. October was chosen as a typical worst-case month to exemplify the maximum impact of SO₂ emissions on regional air quality because this month is characterized by lighter mean wind speeds than the annual average for

TABLE 1.15. Maximum Annual Average Concentrations Calculated for a 3000 MWe Plant^(a)

Meteorological Station	Maximum SO ₂ Concentration µg/m ³ (b)	Distance and Direction of Maximum from Source
Bellingham, WA	2.6	10 km N
Ephrata, WA	1.7	10 km N, 10 km S
Hanford, WA ^(c)	7.1	5-10 km SE
Hoquiam, WA	1.6	20-40 km NW
Kelso, WA	1.6	25-40 km NNW
Moses Lake, WA	3.2	10 km NNE
Spokane, WA	1.4	25-35 km NE
Toledo, WA	2.1	10 km SSE
Astoria, OR	0.8	5-10 km S
Boardman, OR ^(d)	2.9	5 km E
North Bend, OR	2.0	25 km S
Ontario, OR	2.7	10 km W
Pebble Springs, OR ^(d)	5.7	5 km E
Pendleton, OR	1.9	5-10 km SE
Trojan, OR ^(d)	6.4	5 km S
Boise, ID	3.8	5-10 km SE
Idaho Falls, ID	2.2	5-10 km NE
Mountain Home, ID	2.6	5-10 km SE
Pocatello, ID	1.4	5-30 km NE, 5 km SE

^(a)Based on NSPS standards. Emission rate for a 3000 MWe plant with a 60% load factor is 85.2 tons/yr.

^(b)Stack height = 244 m. Grid resolution is 5 km.

^(c)Stability classified as a function of wind speed only by day/night; no account was made for cloud cover.

^(d)Tower data with stabilities based on temperature change with height.

TABLE 1.16. Total SO₂ Emissions Used in the Western Region Assessment

	SO ₂ Emissions, kilotons/yr
Existing (Industrial + Power Plants)	3647.5
Year 2020 "High Coal Electric" Scenario	3367.0
Total	7014.5

most of the western United States.⁴¹ Thus, the computed concentrations for October 1974 are assumed to equal or exceed typical annual average concentrations, since higher concentrations (on a regional scale) and air pollution episodes are normally associated with periods of low wind speeds and shallow mixing heights.⁴² Of course, the patterns shown may vary somewhat from annual patterns due to seasonal variations in prevailing wind directions.

Maximum annual average air concentrations of SO_2 allowed under the National Ambient Air Quality Standards (NAAQS) are $80 \mu\text{g}/\text{m}^3$. State standards must at least meet the NAAQS and may be more restrictive. Figure 1.25 shows the estimated existing SO_2 concentrations. The highest concentrations are primarily in areas with large smelters, refineries, or power plants. The maximum existing concentrations calculated for the western region are $15 \mu\text{g}/\text{m}^3$ in southeast Arizona because of the large smelters in that area. This level is well below the NAAQS maximum allowable annual concentrations ($80 \mu\text{g}/\text{m}^3$) and state standards. However, the model calculations

are averaged for an area of 4075 km^2 , and much higher concentrations are actually being observed in the vicinity of these sources.⁴³ This situation could also exist for other areas with large point SO_2 emissions in this assessment as well. Also, the estimates may understate actual conditions since mobile sources and small stationary sources have not been included.

Figure 1.26 shows the combined SO_2 concentrations of the existing plus the High Coal Electric scenario to the year 2020. Although the background concentrations from the industrial and other non-coal related SO_2 emitters can be expected to change considerably by the year 2020, this analysis shows where problem areas can develop under the assumed siting patterns. For the most part, new candidate siting areas are located where existing concentrations are low. However, the analysis shows that regional scale interactions of concentrations greater than $1 \mu\text{g}/\text{m}^3$ would cover a significant portion of the West, and this could pose problems for regulations preventing significant deterioration (see "Summary and Conclusions" of this report).

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FIGURE 1.25. Estimated SO_2 Concentrations for Existing Large Point Sources West of 100°W . Values represent average for October 1974 meteorological data.

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FIGURE 1.26. Estimated SO₂ Concentrations for Existing Sources West of 100°W and the High Coal Electric Scenario Projected to the Year 2020. Values represent average for October 1974 meteorological data.

Figure 1.27 shows the combined sulfate concentrations resulting from the existing plus the High Coal Electric scenario to the year 2020. Largest sulfate concentrations are calculated to be 5 to 6 µg/m³. In contrast to SO₂ concentrations, the sulfate concentrations decrease much more slowly with distance from the source. Thus, regional scale interactions are more apparent with sulfate than with SO₂ concentrations, as sulfate concentrations of 3 µg/m³ or greater cover large areas in the Great Plains, Southwest and Northwest.

NAAQS for sulfates have not been established. However, there is some evidence that sulfates can cause health problems, and some states have established their own air quality standards for sulfates (e.g., Montana and North Dakota, where the maximum allowable annual sulfate concentration is 4 µg/m³). Thus, sulfate concentrations could limit future siting in some regions if emission levels up to the Federal New Source Performance Standards are permitted.

SUMMARY AND CONCLUSIONS

Although the results of this study show no apparent violation of the National Ambient Air Quality Standards in the western United States due to intensive coal development, additional analyses are required in local regions of complex terrain to improve the estimates, particularly where concentrations can be potentially high. In addition, the results of this study should be examined in the context of the Clean Air Act Amendments of 1977 recently enacted by the Congress. These amendments include regulations for the Prevention of Significant Deterioration, mandating limits to incremental increases in SO₂ and particulate concentrations in three types of regional Classes. Table 1.17 shows the allowable increases for each Class.

It is evident from the results shown in this study, which can only be considered as preliminary at this time, that the Clean Air Act Amendments present a potential constraint on energy development in the West

Neg. 77E217-4

FIGURE 1.27. Estimated Sulfate Concentrations for Existing Sources West of 100°W and the High Coal Electric Scenario Projected to the Year 2020. Values represent average for October 1974 meteorological data.

TABLE 1.17. Allowable Air Quality Increments Specified in Clean Air Act Amendments of 1977

Pollutant	Averaging Time	Allowable Increments, $\mu\text{g}/\text{m}^3$		
		Class I	Class II	Class III
SO ₂	Annual	2	20	40
	24-hour max.	8	91	182
	3-hour max.	25	512	700
Part.	Annual	5	19	37
	24-hour max.	10	37	75

that reaches far beyond the National Ambient Air Quality Standards. This is particularly true of Class I areas, which are established to be protected areas with little allowable development (National Parks and Wilderness areas of greater than 6000 acres are mandated to be Class I areas by the Clean Air Act Amendments). Regulatory constraints such as this warrant additional, more detailed assessments of energy development in the West, using best state-of-the-art modeling and analytical techniques.

PLUME TRAJECTORIES USING SATELLITE IMAGERY

N. S. Laulainen

The feasibility of using high resolution satellite imagery as an aid to defining the transport of particulate air pollution plumes and the determination of turbidities for widespread haze layers is discussed. Limitations of the technique are also noted.

Recent work has demonstrated the usefulness of satellite imagery for the detection of particulate air pollution plumes.^{44,45,46,47} In particular, the Landsat satellites have been providing high resolution, multi-spectral images of various portions of the earth's surface since late 1972. These satellites orbit the earth sun-synchronously at an altitude of ~1000 km, making 14 revolutions of the earth each day with crossings of the equator at 0942 hr local time. Every 18 days, a given satellite roughly repeats its traverse over a given area. The images, providing resolution detail to 100-200 m, cover a square surface area roughly 185 km by 185 km.

This report describes efforts to assess the feasibility of using high resolution satellite imagery as an aid to defining the transport of power plant particulates in the atmosphere and to take advantage of the digi-

tal image processing expertise available at Pacific Northwest Laboratory (PNL). This study was intended to support Multi-State Atmospheric Power Production Pollution Study (MAP3S) field programs in the Northeast.

Microfilm images taken by Landsats I and II over Lake Michigan were scanned to select those images showing some type of plume development. One image for lower Lake Michigan taken on 10 June 1973 (Image ID 1322-16045) was selected for detailed analysis with a variety of computer enhancement techniques. A black-and-white composite, made up of images from the four multi-spectral scanner (MSS) bands (0.5-0.6 μm , 0.6-0.7 μm , 0.7-0.8 μm , and 0.8-1.1 μm), is shown in Figure 1.28. Individual plumes originating from the shoreline are easily discernible out to 50-100 km, where they merge into an overall haze. There is also evidence in this photo of albedo vari-

FIGURE 1.28. Black-and-White Composite of Lower Lake Michigan from a Landsat I Image Taken on 10 June 1973 is Shown. Pollutant plumes are readily identified emanating from the shoreline.

ations of the surface water, indicated by some of the streak patterns near the western edge of the lake and by the fact that ship wakes near the center of the photo are easily recognized.

This particular example points out some of the difficulties of interpreting satellite imagery, viz. the variety of factors which can influence the discrimination of plumes against the underlying surface, in this case water. Some of the factors which affect overall image contrast include the spectral albedo of the lake surface through water turbidity or water depth (nearshore) variations, the amount of radiation backscattered by the plume, and diffusive light scattering by atmospheric molecules and haze layers, the latter being most important in heavily polluted regions.

The spectral dependence of light scattering by aerosols is also illustrated in Figure 1.28. Condensed water plumes, which are dominated by particles/droplets larger than 1-2 μm , are nearly neutral in their spectral scattering characteristics, while smoke plumes, which are dominated by particles smaller than 1 μm , scatter light preferentially in the blue. Thus, smoke plumes should show up in the 0.5-0.6 μm and 0.6-0.7 μm bands while condensed water plumes (like clouds) should appear equally well in all spectral bands.

Since each spectral channel is calibrated, it is possible, in principle, to determine aerosol optical depth (turbidity) from the upwelling radiance measurements. Analysis of this type has been applied to the study of dust storm aerosol⁴⁸ and to regional and global mapping of atmospheric turbidity.^{49,50,51}

Ground-truth turbidity measurements, made simultaneously during satellite overpass, provide an independent measure and probable error for this technique.⁵¹ The method is most effective over low-albedo surfaces such as water. Turbidity values obtained from background or well-mixed urban haze by this method are reasonable, considering the number of assumptions about aerosol scattering phase functions, aerosol size distribution and aerosol composition that must be made because these quantities are fairly well defined and do not change rapidly with time. The situation for obtaining meaningful plume turbidity is questionable since, not only is the aerosol size distribution changing with time, but the condensed water plume also changes with time through dispersion and evaporation.

Use of satellite images for detection of particulate air pollution plumes and for estimating turbidity from areas of diffuse, well-mixed pollution appears promising. Additional images need to be examined to define the variability of surface albedo in order to extract the most information about plume dimensions and turbidities of widespread haze blobs. It would also be useful to speed up turnaround time for image reproduction -- at present it is on the order of one to two months. An idea, which is presently being explored by PNL, is to study the feasibility of additional ground receiving facilities.

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PLUME RISE -- COMPARISON AND USE OF BRIGGS' FORMULAS

D. R. Drewes

Several widely used plume rise prediction equations are compared, and figures are presented to show the extent of the differences between them. One of the equations, which is applicable for buoyant plumes in a neutral atmosphere, is considered further. Graphs are presented to show the rise predicted under a range of meteorological and source conditions. These results can be used to estimate plume parameters in other applications.

In most atmospheric work involving plumes from point sources, it is extremely important to consider the effects caused by the rise of the plume. Plume rise is dependent in a complex (and not well understood) manner upon many factors, including atmospheric stability, wind speed, source height, heat content, and entrainment of outside air by the plume. A number of equations for plume rise have been presented in the literature, and some of the more widely used are those discussed by Briggs.^{52,53,54} Buoyant plumes in unstratified surroundings are found to fit the "2/3 Law of Rise" near the source (the so-called first stage of rise):

$$\Delta h = 1.6 F^{1/3} x^{2/3} / u$$

where Δh is the plume rise (cm) above the source height, F is the initial buoyancy flux divided by $\pi \rho$ ($\text{cm}^4 \text{ sec}^{-3}$), x is the downwind distance (cm), and u is the wind speed (cm sec^{-1}). The constant, 1.6, is dimensionless and represents a best fit to observations of this particular type of plume; except for this concession to empiricism, the equation is primarily theoretical.

At some distance downwind, entrainment of outside air is assumed to have occurred to such an extent that the plume rise is slowed or stopped. Several equations are presented for this second stage of rise.

Case 1. In the most rigorously developed equation, the transition point x^* (to the second stage) is dependent upon the stack height h_s :

$$x^* = 8.6 \times 10^{-3} F^{2/5} h_s^{3/5}$$

The second stage of rise is then given according to

$$\Delta h = 1.6 F^{1/3} x^{2/3} R(x/x^*) / u$$

where

$$R(x/x^*) = \left(\frac{2}{5} + \frac{16x}{25x^*} + \frac{11x^2}{5x^{*2}} \right) / \left(1 + \frac{4x}{5x^*} \right)^2$$

Case 2. A second parameterization, valid for fossil-fueled plants with 20 MW or greater heat emission, is applied at distances over $10h_s$. Briggs claims better fit to observations for this equation than for the previous one:

$$\Delta h = 1.6 F^{1/3} (10h_s)^{2/3} / u$$

Case 3. For other types of sources, a "conservative approximation" to Case 1 is applied at distances greater than $3x^*$ (as defined above):

$$\Delta h = 1.6 F^{1/3} (3x^*)^{2/3} / u$$

Case 4. For buoyant plumes in a neutral atmosphere, Briggs claims good approximation when the formula

$$\Delta h = 1.6 F^{1/3} (3.5x^*)^{2/3} / u$$

is applied at distances greater than $3.5x^*$. In this case x^* is redefined as

$$x^* = \begin{cases} 0.014 F^{5/8} & \text{if } F < 5.5 \times 10^9 \text{ cm}^4 \text{ sec}^{-3} \\ 2.145 F^{2/5} & \text{if } F > 5.5 \times 10^9 \text{ cm}^4 \text{ sec}^{-3} \end{cases}$$

Figure 1.29 presents several graphs of the predictions of the 2/3 Law and each of the second stage equations for a variety of values of F and u ; the value of F shown at the top of each column applies to the three graphs in the column, and the value of u at the right side of each row applies to each graph in the row. The figure reveals that Cases 3 and 4 give the least difference in predicted rise. The difference decreases as F increases and depends on the stack height. In the case of a 150 m stack (assumed in the figure), the difference is about 10% for high buoyancy fluxes. These two cases also predict the least rise of the four equations.

The conditions for which Case 4 is applicable are of particular interest in many applications, so this case was considered further. Calculations were made for various buoyancy flux and wind speed values, and the results are presented in Figures 1.30 and 1.31. Use of the equation in modeling applications requires a knowledge of F . The nature of F , however, makes it a difficult quantity for which to specify a meaningful value, either from simple plume observations or from a conceptual basis. The main value of these figures, then, lies in the estimation of F . If a wind speed is known (or assumed), the figures can be used to determine a value for F which gives (approximately) the observed or desired plume rise characteristics. Thus, the figures provide a useful tool for plume rise modeling applications.

FIGURE 1.29. Comparisons of Plume Rise Predictions Using Briggs' Equations for Several Wind Speeds and Plume Buoyancies

FIGURE 1.30. Plume Rise Predictions for Various Buoyancies at Several Wind Speeds

FIGURE 1.31. Plume Rise Predictions for Various Wind Speeds at Several Plume Buoyancies

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FISSION AND FUSION

- **Particle Resuspension and Translocation**
- **Air Pollution Dry Deposition**
- **Comprehensive Study of Atmospheric Diffusion Prediction from Meteorological Parameters**
- **Alternate Fuel Cycle Technologies/Thorium Fuel Cycle Technologies (AFCT/TFCT)**
- **Hemispheric Pollution Behavior Studies of Chemical and Radiation Substances**
- **Fallout Rates and Mechanisms**
- **Effects of Inadvertent Weather Modification from Large Energy Releases**
- **Hanford Nuclear Energy Center**
- **ALE/Ecosystems**

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

Research activities at PNL include studies of atmospheric diffusion and deposition of material on the earth's surface, resuspension of particulate matter, the climatic impacts of waste heat and water vapor, and the regional and global transport of material. The diffusion studies consist of the modeling of vertical diffusion from a surface source, the comparison of Gaussian diffusion-deposition models, and the final compilation of a data volume of field diffusion experiments to be used for model evaluation and validation.

WIND RESUSPENSION EXPERIMENTS

G. A. Sehmel and F. D. Lloyd

The effects of wind speed, particle size, and atmospheric stability on wind resuspension rates are being determined in three field experiments using both six-micrometer and submicrometer tracer particles. The effect of long-term weathering is also being determined as part of these experiments.

Though wind stresses will resuspend or suspend surface contamination such as plutonium and americium and other pollutants, the transport rates for these materials are unknown. Some resuspension rates are known as a function of wind speed for submicrometer tracer simulant particles,¹ but the effects on resuspension of tracer particle diameter and atmospheric stability and the changes in resuspendibility with time are unknown. Thus, three field experiments are being conducted to determine the effects of wind speed, particle diameter, and atmospheric stability on resuspension. Long-term weathering effects are also being studied.

Results from these experiments will give the initial resuspension rate dependency on wind speed, source particle diameter, and atmospheric stability for transport on nonrespirable particles. The experiments will continue to determine the long-term weathering effects on respirable particle resuspension rates.

The first experiment is determining long-term changes in resuspension rates as a function of wind speed increments using submicrometer tracer particles. The measuring device, which uses impactor cowl air samplers,² is shown in Figure 2.1. Tests began on 3 October 1973 when a calcium molybdate tracer was deposited. Resuspension rates have been determined from measurements of airborne concentrations of both respirable and nonrespir-

able particles since that time. Resuspension is still occurring from the sagebrush and cheatgrass test area.

Neg. 7711495-13

FIGURE 2.1. Submicrometer Tracer Source and Resuspension Sampling Tower

In the second experiment, resuspension of larger 6- μm tracer particles is being determined at a cheatgrass area with the equipment schematically shown in Figure 2.2. Collected airborne samples from the initial experiment are being analyzed for the experiment started on 22 June 1977. The results from this experiment will be the first indication of the effect that particle diameter has on the resuspension rate. A second experiment has been started to determine the initial weathering effect for these larger particles.

The third experiment is determining resuspension rates as a function of atmospheric stability for submicrometer tracer particles by using the equipment shown schematically in Figure 2.3. For these long-term experi-

ments, atmospheric stabilities are approximated by the temperature lapse rate shown. Suspended tracer is sampled as a function of wind speed increments and stability as the wind blows across the tracer from 190 to 260°. Since actual sampling times are relatively short compared to the length of time air samplers are in the field, air sampler inlets^(a) are only opened during sampling periods. Closing the inlets minimizes flow through filters caused by air impact pressure for other wind speeds, directions, and stabilities. Since the inlets are automatically closed, experimental results will also indicate the resuspension rates for transport on nonrespirable particles as a function of the controlled variables.

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FIGURE 2.2. Six-Micrometer Tracer Source and Resuspension Sampling Tower

(a) See G. A. Sehmel, "Isokinetic Air Sampler," p. 1.32.

OFFSITE PLUTONIUM RESUSPENSION NEAR HANFORD

G. A. Sehmel

Airborne plutonium concentrations were measured for offsite resuspension by sampling only southwest winds near the Prosser barricade in the Hanford area. High southwest wind caused the resuspension of plutonium which resembled weapons grade.

Plutonium resuspension is usually studied onsite at DOE installations because of the number of plutonium source areas present. However, since plutonium resuspension can also occur from areas offsite due to accumulated fallout and onsite low level releases, a field experiment was conducted to determine airborne plutonium concentrations caused by wind resuspension from offsite near the Hanford area.

Resuspension of plutonium was studied near the Prosser barricade about 19 to 20 km southeast (130 to 160°) from the fuel processing areas. Airborne solids were collected with filters, at varying heights. Air was sampled only when the wind was blowing from 190-260° offsite towards the Hanford area. All southwest winds were continuously sampled for nonrespirable particles with rotating cowl inertial collection systems.¹ Respirable particles were sampled with particle cascade impactors for wind speed increments of 3 to 5, 5 to 7, and 7 to 11 m/sec at a height of 1.5 m.

Airborne plutonium concentrations in $\mu\text{Ci}/\text{cm}^3$ and $\mu\text{Ci}/\text{g}$ of collected solid are shown in Figure 2.4.¹ Concentrations are shown as a function of wind speed increments for plutonium collected on the impactor backup filter, the 1.1 plus 2.0 μm stages, and the 3.3 plus 7 μm stages.

Airborne plutonium concentrations increased 1 to 2 orders of magnitude as wind speed increased from a low range of 3 to 5 to a high of 7 to 11 m/sec. For wind speeds below about 5 m/sec, airborne submicrometer plutonium concentrations increased nearly linearly with wind speed. Above 5 m/sec, plutonium concentrations increased with wind speed to the 4.2 power.

In other portions of Figure 2.4, selected straight lines suggesting wind speed dependencies are shown only for wind speeds above 5 m/sec. Airborne concentrations increased with wind speed up to the 9.3 power.

Nonrespirable airborne plutonium blowing from offsite onto the Hanford area was also measured^{1,3} by collecting all nonrespirable solids in cowls at each test height.

Plutonium-239 concentrations in $\mu\text{Ci}/\text{g}$ and fluxes in $\mu\text{Ci}/(\text{m}^2 \text{ day})$ for nonrespirable particles are shown in Table 2.1. Concentrations ranged from 1.3×10^{-7} to 2.1×10^{-7} $\mu\text{Ci}/\text{g}$. Horizontal flux calculations were made for both the total times wind was between 3 and 11 m/sec and 190° to 260° and for the total time cowl air samplers were in the field. When the shorter time period (3 to 11 m/sec winds) was used for calculating the horizontal plutonium flux, fluxes ranged from 3.9×10^{-6} to 1.4×10^{-6} $\mu\text{Ci}/(\text{m}^2 \text{ day})$.

The data in Figure 2.4 show that plutonium is resuspended from offsite locations and that resuspension is dependent on high wind speed. Data cross comparisons show a comparable airborne plutonium flux on nonrespirable particles from offsite at Hanford, onsite at Hanford U-Pond, and onsite at Rocky Flats for time periods investigated.¹

Most airborne plutonium collected appears not to have originated from fallout; most resembles weapons grade plutonium.^{4,5} Plutonium isotopic ratios ($^{240}\text{Pu}/^{239} + ^{240}\text{Pu}$) in atom percent for these nonrespirable samples were 6.10 ± 0.02 at 0.3 m height, 6.31 ± 0.02 at 2 m height, and 6.28 ± 0.03 at 5.8 m height. In comparison, the isotopic ratio determined from a sample of forest fire smoke plume near Mt. St. Helens, WA, was 13.82 ± 0.05 . Isotopic ratios for respirable particles sampled near the Prosser barricade were not determined.

Neg. 776114-1

FIGURE 2.4. Airborne ^{239}Pu Concentrations Near Prosser Barricade at Hanford from 21 April to 29 June 1976 when Sampling Only 190 to 260° Winds

TABLE 2.1. Plutonium Transport on Nonrespirable Particles from Offsite near the Prosser Barricade on the Hanford Reservation

Sampling Height, m	^{239}Pu on Airborne Solids		Airborne ^{239}Pu Nonrespirable Flux, $\mu\text{Ci}/(\text{m}^2 \text{ day})$ for Times Of	
	dpm/g	$\mu\text{Ci/g}$	Only for 190 to 260° Winds, 3 to 11 m/sec	For Total Field Time
0.3	0.29	1.3×10^{-7}	3.9×10^{-6}	8.0×10^{-7}
2	0.46	2.1×10^{-7}	4.0×10^{-6}	8.3×10^{-7}
5.8	0.32	1.5×10^{-7}	1.4×10^{-6}	2.8×10^{-7}

RESUSPENSION AND TRANSPORT OF RESPIRABLE
PARTICLES ON NONRESPIRABLE PARTICLES

G. A. Sehmel

Respirable particles as small as $0.2\ \mu\text{m}$ diameter can be dislodged from larger, nonrespirable particles when a nonrespirable particle ($>1\ \text{mm}$) is dropped only a few cm. This fact suggests that plutonium particles attached to nonrespirable soil particles may be subsequently transported as respirable particles.

Plutonium on airborne and surface soils has been classified as respirable and nonrespirable in order to determine the potential inhalation concentrations or to determine source characteristics for resuspension. Although plutonium can be attached to most soil particle sizes,¹ plutonium is usually transported by submicrometer plutonium particles attached to larger host soil particles. (The submicrometer size is inferred from collected dpm, rather than from direct measurement of discrete particles.)

The potential resuspension source changes in physical properties are important as plutonium is transported. Since nonrespirable soil particles can be shed from larger soil particles upon impact,⁶ it is possible that small particles can become airborne when a larger host particle strikes a solid surface such as the ground. In this way, respirable plutonium might become airborne. The objective of this study was to determine if attached respirable soil particles could become airborne when host particles hit the ground.

The release of submicrometer soil particles from larger host soil particles was determined with an optical particle counter^(a) and a clean air hood. For the measurements, the inlet to the optical counter was placed within the operating clean air hood so that no particles in the background air were sampled.

Three large rocks were selected for the test. After they were picked up from the ground, the rocks were rotated to release easily detached adhering particles. Afterward, they were dropped in the clean air hood.

To determine if attached particles were released, the host rock was dropped 51 cm to impact on a solid surface near the optical counter inlet sampling at $0.31\ \text{m}^3/\text{min}$. The

cloud of released particles was counted as it passed through the optical counter. Subsequently, the counting rate decreased to background.

The total number of particles released during four drops was determined from the flow rates and recorded counts. It was assumed that all particles shed from the host particle were drawn into the optical particle counter. The optical particle counter does not size the entire $0.31\ \text{m}^3/\text{min}$ but only a small fraction of $0.094\ \text{cm}^3/\text{sec}$. This small sampling volume, however, is within the center of a cylindrical sampling volume. In the present analysis, it was assumed that released particles were uniformly distributed throughout the cylindrical sampling volume. In this case, the actual counts, determined by the optical counter, were multiplied by the scaled ratio of $0.31\ \text{m}^3/\text{min}$ divided by the flow rate of $0.094\ \text{cm}^3/\text{sec}$. Thus, one particle counted was equivalent to 5.5×10^4 particles released.

The total number of particles released from three rock particles dropped from a 51 cm height is shown in Figure 2.5 as a function of small diameter increments ranging from 0.17 to $0.79\ \mu\text{m}$. The number of particles released was large and decreased during subsequent droppings. The total number of particles released is shown as a function of particle mass in Figure 2.6. The increase with mass to the 0.63 power suggests a host particle surface-to-volume release relationship.

The majority of the particles released when the rock struck a solid surface ranged in size from 0.1 to $0.8\ \mu\text{m}$ in diameter. This result suggests that airborne particle size distributions may be modified by any sampling technique in which particles strike solid surfaces. The resuspension source characteristics of particles will also be influenced by the continual shedding and redeposition of nonrespirable particles.

(a) Active Scattering Aerosol Spectrometer, Particle Measuring Systems, Inc., Boulder, CO.

Neg. 7711495-7

FIGURE 2.5. Number of Particles Released as a Function of Released Particle Diameter from Host Particles

Neg. 7711495-15

FIGURE 2.6. Number of Particles Released as a Function of Host Particle Mass

AIRBORNE SOIL CONCENTRATIONS

G. A. Sehmel and K. L. Richmond^(a)

Airborne soil concentrations downwind of an isolated field were measured during one dust storm. Concentration increased as the friction velocity increased, but the increase was a function of particle diameter.

Since plutonium in the environment is usually resuspended on a host soil particle, some study of soil suspension can give insight into the physics of plutonium resuspension and transport. A dust storm is the ideal time for such a study because of the volume of soil transported. Previous studies have shown that airborne concentration of respirable particles increase downwind;⁷ however, they have not related the increase to air stresses at the suspension source. The objective of this study was to relate airborne soil concentrations to the friction velocity at an isolated blowing field.

Though the relationship of airborne soil concentrations and friction velocity is unknown for conditions other than during one dust storm, airborne soil concentrations did increase in that instance with the 2.6 to 6.4 power of friction velocity. The experiment will be repeated to determine the generality of these results.

Soil suspension experiments were conducted in an isolated field approximately 1 km in length. The field had been burned and all vegetation removed. Upwind suspension was minimal because an alfalfa crop was growing in the upwind field.

Airborne soil concentrations were related to the air friction velocity which was measured with a Gill UVW anemometer. Concentrations were measured for 14 diameter ranges from 0.3 to 2.9 μm with an optical particle counter.^(b) Both airborne concentrations and friction velocity were measured at a height of 2.5 m.

Airborne concentrations were measured for four friction velocities, and the typical results are shown in Figure 2.7 for four particle diameter ranges. For each diameter range, airborne concentrations increased with the 2.6 to 6.4 power of friction velocity. The friction velocity power dependency for all diameter ranges measured is shown in Figure 2.8.

Neg. 7711495-17

FIGURE 2.7. Airborne Soil Concentration as a Function of Particle Diameter and Friction Velocity

(a) K. L. Richmond was a NORCUS student from the University of Washington during January-June 1976.
(b) Active Scattering Aerosol Spectrometer, Particle Measuring Systems, Inc., Boulder, CO.

Neg. 7711495-14

FIGURE 2.8. Airborne Soil Particle Concentration Dependency on Particle Diameter and Friction Velocity [Number/cm³ ∝ (U_{*}) Power]

Airborne concentrations were only a low power (2.6 and 3.4) of friction velocity for particle diameters from 0.35 to 0.55 μm. In contrast, for the next diameter range (from 0.55 to 0.70 μm), the power increased rapidly to 5.0. For larger particle diameters, the power increased to 6.4.

Resuspension characteristics differed for particles with a diameter of about 0.6 μm. For smaller particles, the friction velocity power increased with particle diameter to the 1.5 power; that is

$$\chi \propto (U_*)^f (d)^{1.5}$$

For larger particles, the friction velocity power increased with particle diameter to the 0.16 power; that is

$$\chi \propto (U_*)^f (d)^{0.16}$$

AIRBORNE PLUTONIUM-239 TRANSPORT MEASURED FROM THE 125-m HANFORD METEOROLOGICAL TOWER

G. A. Sehmel

Airborne ²³⁹Pu concentrations and fluxes were measured at six heights from 1.9 to 122 m at the Hanford meteorological tower. The data show that ²³⁹Pu was transported on nonrespirable and "small" particles at all heights.

Studies at Rocky Flats and Hanford have shown that plutonium is resuspended.^{8,1,9} For the time periods investigated in these studies, airborne concentrations were above fallout levels but below 0.03% of the maximum permissible air concentration, and plutonium was transported on both respirable and nonrespirable particles to at least a 30 m height. However, the maximum plume height was unknown and the relative transport on respirable and nonrespirable particles could only be estimated. The objective of this study was to measure the average relative plutonium concentrations and fluxes on both nonrespirable and "small" particles for sampling heights up to 122 m more directly.

This study determined that ²³⁹Pu was transported mainly on large particles. Therefore, any prediction of how far resuspended respirable particles will travel downwind must rest on a determination of the amount of plutonium attached on nonrespirable particles. Since these particles are transported at least to a 125-m elevation, models are also needed to describe how nonrespirable particles are transported and plutonium is detached.

Air samples were taken continuously with particle impactor cowl air sampling systems (see Figure 2.15)(a) located at six sampling heights from 1.9 to 122 m on the Hanford 125-m meteorological tower. Samples were collected for all wind speeds and directions for the

(a) G. A. Sehmel, "Field Particle Deposition Measurement by Mass Balance Using Ambient Radioactive Particles and Airborne Soil," p. 2.13.

time period between 13 August and 12 November 1976. In this report, particles settling within the cowl inlet and those collected as interstage impactor losses (nonrespirable) are called "large" cowl collected particles. The sum of all particles collected on the cascade impactor collection surfaces plus backup filter are called "small" particles.

Airborne ^{239}Pu concentrations per unit air volume, concentrations per gram of airborne solid, and horizontal fluxes were calculated. Figure 2.9 shows concentrations in $\mu\text{Ci}/\text{cm}^3$ for small particles. Local versus fallout resuspension is also indicated since concentrations were largest at a 1.9 m sampling height and decreased with sampling height to 91 m. It is unclear why the concentration at 122 m was greater than at 91 m. A similar increase in concentration was observed at Rocky Flats at sampling heights between 10 and 30 m.¹ The increased airborne concentration at 122 m might be caused in part by a greater ^{239}Pu $\mu\text{Ci}/\text{g}$ concentration at 122 m than at 91 m.

Concentrations per gram of airborne solids were greater on small than on large particles, as Figure 2.10 shows. The $\mu\text{Ci}/\text{g}$ for small particles did decrease with height up to 91 m. This decrease might be explained by sampling both "more contaminated" locally resuspended particles and "less contaminated" soil blowing in from a greater distance. For

Neg. 7711495-8

FIGURE 2.9. Airborne ^{239}Pu Concentrations at the Hanford Meteorological Tower From 13 August to 12 November 1976 (Impactor Stages Plus Backup Filter)

Neg. 7711495-12

FIGURE 2.10. ^{239}Pu on Airborne Solids Collected at the Hanford Meteorological Tower From 13 August to 12 November 1976

large particles, the maximum $\mu\text{Ci}/\text{g}$ occurred for a sampling height of 30 to 60 m. This plume concentration profile might be explained if contaminated large particles were resuspended at a distance upwind and deposited between the resuspension and sampling sites or if "less contaminated" local soil particles were resuspended, and the average of both upwind and soil decreased the average $\mu\text{Ci}/\text{g}$ from upwind. These explanations carry one qualification: that these average profiles are for samples collected for all wind speeds and directions rather than from only one source.

Figure 2.11 shows average ^{239}Pu fluxes in $\mu\text{Ci}/(\text{m}^2 \text{ day})$ for both large and small particles. Two isokinetic sampling assumptions were made for these calculations: 1) for the large particles, particle inertia was assumed sufficient to cause particle collection within the cowl inlet for all wind speeds, and 2) for small particles, the particles were assumed to be without inertia and followed airflow. Thus for small particles, the flux was calculated from the product of measured concentration and average wind speed at each sampling height.

For all heights, the ^{239}Pu flux was greater on the large particles. As shown on the right side of Figure 2.11, the flux on small particles ranged from only 9.6 to 46% of the total calculated flux.

Neg. 7711495-16

FIGURE 2.11. Airborne ^{239}Pu Fluxes at the Hanford Meteorological Tower From 13 August to 12 November 1976

IMPROVED PREDICTIVE DRY DEPOSITION VELOCITY CORRELATION

G. A. Sehmel and W. H. Hodgson

An improved predictive deposition velocity correlation was developed by including a single dimensional term with non-dimensional groupings in the correlation.

Though two predictive dry deposition velocity resistance correlations reported in previous studies were equally good statistically,¹⁰ there was no basis for selecting which correlation to use since both were based on the same experimental data. Because predictions from these two correlations were not identical, however, this study attempted to develop an improved predictive deposition velocity resistance correlation.

With the correlation model developed, predicted dry deposition velocities are bounded by a lower limit for small particles ($<10^{-2} \mu\text{m}$) and an upper limit for large particles ($>0.1 \mu\text{m}$). In between, predictions are less

certain. Predictions can be calculated from the model for most variables except available surface area, but geometrical terms are needed to describe the available area in a canopy.

The term correlated in this study, Int , is based on a dimensionless description of particle deposition and transport within the 1 cm adjacent to a surface; above 1 cm is described by meteorological diffusion.¹¹

Least squares techniques were used to develop the improved correlation of surface mass transfer resistances derived from wind tunnel tests.¹⁰ The new correlation for predicting this surface mass transfer resistance is

$$\begin{aligned} \text{Int}_{1977W} = \exp \left\{ -378.051 + 16.498 \ln(\text{Sc}) \right. \\ \left. + \left[\ln \tau^+ \right] [-11.818 - 0.2863 \ln \tau^+ \right. \\ \left. + 0.3226 \ln\left(\frac{d}{z_0}\right) - 0.3385 \ln\left(\frac{D}{z_0 u_*}\right) \right] \\ \left. - 12.804 \ln d \right\} \quad (1) \end{aligned}$$

The multiple correlation coefficient is 0.92 and all coefficients are statistically significant at the 99% level.

Deposition velocities were predicted using this mass transfer resistance correlation combined with the meteorological mass transfer resistances from heights of 1 cm to 1 m.¹¹ Predicted velocities, K_{1-M} , are shown in Figure 2.12 for a 1-m concentration reference height and a 30 cm/sec constant friction velocity. Two lower limit curves were also calculated, and in this case, the correlation was replaced by an expression to account only for mass transfer by Brownian diffusion in the 1 cm next to the deposition surface. These deposition velocities increase with a decrease in particle diameter. Also shown is a curve for Brownian diffusion in the lower 0.01 cm. Predicted deposition velocities for these small particles are obviously dependent upon Brownian diffusion.

An upper limit to dry deposition velocities was also calculated. In this case, the mass transfer resistance in the 1 cm adjacent to the deposition surface was assumed to be zero.

NOMENCLATURE

- a = particle radius, cm
- d = particle diameter, cm
- D = Brownian diffusion coefficient cm²/sec, see Equation 2
- Int = integral mass transfer resistance next to surface, dimensionless
- k = Boltzmann's constant, 1.32×10^{-16} erg/(molecule °K)
- p = pressure, cm of mercury (76.0 cm used)
- Sc = Schmidt number, ν/D
- T = temperature, °K (296°K used)
- u_* = friction velocity, cm/sec
- v_t = monodispersed particle gravity settling velocity, cm/sec
- z_0 = aerodynamic surface roughness, cm
- μ = air viscosity, g/(cm·sec) [1.78×10^{-4} g/(cm·sec) used]
- ν = kinematic viscosity, μ/ρ
- ρ_p = particle density, g/cm³ (1.5 g/cm³ used)
- τ^+ = dimensionless relaxation time, $\frac{\rho_p d^2 u_*^2}{18\mu \nu}$

where Brownian diffusivity was calculated¹² from

$$D = \frac{kT}{6\pi\mu a} \left\{ 1 + \frac{10^{-4}}{pa} [6.32 + 2.01 \exp(-2190pa)] \right\} \quad (2)$$

Neg. 7711495-2

FIGURE 2.12. Predicted Deposition Velocities at 1 m for $u_* = 30$ cm/sec and Particle Densities of 1, 4 and 11.5 g/cm^3

FIELD PARTICLE DEPOSITION MEASUREMENT BY MASS BALANCE USING AMBIENT RADIOACTIVE PARTICLES AND AIRBORNE SOIL

G. A. Sehmel

Airborne flux changes and radioactive soil particles across a sagebrush and cheatgrass area are being measured with air samplers mounted on towers at heights from 0.3 to 20 m. Deposition will be calculated from vertical profile changes at two towers located 305 m apart.

Though air pollution particle removal rates by dry deposition can be predicted from correlating wind tunnel data,^(a) direct field measurements have often lacked adequate con-

trol of variables or have been based on modeling concepts which had not been validated. Atmospheric transport and diffusion models have not been validated thoroughly

(a) See G. A. Sehmel and W. H. Hodgson, "Improved Predictive Deposition Velocity Correlation," p. 2.11.

even for distances between 1 to 10 km and not at all at greater distances. Much research has compared predictions from different transport models, rather than obtained validation data. Some also feel that predictions from simple transport models (i.e., complete mixing to the mixing height and transporting downwind by the average air velocity) compared to concentration data may be as good as some predictions from more complex transport models. Obviously transport model validation and concentration validation data are needed to increase confidence in transport model predictions.

One potential problem with most existing field deposition data and transport models is that dispersion coefficients (σ 's) and deposition velocities have always been measured simultaneously. For instance, when σ_z is assumed, a constant deposition velocity is calculated to fit transport data. A constant deposition velocity of 1 cm/sec may often be assumed even though deposition mass transfer rates should be a function of particle diameter.

To decrease some uncertainty in deposition mass transfer calculations, particle deposition between towers was determined for a surface release of submicrometer particles.

There was 65% deposition for a plume travel distance of 91 m across sagebrush.¹³ Also, a plume loss of 80% was reported for a surface release with a mean diameter of 1 to 1.5 μm .^{14,15} These surface release losses are larger than usually expected. A deposition experiment was begun in October 1977 which uses ambient radioactivity as an airborne tracer to simulate plume depletion for a source far upwind.

This experiment is designed to measure deposition of both respirable and nonrespirable particles across a 305 m fetch of cheatgrass and sagebrush. Since fallout concentrations are assumed uniform in the crosswind direction, calculated deposition results from this experiment will be independent of tracer release crosswind integration, noncontainment of tracer plume height, and equality of turbulent diffusion transport required in other experimental measurement techniques.

The experiment has been set up near the Hanford area's southwest perimeter, as Figure 2.13 shows. Air samplers on the site are automatically activated when the wind direction is $233^\circ \pm 45^\circ$. This direction was selected to emphasize fallout measurement and minimize potential effects from local resuspension and transport from the 200 areas.

Neg. 7711495-6

FIGURE 2.13. Location of Air Sampling Towers to Measure Particle Deposition

The two air sampling towers, shown schematically in Figure 2.14, are activated when the air direction ($233^\circ \pm 45^\circ$) and wind speed increments (3 to 5, 5 to 7, and 7 to 11 m/sec) are identical at each tower site. In this manner, the experiment is designed to measure changes in horizontal flux profiles and deposition for the same air mass as air is transported from the upwind to the downwind tower.

Neg. 7711495-4

FIGURE 2.14. Deposition Measurement by Sampler Activation with Same Wind Direction and Speed at Each Tower

Air samplers are solenoid operated isokinetic air samplers, (a) particle cascade impactors (shown in Figure 2.15) with cowl inlets which can only rotate within $233^\circ \pm 45^\circ$,¹ or particle cascade impactors without cowls which are faced horizontally into the 233° wind.

Neg. 744581-12

FIGURE 2.15. Rotating Cowl and Impactor

(a) G. A. Sehmel, "Isokinetic Air Sampler," p. 1.32.

LAGRANGIAN SIMILARITY MODELING OF VERTICAL DIFFUSION FROM A SURFACE SOURCE

T. W. Horst

The crosswind-integrated surface air concentrations of a diffusing tracer were calculated from Lagrangian similarity theory and compared to the data from Project Prairie Grass at distances from 50 m to 800 m downwind of the surface tracer source. The agreement between theory and data is quite good, supporting the assumption that inert contaminants have an eddy diffusivity corresponding more closely to that of heat than momentum. The best fit to the data was also found for an assumed vertical distribution of tracer proportional to $\exp(-z^r)$ where $r = 1.5$, rather than a Gaussian value of 2.

Chaudhry and Meroney have recently shown that the vertical spread of a material diffusing from the surface can be predicted by Lagrangian similarity theory, if it is assumed that the eddy diffusivity of the material corresponds to that of heat rather than momentum.¹⁶ They compared the predicted mean height of the diffusing material $\bar{z}$ with that measured 100 m downwind of the source during the Prairie Grass diffusion experiments.

By assuming a vertical distribution of the material equal to

$$\chi(z) = \chi(z=0)\exp[-(z/b\bar{z})^r] \quad (1)$$

no loss of the material

$$\int_0^\infty u(z) \chi(z) dz = Q \quad (2)$$

and by using this definition of $\bar{z}$

$$\bar{z} = \frac{\int_0^\infty z \chi(z) dz}{\int_0^\infty \chi(z) dz} \quad (3)$$

the crosswind-integrated air concentration $\chi(z)$ may also be predicted. Here z is the vertical coordinate, Q is the rate of release of the tracer material, and u is the horizontal wind speed. Elliot has investigated the vertical distributions of tracer measured at 100 m downwind of the source and found that a value of 2 for r is an overestimate except in fairly stable conditions, that 1.5 was more appropriate for near-neutral conditions, and that r was less than 1.5 for unstable conditions. He also found that the continuity condition (2) was satisfied by the measured distribution of χ and u .¹⁷

During Project Prairie Grass, the surface air concentrations were measured at distances of 50 m, 100 m, 200 m, 400 m, and 800 m

downwind of the source. Concurrent vertical profiles of wind speed and temperature permitted the estimation of the friction velocity u_* and the Monin-Obukhov length L , basic independent variables of the Lagrangian similarity theory. Comparisons were then made between the measured values of the normalized, crosswind-integrated surface air concentration $u_*\chi/Q$ and theoretically predicted values. At all distances the agreement between measurements and theory was good, being poorest for strongly stable conditions. Figure 2.16 shows the measured and predicted values of $u_*\chi/Q$ at a downwind distance of 400 m as a function of atmospheric stability parameterized by $1/L$.

FIGURE 2.16. Predicted and Measured Contaminant Concentrations as a Function of Atmospheric Stability

Theoretical calculations were made assuming values of the exponent r equal to 1.0, 1.5 and 2.0 and assuming that the eddy diffusivity for the tracer was equal to either that for momentum K_m or for heat K_h . In Figure 2.16 the solid theoretical curve corresponds to $r = 1.5$ and K_h , and the broken curve corresponds to K_m . While the curves are little different for strongly stable

conditions (L negative), the superiority of K_h is quite obvious for near-neutral ($L = 0$) and unstable conditions (L positive). The predicted values of $u_* \chi / Q$ are not nearly as sensitive to the value of r , but for all distances and all stabilities a value of 1.5 appears to give the best overall match between data and theory.

A FURTHER COMPARISON OF GAUSSIAN DIFFUSION-DEPOSITION MODELS

T. W. Horst

The Gaussian diffusion-deposition models of Chamberlain and Overcamp have been compared with the exact solution of Horst. Overcamp's model is found to be a useful improvement over Chamberlain's at short distances downwind of the source. At large distances, however, Chamberlain's model is superior to Overcamp's.

Csanady's model for the deposition of particulates with an appreciable settling velocity from a Gaussian diffusion plume¹⁸ has been extended by Overcamp to the case of gases and fine particulates, where the deposition velocity does not equal the settling velocity.¹⁹ Overcamp's model selectivity reduces the strength of the conventional image source to account for the loss of material due to dry deposition. Because it results in a relative depletion of the material near the ground, his model is intended to be an improvement over Chamberlain's source depletion model,²⁰ which distributes the loss throughout the entire vertical extent of the plume by reducing the strength of both the real and image sources.

As Csanady discussed, however, his model is only an approximate solution.²¹ Horst, on the other hand, has developed a surface depletion model which has been used to calculate exact solutions for the deposition of nonsettling particles from a Gaussian plume.²² Nevertheless, the models of Chamberlain and Overcamp are still of interest because they are computationally much less complex than Horst's. Therefore, Overcamp's model predictions have been compared with Chamberlain's and Horst's.²³

For the case of gases and fine particulates with negligible settling velocities,

Overcamp's model predicts that dry deposition reduces the surface air concentration by a factor

$$\frac{1}{2} \left[1 + \alpha_0(x) \right] = \left[1 + \frac{v_d}{u} \left(\frac{h}{\sigma_z} \frac{d\sigma_z}{dx} \right)^{-1} \right]^{-1}$$

where the coefficient $\alpha_0(x)$ specifies the fractional reduction of the image source strength; the x axis is aligned in the direction of the mean wind u ; v_d is the deposition velocity; h is the effective height of pollutant emission; and σ_z is the vertical standard deviation of a nondepositing Gaussian diffusion plume. Figure 2.17 shows this quantity for $h = 100$ m, $u = 5$ m s⁻¹, $v_d = 0.01$ m s⁻¹, and Pasquill stability categories A, C and E. Predictions of Chamberlain's source depletion model and Horst's surface depletion model are also shown.

For all stabilities, both Horst's and Overcamp's models correctly predict greater reductions in the surface air concentration than Chamberlain's model. However, Overcamp's model still underestimates the air concentration reduction at short distances and greatly overestimates the reduction at large distances. For a release height of 100 m, the crossover occurs at $x \approx 10^4$ m. This distance is reduced by a factor of 5 to 10 for a release height of 10 m.

FIGURE 2.17. The Reduction of the Surface Concentration Due to Deposition as Predicted by Several Gaussian Models

Just as with Chamberlain's model, this difficulty in accurately predicting at short distances results from Overcamp's model accounting for the material loss due to deposition by reducing the strength of a source at $x = 0$. Nevertheless, at short distances

downwind of the source, Overcamp's model is an improvement over Chamberlain's model because reduction of only the image source strength results in more depletion of the material near the surface.

For large distances, however, Overcamp's model, in contrast to Horst's and Chamberlain's, predicts a sharp decrease in the surface air concentration. This prediction occurs because the model neglects the additional downward flux of material produced by the reduced air concentration at the surface. Because excessive depletion of the surface air concentration results, Overcamp's model in fact predicts that deposition ceases at large downwind distances and that the amount of undeposited material unrealistically approaches a constant value.

In summary, Overcamp's extension of Csanady's model is a useful improvement over Chamberlain's model at short downwind distances. His predictions lie midway between those of Chamberlain's model and Horst's, and his model is even simpler to apply than Chamberlain's. At large distances, however, Overcamp's model greatly overestimates the reduction of the surface air concentration and unrealistically predicts that the loss of material due to deposition has an upper limit. At these distances Chamberlain's model is superior to Overcamp's.

DOCUMENTATION OF THE HANFORD 67-SERIES ATMOSPHERIC FIELD DIFFUSION EXPERIMENTS

P. W. Nickola

A data volume documenting the Hanford 67-Series atmospheric field diffusion experiments has been published. The documented experiments were carried out on the Hanford field diffusion grid from 1967 to 1973.

During the period 1959 to 1973, more than 300 atmospheric field diffusion experiments were conducted at the Hanford area near Richland, WA. A volume published in 1977 has documented the diffusion data and concurrent meteorology for the latter third of these experiments.²⁴

Prior to the publication of this volume, some of the earlier of the 300 experiments were described in reports or journal articles. The 1959 experiments, dubbed the Green Glow diffusion program, were documented by Barad

and Fuquay.²⁵ They included detailed diffusion and meteorological data for the 27 Green Glow field tests.

In the Green Glow tests, tracers were released from an elevation of 2 to 3 m, and samples were taken in two arrays: ground-level and tower. These near ground-level tracer releases, with both horizontal and vertical sampling arrays, continued at Hanford with a series of 42 field experiments in 1960, 1961 and 1962 known as the Hanford 30-Series. Selected ground-level diffusion data

and meteorological data from both the Green Glow and 30-Series experiments were collected by Fuquay, Simpson and Hinds in a 1964 journal article.²⁶

While the 30-Series experiments were being run, another group of field experiments began at Hanford in the fall of 1960. Primarily elevated-source experiments, these tracer releases, numbering more than 200, were designed to investigate specific areas in the more general realm of diffusion. Results of these investigations have been presented in a variety of forums -- including this series of annual reports to the sponsor. However, because measurements made during these field experiments are pertinent to areas beyond the narrower objectives of the original experiments, the 1977 data volume was published for general use.

The experiments documented in the 1977 volume are a segment of tracer releases mentioned in the preceding paragraph which were carried out between July 1967 and November 1973, and have rather arbitrarily been labeled the Hanford 67-Series. With publication of the 67-Series report, there remain approximately 100 Hanford field diffusion experiments (carried out between 1960 and 1967) which have not been published in a form convenient for general research use.

The 103 tracer releases of the Hanford 67-Series were carried out during 54 different experimental periods. Multi-tracer releases (generally from two different elevations) account for the fact that the number of tracer releases is greater than the number of experimental periods. Vertical profiles of wind speed, wind direction and temperature were made during the tracer releases which generally lasted 30 min. As few as 63 and as many as 718 field locations were employed in sampling tracer concentrations during a given release. In 32 of the 103 releases, 10 or more towers were employed downwind of the source in an attempt to define vertical concentration distributions. Tower height varied from 27 m to 62 m. As few as 2 and as many as 10 sampling arcs, concentric about the release point, were used in deploying tracer samplers. The radii of these arcs varied from 0.2 km to 12.8 km from the source. Tracers were released at an elevation of 1 m, 2 m, 26 m, 56 m or 111 m.

Table 2.2, reproduced from the report documenting the Hanford 67-Series, should aid researchers in selecting field experiments which may be pertinent to their specific interest. Details of the meteorology and tracer dispersal, sampling and assay follow in the narrative and appendices of the report.

TABLE 2.2. General Grid, Source, Release and Meteorological Information

Test	Grid Source	Type(a) and Duration(b) of Tracer Release					Ubar, m/sec		Sig $\odot$, Deg		Delta T, F Deg.		Sampling Distance from Source, m		Total Towers Active
		1m	2m	26m	56m	111m	1.5m	61m	1.5m(c)	61m(d)	0.9 to 30m	15 to 61m	Nearest	Farthest	
D1	S		ZF				1.1	3.9	22	10	1.7	0.7	200	3,200	20
D2	S		ZF				1.6	6.6	10	5	6.4	2.0	200	3,200	20
D3	S		ZF				2.4	4.7	33	17	1.9	0.6	200	3,200	20
D4	S		Z16,F16				1.2	4.8	37	4	1.3	0.6	200	3,200	20
C1	S	K15					1.2	5.8	7	3	6.9	6.9	200	800	6
C2	S	K15					3.9		6		-1.6	-1.2	200	800	6
C3	S	K14					7.6		10		-3.2	-2.4	200	800	6
C4	S	K10					3.8		13		-4.6	-2.3	200	800	6
C5	S	K20	Z20				2.6	7.5	7	3	1.0		200	800	10
U56	U		Z	F			1.6	6.0	22	5	2.1	2.3	200	3,200	None
U57	U		Z	F			1.3	3.4	9	4	5.9	0.7	400	3,200	None
U58	U		Z	F			2.9	6.9	8	6	1.7	0.6	400	3,200	None
U59	U		F	Z			1.8	3.9	15	8	1.3	0.1	800	12,800	None
U60	U		F	Z			1.2	5.7	14	6	4.1	1.5	800	12,800	None
U61	U		F15	Z			5.0	10.1	6	3	0.4	0.0	800	12,800	None
U62	U		F20	Z			5.2	9.2	6	6	-0.4	-0.5	800	12,800	None
U63	U		F25	Z			2.9	7.5	6	2	1.2	0.6	400	12,800	None
U64	U		F20	Z			1.4	5.3	5	2	6.7	5.5	400	7,000	None
U65	U		F	Z			2.7	6.7	15	7	0.5	0.1	400	12,800	None
U66	U		F	Z			1.3	3.9	31	9	2.6	1.7	400	7,000	None
U67	U		F	Z			2.1	5.3	11	5	0.2	0.4	400	7,000	None
U68	U		F	Z			2.1	6.0	13	4	1.4	0.5	400	12,800	None
U69	U			Z			2.6	5.8	9	5	0.6	0.1	400	12,800	None
U70	U		F	Z			4.6	8.7	8	5	0.1	0.0	400	12,800	None
U71	U			Z	F15		1.5	4.3	15	11	1.3	0.3	400	12,800	None
U72	U			Z	F20		1.9	6.2	20	5	2.1	1.1	400	7,000	None
U73	U			Z	F20		2.7	6.7	8	4	0.9	0.7	400	12,800	None
U74	U			Z	F		1.7	6.5	10	3	2.3	1.3	400	7,000	None
U75	U			Z	F		1.3	2.5	6	8	5.4	3.9	400	12,800	None
U76	U			Z	F		3.2	7.2	9	5	0.8	0.8	400	7,000	None
U77	U			Z	F		1.6	3.7	9	4	0.3	0.0	400	12,800	None
U78	U			Z	F		3.8	7.4	6	2	-0.5	-0.3	400	7,000	None
U79	U			Z	F		1.9	6.8	7	4	2.5	2.1	400	12,800	None
U80	U			Z	F		0.9	2.9	16	7	6.7	3.4	400	3,200	None
U81	U			Z	F		2.8	7.1	14	11	1.3	1.3	400	12,800	None
U82	U			Z	F		1.1	4.5	22	5	6.3	3.1	400	12,800	None
U83	U			Z	F		1.5	6.7	7	3	6.1	5.2	400	7,000	None
U84	U				Z		0.9	5.1	11	5	2.0	1.2	400	12,800	None
U85	U				F	Z	3.0	8.3	13	6	-0.4	1.3	400	12,800	None
U86	U				F	Z	0.8	5.0	11	5	5.4	2.3	400	12,800	None
U87	U				Z26		1.0	3.6	30	17	5.4	1.7	400	12,800	1
U88	U				F16	Z	1.3	7.6	11	3	2.0	4.2	400	12,800	1
U89	U				F	Z	0.9	3.1	26	14	5.9	1.0	400	12,800	1
U90	U				F4	Z	0.9	4.3	13	6	6.6	0.5	400	12,800	1
U91	U				F	Z	0.9	3.8	12	10	2.8	1.2	400	12,800	1
U92	U				F	Z	0.9	4.8	8	4	3.4	1.1	400	12,800	1
V1	U			ZR			3.5	6.1	14	8	-3.4	-1.6	200	3,200	20
V2	U			ZRK			2.8	4.9	12	11	-1.0	-1.0	200	3,200	20
V3	U			ZR			2.5	4.3	10	8	-2.1	-1.4	200	3,200	20
V4	U			Z29,R31			1.8	2.9	11	5	-0.2	-1.2	200	3,200	17
V5	U			ZRK			1.2	7.4	14	3	4.9	3.3	200	3,200	20
V6	U			ZRK			2.9	8.8	6	4	1.7	2.1	200	3,200	20
V7	U			Z K			3.2	4.6	20	19	-4.1	-2.0	200	3,200	20
V8	U			Z10			1.8	3.4	9	4	-0.9	-0.7	200	3,200	20

(a) Z,F,R and K indicate zinc sulfide, fluorescein, rhodamine B and Krypton-85 respectively.

(b) Number following tracer type indicates duration of release in minutes. If no time is indicated, duration is 30 min.

(c) Beckman & Whitley vane.

(d) Aerovane.

TECHNICAL PROGRESS IN THE ALTERNATE FUEL CYCLE PROGRAM

W. E. Davis, W. J. Eadie and D. C. Powell

A U.S. scale assessment model developed at Pacific Northwest Laboratory (PNL) was used in an annual assessment for a simulated release from Savannah River Laboratory (SRL) for 1975. The estimated concentrations of pollutants and deposition were sent to Savannah River to be used to determine the effects of a release from the site.

With the increasing number of light water reactors, ~60 in the U.S. in 1976 and approximately 100 being built, the number of spent fuel elements from these reactors will increase. A study in progress is attempting to evaluate the environmental effects of reprocessing these spent fuel elements. Part of the study is determining the doses to man from the release of pollutants to the atmosphere during reprocessing. Pacific Northwest Laboratory's U.S. scale assessment model has been selected to describe the transport, diffusion and deposition of pollutants from a simulated release during reprocessing. Air concentration and surface deposition values calculated from this simulation will be used to estimate doses to man.

PNL's model, a variation of one developed by L. L. Wendell, et al., has been selected to describe the effects of nondepositing by-products, such as Kr^{85} , and depositing by-products, such as I^{129} , Pu and HTO .^{27,28} This model has been modified to estimate diffusion and wet removal of pollutants.

The model operates on a fixed two-dimensional advection grid, where the dimensions of each square are 2° latitude and 2° longitude for an 18 x 17 grid. The winds used for advection are the mean winds from radiosonde sites for a layer 300-1500 m above the surface. The winds, measured every 12 hr, are then interpolated to a grid using an interpolation scheme based on a distance-squared weighting function. The hourly values are calculated by linear interpolations in time. The advection of the parcels is provided by trajectories using winds bilinearly interpolated from the grid points. No allowances are made for vertical transport in the model.

Diffusion is calculated by assuming a three-dimensional Gaussian spread under vertical constraints. For transport distances less than 100 km, the sigma values were calculated using equations by Eimutis and Konicek.²⁹ Beyond 100 km, diffusion is handled as a function of time. Specifically,

$$\sigma_z = \sqrt{2K_z t}$$

$$\sigma_y = 0.5t$$

where K_z is the vertical diffusion coefficient and t is time since release.³⁰

Dry deposition is estimated by assuming source depletion according to the following:

$$\frac{dQ}{dt} = -Q v_d z^{-1}$$

where Q is the mass of pollutant over a unit area, v_d is the dry deposition velocity, and z is a length of particle diffusion.

Wet deposition is estimated by assuming source depletion and a washout ratio such that the mass change due to wet deposition is given by

$$\frac{dQ}{dt} = -\frac{Q}{L} WP$$

where Q is the mass of pollutant over a unit area, W is the washout ratio, L is the depth of the mixing layer, and P is the precipitation rate. The pollutant is assumed to be entrained into a cloud with uniform feeding from the mixing layer of depth L .

Hourly values of precipitation rates, P , were gridded for the year 1975. These values were used in the annual assessment.

Dry and wet depletion are calculated with the equation

$$\frac{dQ}{dt} = -\left(\frac{v_d}{z} + \frac{WP}{L}\right) Q$$

where Q is puff mass, v_d is deposition velocity, z is a current vertical distribution factor (length), W is a washout ratio, P is the current precipitation rate, and L is the mixed layer depth.

In the simulations, pollutants were released at a height of 62 m and at a rate of

1 Ci/sec. Estimates were made about the depth of the mixing layer by averaging monthly values of mixing depth. The results of these simulations are shown in Figures 2.18-2.22; Figures 2.18-2.20 show air concentrations (Ci/m^3), and Figures 2.21 and 2.22 the total mass deposited (Ci/m^2).

These results have been sent to SRL to be used in making dose calculations to determine the effects of a release from the Savannah River site.

FIGURE 2.18. Surface Air Concentrations for 1975 for Simulated Release from SRL of 1 cm/sec with No Deposition. Height of release was 62 m. Lines of concentrations are for Ci/m^3 .

FIGURE 2.19. Surface Air Concentrations for 1975 for Simulated Release from SRL of 1 Ci/sec with a Dry Deposition Velocity of 1 cm/sec (See Figure 2.18)

FIGURE 2.20. Surface Air Concentrations for 1975 for Simulated Release from SRL of 1 Ci/sec with a Dry Deposition Velocity of 1 cm/sec and Washout Ratio of 5×10^5 with the Use of Hourly Precipitation (See Figure 2.18)

FIGURE 2.21. Ground Deposition for 1975 from a Simulated Release from SRL of 1 Ci/sec with a Dry Deposition Velocity of 1 cm/sec. Lines of deposition are for Ci/m^2 .

FIGURE 2.22. Ground Deposition for 1975 from a Simulated Release from SRL of 1 Ci/sec with a Dry Deposition Velocity of 1 cm/sec and a Washout Ratio of 5×10^5 with the Use of Hourly Precipitation (See Figure 2.21).

COMPARISON OF WET VERSUS DRY DEPOSITION IN
SINGLE AND MULTI-LAYER U.S. SCALE ASSESSMENT MODELS

W. E. Davis

Single-layer and eight-layer U.S. scale assessment models were used to estimate the relative importance of wet removal versus dry removal of pollutants. A constant dry deposition velocity of 1 cm/sec was used. In the single-layer model, the mass deposited by dry removal was greater than with wet removal. However, when a multi-layer model was used with simulated vertical motions, the mass deposited by wet and dry removal was approximately the same.

Recently, a number of computer models have been developed to provide surface air concentration and surface deposition data for use in air pollution studies.^{31,32,33,34} To determine surface deposition, one component considered is precipitation removal. Whether hourly precipitation is as important as dry deposition in making these calculations, however, has never been established. A preliminary study has been made using simulations with a constant dry deposition velocity of 1 cm/sec and a simulated precipitation removal in both the single-layer model and a more complex eight-layer model to establish the relative importance of hourly precipitation.

In order to make a comparison of the role of wet and dry removal, several assumptions had to be made. One was that the dry removal could be adequately described by using a dry deposition velocity of 1 cm/sec. Another was that a washout ratio of 5×10^5 could be assumed during cases of rain.³⁵ A third was that the removal was in a cloud where uniform entrainment throughout the mixed layer occurred. The final assumption was that no dry deposition occurred over ocean areas, so that an unbiased comparison could be made only in areas where both wet and dry deposition were simulated.

The single-layer model, described in a report by Davis, Eadie and Powell,^(a) uses 12-hr gridded winds over a 300-1500 m layer to transport puffs. The eight-layer model, described in a report by Davis and Wendell,³⁶ is based on isentropic considerations and uses 12-hr layer averaged winds between 100-300, 300-500, 500-700, 700-1000, 1000-1500, 1500-2000, 2000-2500, and 2500-3000 m. These winds are interpolated to a grid and then are used to transport the puffs in hourly steps. The eight-layer model simulates vertical

motion by examining changes in the mean potential temperature in the transport layer. If a closer match in potential temperature occurs in a layer above the present layer, the puff is moved to the layer above. The same is true for downward transport.

The eight-layer model was run in two modes. The first looked at puff diffusion without considering vertical layering of the atmosphere; that is, the plume was allowed to spread vertically to the ground if the vertical diffusion estimation (σ_z) exceeded the puff's height above ground. The second mode simulated vertical diffusion over fronts where the air mass below the puff would be cooler and more stable. The vertical diffusion through this lower layer to the ground was calculated with a Pasquill class F stability. No dry deposition was allowed until the puff diffused through this surface layer.

The single-layer and eight-layer models with two modes were run for 15 days in April 1974. The single-layer results in Table 2.3 show that the wet removal is about one-half the dry removal. When the eight-layer mode was used in mode one, the wet removal increased to about three-fourths of the dry. When mode two was used, the wet removal exceeded the dry.

The difference between the single-layer and the multi-layer model is in the transport section. This difference can result in a closer alignment of the puff with precipitation, as Table 2.3 shows. The amount deposited in the multi-layer model between mode one and mode two changed because of the handling of vertical diffusion and dry deposition. This change decreased the dry deposition in the mode two results and increased the amount deposited by precipitation.

(a) "Technical Progress in the Alternate Fuel Cycle Program," p. 2.21.

TABLE 2.3. Comparison of Wet Versus Dry Deposition

Precip. Averaging Time	Model	Washout Ratio	Dry Deposition Velocity	% Total Mass Deposited	% Total Wet Deposited	% Total Dry Deposited
1 hr	Single- layer model	5×10^5	1 cm/sec	78	27	51
1 hr	Eight- layer model	5×10^5	1 cm/sec	74	32	42
1 hr	Eight- layer model	5×10^5	1 cm/sec	80	42	39

HEMISPHERIC POLLUTION BEHAVIOR STUDIES OF CHEMICAL AND RADIATION SUBSTANCES

J. D. Ludwick

Researchers at an air monitoring station in Quillayute, WA, measuring aerosol levels in the upper atmosphere, have determined from data collected from 1975 to 1977 that ozone concentrations peak in the spring and fall. The high ozone level in the spring probably results from ocean-trajectory air being brought in by strong off-shore flows; high levels in September and October may result from upper air being pushed down by upper-level high pressure areas and from polluted, stagnant air from the Puget Sound industrial area pushed westward by shifting flows.

Over the past few years, an air monitoring station in northwestern Washington State has been equipped to measure hemispheric levels of aerosol materials. After these aerosols are identified, the station tracks them to their source regions. Since the complex history of air parcels being sampled requires the identification of all possible components for clues to their sources, the task is complex. The modifying processes that take place in atmospheric travel with time also have to be considered. With sufficient time to establish seasonal patterns and longer-term fluctuations and to maintain an awareness of the changing anthropogenic pollutants introduced, the station can effectively evaluate the hazard to the biosphere presented by major industrial emissions.

In 1974 we reported the first observation that ozone concentrations in the lower troposphere at Quillayute were occasionally well correlated with ^7Be measurements. Since the upper level origin of this ^7Be was known, we believed that stratospheric ozone was

contributing significantly to low-level ozone concentrations. The National Air Quality Standard for this material, set at 80 ppb, provided criteria for limiting the atmospheric input of this pollutant from industrial emitters. In the clean air environment at the air monitoring station, levels of natural ozone in excess of 50 ppb were found to correlate with other high altitude products, suggesting that the standard might well be unreasonably restrictive.

Recently, we have processed and examined the data accumulated from ozone and radionuclide measurements for the period of January 1975 to January 1977. A brief comparison of this formalized data with the raw 1977 data to date shows essentially the same features.

- Ozone concentrations start the year in the 35-40 ppb range. They rise to average daily maximum levels of 50 ppb for periods of as long as a month during the April-May period.

- Observations have shown that continuous air concentrations of ozone may exceed 45 ppb for intervals of four consecutive weeks during spring. Although slight amounts of nearby air pollution occasionally contribute to the values at Quillayute, the strong on-shore flows at this time of year make ocean-trajectory air the predominate feature and source of these high ozone levels.
- Daily maximum averages of air-ozone concentrations decline to less than 30 ppb about August and rise to 40 ppb or more during September and October. The levels then reduce to values reported above at the start of the year. Two features of the air-flow patterns are suggested to account for the unusual rise in ozone during September and October. First, high ozone values are often associated with upper-level high pressure areas over Quillayute and the vicinity during that time of year. This factor and the high ^7Be values for many of these days suggest the downward transport of upper air to the station. Second, persistent stagnation of air over the Puget Sound industrial area produces pollution-derived ozone. Flow patterns, depending upon the position of the upper-level high pressure region, can occasionally carry this air westward through the Straits of Juan de Fuca to the monitoring station. These ozone concentrations are often correlated with the general airborne particulate levels measured at Quillayute rather than the radionuclide concentrations.

FIGURE 2.23. Ozone and Radionuclide Concentrations at Quillayute, WA in May 1975

- Correlations in the ozone-radionuclide data occur during other periods as well, as Figure 2.23 illustrates for May 1975. Also significant is the strong correlation between elevated ^7Be values and elevated ozone days -- one day removed from the arrival of the ^7Be . Evidently, the particulate and gaseous materials may also undergo some separation in their downward transport to the station. If it were not for this effect, a much stronger correlation of the ozone and ^7Be throughout the collected data at Quillayute would be evident.
- Throughout 1976, levels of ^{144}Ce and ^{95}Nb measured at Quillayute were the lowest observed at this site. In October, however, the effects of a medium-energy nuclear device affected the materials being measured, as Figure 2.24 shows. Very high levels of ^7Be were observed without any corresponding correlations in the ozone levels. Excursions of this magnitude in the data almost invariably indicate the presence of upper-level air. At least 80% of the ^7Be from air influenced by the nuclear device was derived from upper-level air.

As the nations of the industrial world are forced to overlook environmental concerns in developing energy sources rapidly and economically, monitoring stations like the one at Quillayute will provide the data necessary for evaluating hemispheric pollutant concentrations and will sound the alarm in the event that natural atmospheric cleanup mechanisms cannot cope with the increasing levels of anthropogenic poisons.

FIGURE 2.24. Ozone and Radionuclide Concentrations at Quillayute, WA in October 1976

SHORT-LIVED DEBRIS FROM THE CHINESE NUCLEAR TEST OF SEPTEMBER 17, 1977

C. W. Thomas

Pacific Northwest Laboratory has measured the ground-level air concentrations and deposition of short-lived radionuclides from a radioactive cloud, produced by Chinese nuclear tests on 17 September 1977, that passed over Richland, WA, on 22-23 September. A sizable percentage of the debris particles were attached to large particles. The deposition of short-lived radionuclides was also high, especially of ^{131}I .

The People's Republic of China detonated a low-yield atmospheric nuclear device on 17 September 1977, with a reported yield of approximately 15 kilotons. Trajectory analysis predicted that the leading edge of the radioactive cloud would arrive at the west coast of Washington on 21 September. However, Pacific Northwest Laboratory confirmed the first observation of radioactivity from an air sample collected at Richland, WA from 1520 hr on 22 September to 1400 hr on 23 September. As with other tropospheric detonations, this test produced significant ground-level air concentrations and depositions of short-lived radionuclides.

The atmospheric radionuclide concentrations at Richland between 22 September and 15 October 1977, are reported in Table 2.4. Most concentrations reached a maximum in the sample collected from 1400 hr on 23 September to 1140 hr on 24 September, and then decreased. The maximum ^{131}I concentration of 317 dpm/ 10^3M^3 was measured in this sample.

The particle-size distributions of the short-lived radionuclides in the debris are reported in Table 2.5. The data were obtained from the analysis of the stages of a 20 cfm Anderson impactor operated from 20 September through 25 September. With the exception of neptunium, between 20% and 40% of the radionuclides were associated with particles greater than $7\ \mu$ in diameter. These particle-size distributions are similar to ones measured soon after the Chinese test of 26 September 1976.

The physical and chemical forms of ^{131}I were measured by a special iodine sampler with three filters: 1) a particle filter, 2) a charcoal filter for removing inorganic iodine, and 3) a charcoal bed for removing organic iodides, such as methyl iodide. Seventy-six percent of the iodine was associated with particles, 13% was gaseous inorganic iodine, and 11% was gaseous organic iodine.

A fallout collector covered by a thin film of water was used to collect both wet and dry

deposition. The activities of radionuclides deposited in these collectors are reported in Table 2.6. The percentages of the activities associated with particles and the percentages present in cationic and anionic form in the water of the tray are also reported. The high measured depositions of short-lived radionuclides were comparable to the depositions measured on the East Coast of the United States following the Chinese nuclear test of September 1976.

The physical and chemical form of iodine in the water of the fallout tray is particularly interesting. Approximately 80% of the iodine was in particulate form, even though distilled water was used in the tray, and the iodine was allowed to remain in the water a few days prior to analysis. The fact that ^{140}Ba was mainly soluble is also of interest.

Iodine-131 is often the largest single contributor to the radiation exposure received by the population from fresh debris since it is transmitted along the air-grass-cow milk-human thyroid pathway. A sample of grass collected in Richland at 0800 hr on 29 September 1977, contained $1000\ \text{pCi kg}^{-1}$ of ^{131}I , one order of magnitude less than some of the concentrations measured on the East Coast following the 26 September 1976 Chinese nuclear test. An infant feeding on milk from a cow which fed exclusively on grass containing this ^{131}I concentration would receive an ^{131}I dose to the thyroid of 30 mrem. This dose is greater than the 15 mrem year limit for the ^{131}I dose to the population from a light water reactor operating under the design objectives contained in Regulatory Guide 1.109.

The chemical and physical forms of radionuclides in debris are related to the type, yield, and altitude of detonation. If we hope to increase our understanding of the chemical and physical properties of fallout radionuclides, which determine the potential dose to man, it is important to determine the correlation between the type of nuclear detonation and the properties of the radionuclides which are produced.

Table 2.4. Radionuclides in the Atmosphere at Richland, WA, Following the Chinese Nuclear Detonation of 17 September 1977

Sampling Period	(dpm/10 ³ meter ³)											
	¹⁴⁴ Ce	⁹⁹ Mo	¹⁴¹ Ce	²³⁷ U	¹³² Te	²³⁹ Np	¹³¹ I	⁷ Be	¹⁰³ Ru	¹⁴⁷ Nd	¹⁴⁰ Ba	⁹⁵ Zr
22/9 (1520)	120	359	90.5	29.6	182	610	114	279	45.8	149	143	113
23/9 (1400)												
23/9 (1400)	136	170	195	65.6	142	1102	317	327	119	193	383	146
24/9 (1140)												
24/9 (1140)	52.0	59.5	31.5	11.7	47.7	104	38.7	158	15.3	36.4	44.0	42.1
25/9 (1315)												
25/9 (1315)	43.3	37.6	21.4	7.73	25.4	85.6	25.4	108	9.16	20.3	27.8	28.2
26/9 (1325)												
26/9 (1325)	77.1	48.1	36.3	10.3	44.0	127	49.6	198	22.5	29.1	54.1	51.7
27/9 (1303)												
27/9 (1303)	125	87.7	85.2	22.7	121	180	103	305	51.5	64.5	133	95.9
28/9 (1405)												
28/9 (1405)	113	40	65.0	<1.5	49.1	103	68	295	49	34	110	162
29/9 (1400)												
29/9 (1400)	87	32	60	2.2	36	59	52	267	41	30	89	133
30/9 (1500)												
2/10 (1230)	125	6	25	<2	5.2	31	19	368	24	7.5	32	171
4/10 (0740)												
4/10 (0740)	89	1.9	13	<1	2	7.5	9	305	14	4.4	13	144
5/10 (1316)												
6/10 (1028)	32	3.6	36	<1.2	5	11	24	261	27	15	37	120
7/10 (0925)												
7/10 (0925)	67	2.4	35	<1.6	3.0	<3.4	15.9	185	24	14	37	97
9/10 (1205)												
9/10 (1205)	100	1.1	34	<1	<1.9	<3.3	14	336	32	9	34	144
11/10 (0725)												
11/10 (0725)	87	0.9	27	<1.3	<0.9	<6	10	262	26	7	30	116
13/10 (0735)												
13/10 (0735)	49	<1.0	21	<0.73	<0.7	<5	6	151	23	3	21	65
15/10 (0940)												

TABLE 2.5. Particle Size Distribution of Radionuclides in Initial Fallout from the Chinese Nuclear Test of 17 September 1977 (a)

Radionuclide	Percent in Each Size Range				
	>7 μ	7-3.3 μ	3.3-2 μ	2-1.1 μ	< 1.4 μ
⁹⁹ Mo	40.4	24.2	12.6	6.7	16.1
¹⁴¹ Ce	34.6	23.5	12.5	8.4	21.0
¹³² Te	26.9	28.7	16.7	8.9	18.9
²³⁹ Np	36.5	24.4	19.9	10.6	8.5
¹³¹ I	24.5	20.8	12.5	8.6	33.7
⁷ Be	1.5	3.8	5.2	10.1	79.7
¹⁰³ Ru	23.4	21.3	10.7	7.5	37.1
¹⁴⁷ Nd	34.2	17.0	13.8	5.4	29.7
¹⁴⁰ Ba	23.5	28.3	13.3	8.4	26.5
⁹⁵ Zr	24.3	17.2	10.4	10.6	37.6

(a) Aerosol sampling period 20 September 1977 through 25 September 1977 (10 a.m.)

TABLE 2.6. Physical/Chemical Form of Fallout From Chinese Nuclear Test of 17 September 1977 (a) (dpm/meter²)

	Particulate -- %		Cation -- %		Anion -- %		Neutral -- %		Total
¹⁴⁴ Ce	371	37.8	590	60.1	<17.7	<1.8	<3.03	<0.4	982
⁹⁹ Mo	10111	73.8	371	2.7	3203	23.4	<9.92	<0.1	13695
¹⁴¹ Ce	1941	52.3	1758	47.3	7.44	0.2	<7.21	<0.2	3714
²³⁷ U	1384	47.4	1505	51.5	< 8.29	<0.3	<24.8	<0.8	2922
¹³² Te	8294	63.9	1991	15.3	81.8	0.6	2613	20.1	12980
²³⁹ Np	11628	38.1	18690	61.3	99.2	0.3	<72.5	<0.2	30490
¹³¹ I	4830	79.7	200	3.3	1007	16.6	25.5	0.4	6063
⁷ Be	256	18.4	1049	75.3	<12.7	<0.9	75.6	5.4	1393
¹⁰³ Ru	1014	37.9	872	32.6	631	23.6	158	5.9	2675
¹⁴⁷ Nd	2908	72.4	919	22.9	147	3.7	<44.8	1.1	4019
¹⁴⁰ Ba	1805	15.7	9691	84.1	8.23	0.07	<23.4	<0.2	11528
⁹⁵ Zr	2259	75.7	574	19.2	142	4.8	< 7.51	<0.3	2983

(a) Material collected in a fallout tray from 20-9-77 (1520) to 23-9-77 (1430)

POSTULATED WEATHER MODIFICATION EFFECTS OF LARGE ENERGY RELEASES

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D. S. Renné, J. E. Hubbard and K. A. McGinnis

The direct and indirect impacts of large energy releases have been studied to place impacts postulated in the literature in a perspective consistent with existing technical information. Increases in cloudiness and precipitation could occur as far as 30 to 50 km from an energy center, but they might be difficult to identify. Net ecosystem response to large energy releases would be highly site specific, and physiological effects on humans and animals would be minimal. A framework was established for economic analysis of the impacts; however, the specific economic consequences of a given release are site specific.

All forms of thermal power production and many industrial processes release energy to the environment, which inevitably makes its way into the atmosphere. As the magnitude of individual releases increases, it is important to understand the direct impact of these releases on the atmosphere and the indirect impacts on ecosystems and economics. This study places the impacts postulated in the literature in a perspective consistent with existing technical information. In this study, the magnitudes of possible atmospheric impacts and their ecological and economic implications were estimated, assuming that atmospheric changes (in temperature, moisture, wind, etc.), which may or may not occur, do occur. The results of the study were published in February 1977.³⁷

Energy releases from energy centers (10 to 40 power plants at a single site) and individual power plant clusters (1 to 4 power plants) were considered. Energy was assumed to enter the atmosphere as increased temperature (sensible heat), moisture (latent heat), and air motion (kinetic energy). Primary impacts of these increases could be increased cloudiness and fog, and altered precipitation patterns. Potential secondary atmospheric impacts included decreased daytime and increased nighttime temperatures, early initiation of convective clouds, and increased frequency of hail, lightning and convective vortices.

The study determined that large energy releases may increase the temperature of the air by a Celsius degree or more, and that the magnitude of the increase and the volume of air affected would depend on the cooling systems used. A large energy center, for example, may produce a heat island

that is capable of influencing airflow and precipitation patterns; however, it is unlikely that a single cluster of power plants would significantly affect either. The moisture released by evaporative cooling systems could result in increased cloudiness and fog, and a large energy center might increase convective precipitation amounts by up to 30%, depending on its location and the type cooling systems used. These increases might occur as far as 30 to 50 km from the energy center. Figure 2.25 shows the potential increases in convective precipitation resulting from large energy releases, both in millimeters and as a percentage of annual rainfall. The predicted atmospheric modifications would be difficult to identify statistically because of dispersion and the natural variability of atmospheric phenomena.

Changes in atmospheric temperature, humidity and direct solar radiation may result in changes in the diversity of species, the productivity within natural ecosystems, and in agricultural productivity. In natural ecosystems, responses to atmospheric modifications tend to be offsetting. The response to an increase in temperature would be countered by the response to a decrease in direct solar radiation, etc. As a result, the net ecosystems' response to large energy releases would be highly site specific. Agricultural responses would be similar, with specific responses being governed by species, region of the country, and the form in which the energy is released. That these responses could be either positive or negative is demonstrated in Table 2.7, taken from a report on climate modification prepared for the Department of Transportation.³⁸ The estimates in the table were obtained using various models and should be considered as rough approximations. The probability that a single area would experi-

FIGURE 2.25. Increase in Total Annual Precipitation Due to Convective Precipitation Enhancement from a Large (36,000 MWe) Energy Center. Isopleths are increases in millimeters. Percentage increases are also shown.

ence both a 2°C increase in mean temperature and a 10 to 20% increase in precipitation is small.

The direct physiological effects of the postulated atmospheric changes on human and animal populations would be minimal. Humans would more probably be affected by impacts restricting their activities.

The study analyzed the economic impact of the postulated atmospheric modifications on the basis of costs and benefits. Willingness-to-pay was selected as the appropriate measure for valuing each impact. Recreation, transportation, aesthetics, agriculture, and forestry would be most affected by primary and secondary atmospheric modifications. Economic values can be placed on some of the effects, but the willingness of people to pay to gain benefits and avoid damages in many cases can only be determined through extensive surveys. It is anticipated that the specific economic consequences of a given energy release would be a function of the release site.

TABLE 2.7. Summary of Estimates of Impacts on Agricultural Yields Due to a Postulated Change in Mean Temperature of +2°C and Annual Precipitation Increase of 10 to 20%

Crop/State	Change in Yield
Corn	
Indiana	-30%.
Iowa	-11%.
Missouri	-5%.
Illinois	-20%.
Wheat	
North Dakota	-0.69 to -0.49 bushels/acre
South Dakota	-1.13 to -0.67 bushels/acre
Kansas	-0.82 to -0.16 bushels/acre
Oklahoma	-3.45 bushels/acre
Illinois	-4.10 to -5.71 bushels/acre
Indiana	-3.05 to -4.20 bushels/acre
Rice	
California	+20% of average U.S. production
Arkansas	+4% of average U.S. production
Louisiana	-5% of average U.S. production
Texas	-6% of average U.S. production
United States	+3% of average U.S. production
Soybeans	
Ohio	+1%.
Indiana	-12%.
Iowa	+9%.
Cotton	+0.54 bales/acre ^(a)
Sorghum	
Manhattan, KS	+5.8%.
Alfalfa	
Albuquerque, NM	+18% (Oct) to -7% (Jul) ^(b)
Minneapolis, MN	+18% (May) to -2% (Jul, Aug)
St. Louis, MO	+12% (May) to -6% (Jul, Aug)
Raleigh, NC	+16% (Apr) to -9% (Aug)

^(a) Interpolated for change in precipitation from 0 to +30%.

^(b) For 2.5°C temperature increase, no change in precipitation. The extreme values are given for the months during the growing season of April to October.

ATMOSPHERIC STUDIES RELATED TO A HANFORD NUCLEAR ENERGY CENTER

J. V. Ramsdell

Atmospheric studies indicate that if a Nuclear Energy Center were to be established at Hanford, the impact on fog in the Tri-Cities could be significant. The magnitude of the impact would depend on the choice of locations and cooling systems for the reactors in the center. Related studies show that the arrangement of reactor clusters can significantly affect the probability of multiple cluster involvement in the event of a Class-9 reactor accident.

Pacific Northwest Laboratory is currently evaluating the nuclear energy center concept as it might be applied at Hanford. The atmospheric studies portion of the evaluation is estimating the impact of a Hanford Nuclear Energy Center (HNEC) on the atmosphere and the probability that more than a single cluster of reactors would be affected by a Class 9 reactor accident.

A multiple-source diffusion model has been developed to evaluate the potential impact of various reactor cluster locations and cooling options on fog and humidity. The results of the initial modeling work were published in March 1977³⁹ in a report describing the computer code developed and containing a user's guide for the code. Atmospheric measurements are being made to provide better input to the model.

The potential cluster locations in the Hanford Nuclear Energy Center that were evaluated in the fog studies are shown in Figure 2.26. Cooling systems evaluated were low, mechanical draft towers; tall, assisted natural draft towers; cooling ponds; once-through; and once through with helper ponds and towers. The estimated increases in fog in the Tri-Cities are shown in Table 2.8 for several combinations of locations and cooling options. The options are described in Table 2.9. Increases of 50 in total hours of fog would be statistically significant in about 15 years, as would increases of about 20 in hours of visibility less than 1/2 mi. Thus, the data in Table 2.8 indicate that the choice of location and cooling system may have significant impact on fog in the Tri-Cities.

FIGURE 2.26. Potential HNEC Cluster Locations

TABLE 2.8. Predicted Increases in Hours of Reduced Visibility in the Tri-Cities

	All Fog	Hours of Visibility				
		<3	<1	<1/2	<1/8	<1/16
4 Reactor Cluster						
1	15	29	33	31	16	4
20 Reactor Center						
2	98	103	91	79	36	9
7	250	237	198	165	73	17
8	207	199	167	141	62	15
9	78	85	77	67	31	8
10	55	65	61	54	26	6
11	122	124	108	92	42	10
15	74	82	74	65	30	7
16	57	65	62	55	26	6
17	37	49	48	44	22	5
23	60	69	65	57	27	7
25	44	55	53	48	23	6
26	34	46	46	42	21	5
40 Reactor Center						
12	210	202	170	142	63	15
13	112	115	101	87	40	9
14	288	271	224	187	82	19
18	164	161	137	116	52	12
19	117	120	104	89	41	10
20	63	72	67	59	28	7
24	144	144	123	105	47	11
27	90	96	86	74	34	8

The basic multiple source diffusion model has been modified to include deposition and has been used to evaluate the probability that more than a single reactor in the energy center would be involved following a Class 9 reactor accident. Involvement is defined by being in the effluent plume from the accident, but may be limited by either airborne or surface concentration of contaminants.

In the study of multiple reactor involvement following a Class 9 accident, the HNEC was assumed to consist of clusters at locations 1 through 5 in Figure 2.26. The cluster at location 1 was assumed to consist of five reactors, that at location 4 was assumed to consist of three reactors, and the clusters at the remaining locations were assumed to consist of four reactors each. Table 2.10 shows the conditional probability that at least M reactors would be involved following an accident, given that an accident has occurred.

Parametric studies have been conducted to determine the sensitivity of the results to

variations in cluster patterns and spacing, and to changes in modeling assumptions. The results of this study are to be published in a future report.

The results of both studies indicate the importance of atmospheric considerations in the planning of energy centers and the importance of planning an energy center as an entity rather than allowing it to happen. If the specific numerical results indicate significant problems, additional investigation using more sophisticated methods and more complete data should be undertaken. The models used in the current studies are extremely simple and ignore many physically relevant processes, although the results are considered to be conservative.

TABLE 2.9. Site and Cooling System Combinations Simulated

Case	Site														Climatology
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	
1	a														annual
2	a	a	a	a	a										annual
3	a			a		a							a	a	annual
4	a					a		a			a			a	annual
5	a			a		a	a		a						annual
6	a	a	a	a							a				annual
7	a			b		b	b		b						annual
8	a			e		e	e		e						annual
9	a			d		d	d		d						annual
10	a	c	c	c	c										annual
11	a	c	c				e		e						annual
12	a	a	a	a	a	a	a		a	a	a				annual
13	a	c	c	c	c	c	c		c	c	c				annual
14	a	c	c	e	c	e	e		e	e	c				annual
15	a	a*	a*	a*	a*										annual
16	a	a+	a+	a+	a+										annual
17	a	a°	a°	a°	a°										annual
18	a	a*	a*	a*	a*	a*	a*		a*	a*	a*				annual
19	a	a+	a+	a+	a+	a+	a+		a+	a+	a+				annual
20	a	a°	a°	a°	a°	a°	a°		a°	a°	a°				annual
21	a	a	a	a	a										annual
22	a	a	a	a	a	a	a		a	a	a				annual
23	a	c	c				d		d						annual
24	a	c	c	d	c	d	d		d	d	c				annual
25	a	c	c	c				f							annual
26	a	c	c					f					f		annual
27	a	c	c	c	c	c		f	c		c		f		annual

Cooling system types

low tower	a
cooling pond	b
tall tower	c
helper tower	d
helper pond	e
river	f

*75% wet, °50% wet, °25% wet

TABLE 2.10. Conditional Probability that at Least M Reactors would be Involved Following an Accident in an HNEC. Involvement is defined by normalized surface concentration equal to or greater than tabled value.

Involvement (m ⁻²)	Number of Reactors (M)									
	3	4	5	7	8	9	11	12	13	16
1.00 x 10 ⁻⁷	1.000	0.850	0.250							
4.64 x 10 ⁻⁸	1.000	0.852	0.257	0.006						
2.15 x 10 ⁻⁸	1.000	0.853	0.268	0.021	0.008	0.008				
1.00 x 10 ⁻⁸	1.000	0.853	0.290	0.046	0.028	0.016				
4.64 x 10 ⁻⁹	1.000	0.865	0.313	0.069	0.035	0.024	0.011	0.011	0.011	
2.15 x 10 ⁻⁹	1.000	0.876	0.369	0.126	0.088	0.044	0.030	0.016	0.016	
1.00 x 10 ⁻⁹	1.000	0.878	0.397	0.158	0.127	0.081	0.066	0.059	0.056	0.018
4.64 x 10 ⁻¹⁰	1.000	0.881	0.409	0.172	0.138	0.098	0.085	0.079	0.073	0.023
2.15 x 10 ⁻¹⁰	1.000	0.884	0.417	0.182	0.148	0.102	0.090	0.082	0.075	0.024
1.00 x 10 ⁻¹⁰	1.000	0.884	0.421	0.188	0.155	0.108	0.095	0.086	0.078	0.025
>0	1.000	0.884	0.427	0.194	0.165	0.121	0.108	0.100	0.090	0.030

PRECIPITATION VARIATION ON THE HANFORD
NATIONAL ENVIRONMENTAL RESEARCH PARK

J. M. Thorp

Eight years of precipitation data from 26 climatological sites on the Arid Lands Ecology (ALE) Reserve have been regressed and correlated against the precipitation record of the Hanford Meteorology Station (HMS). The regression analyses show that precipitation falls on the ALE Reserve at times when none occurs at HMS. Covariability analyses indicate a precipitation regime above 300 m near the center of the Reserve to be most independent both of the HMS regime and of elevation. Below 300 m the coefficient of determination, r^2 , varies inversely with elevation across the entire Reserve. Rattlesnake Mountain is an important factor in precipitation variability, and its influence appears to extend to at least the HMS located 17 km to the north.

The ALE Reserve portion of the Hanford National Environmental Research Park (NERP) has been under microclimatological examination since 1968.⁴⁰ Its diverse topography ranging in elevation from 120 m to 1100 m MSL produces identifiable microclimates which are important factors in plant distribution.

Most of the Hanford NERP is more homogeneous topographically (120-350 m MSL), and

except for a network of wind measuring instruments, climatic descriptions are based on records from a single station, the HMS, 223 m MSL. Climatological measurements have been taken 24 hours a day⁻¹ since late 1944 at HMS. U.S. Weather Bureau cooperative observers made observations of temperature extremes and precipitation at a site 16 km ENE of the HMS from 1912-1943. Data and statistical analyses from these sites were consolidated where

practical and published by Stone, et al., in 1972.⁴²

Monthly precipitation has been measured at 26 microclimatological sites on the ALE Reserve since 1969, and these data have been compared with the HMS data to provide a regression equation for each station.⁴⁰ Except for five stations very nearly at the same elevation as the HMS, the intercepts are all positive, indicating that most of the ALE Reserve receives precipitation at times when the HMS does not.

The bioyear precipitation from October through May is most significant for growth and reproduction of the plant species found on the ALE Reserve. The distribution of average monthly bioyear precipitation for the years of record is shown in Figure 2.27. Except for 2 sites on the exposed crest of the mountain where wind effects are great, ALE Site #1 and the HMS have less bioyear precipitation than any other sites.

The correlation coefficient, r , of each ALE station compared with the HMS varies only from 0.871 to 0.951, indicating a meaningful correlation at all sites.

The coefficient of determination, r^2 , has been calculated to show the covariability between monthly precipitation at each ALE station and that at the HMS. These values of r^2 have been plotted on an elevational schematic of the ALE network (Figure 2.28) to show the degree of dependence of the precipitation regime at each ALE site on the regime at the HMS. We see that up to about 300 m dependency at most stations is generally inversely stratified with elevation. Above 300 m the dependency pattern is quite different, however, with less effect from altitude, especially on the 4 westernmost transects. The greatest independence comes on the Central transect above 300 m where $r^2 < .800$. This seems to indicate that the effect of Rattlesnake Mountain on precipitation-producing mechanisms is most significant near the center of the ALE Reserve. This influence is likely two-fold: 1) the mountain is instrumental in orographically enhancing precipitation falling on its northeastern slope, and 2) the HMS, which is situated 17 km north ("downwind") from the crest of the mountain, probably lies within a mini-rain shadow of Rattlesnake Mountain. These influences are no doubt largely responsible for the lower bioyear monthly average precipitation at ALE Site #1, which is also about 17 km from Rattlesnake Mountain.

FIGURE 2.27. Average Monthly Bioyear (Oct-May) Precipitation (CM) for the ALE Reserve and HMS (1968-1975)

FIGURE 2.28. Values of the Coefficient of Determination, r^2 , of Mean Monthly Precipitation at ALE Sites Versus HMS (1968-1975)

COOLING TOWER DRIFT: EXPERIMENT DESIGN FOR COMPREHENSIVE CASE STUDY

N. S. Laulainen

A comprehensive experimental study of drift emissions and deposition from a mechanical draft cooling tower is planned for early spring 1978. The field effort is to measure rates of mineral mass and drift water emissions and depositions downwind from a suitable power plant. Measurements of drift droplet size distributions, cooling tower temperature and velocity profiles, and ambient meteorological parameters are also to be made. From these measurements, a data base can be developed which can be used for validating models of drift deposition from cooling towers.

Drift from a cooling tower is that component of circulating water which is entrained by the airflow in small droplets and carried out beyond the tower. The drift droplets are produced mechanically within the tower, but the visible plume condensate droplets are created through cooling of the saturated tower exhaust air. The differentiation between these droplet sources is important because the drift will contain concentrations of dissolved minerals and chemicals similar, if not identical to concentrations of the circulating water. Depending

on the chemicals present in the circulating water, drift may have an adverse effect on the environment. In order to assess this impact, it is important that the amount of drift and its distribution in the air and on the ground be determined.

Each cooling tower can be expected to have a unique drift distribution, depending on the type and size of the units and the design of the drift eliminators. Since meteorological conditions also play an important role in determining drift distribution, a complex

model will be required to assess cooling tower drift for any particular tower under the varied modes of its operation and the range of meteorological conditions to which it will be subjected.

A number of models have been developed to estimate drift distributions; ten of these have been reviewed by Chen.⁴² Using a common set of input parameters, Chen finds that the maximum deposition differs among the models by two orders of magnitude with a wide range in downwind location of peak deposition.

The objective of the experimental work described here is to develop a data base which can be used for validating drift deposition models. The central step of the proposed work is the measurement of the source characteristics and the meteorological conditions responsible for transport and dispersion of the cooling tower plume containing the drift component, and the downwind deposition and air concentrations of drift. The source and transport parameters serve as inputs to the models, while the deposition pattern serves as a comparison to the model outputs. Initially, a comprehensive experimental effort is planned for early spring 1978 on the mechanical draft cooling towers of some suitable power plant site, such as the Pittsburgh (CA), Duane Arnold (Cedar Rapids, IA), Prairie Island (Red Wing, MN), Palisades (South Haven, MI) or Jack Watson (Gulfport, MS) plants.

A major deficiency in evaluating drift transport and deposition is the lack of good field data. As models have become more refined, the need for more accurate measurements of both the source and sink terms has also grown. It is necessary, then, that simultaneous measurements be made of the mineral (or other additive) concentration in the tower basin; the rates of mineral mass and drift water emission and the associated drift droplet size distribution at the tower exit; the ambient meteorological conditions to evaluate plume rise, transport and dispersion; and the spatial distribution of ground drift deposition, including both mineral mass and drift droplet size distribution. In addition, measurements of updraft wet- and dry-bulb air temperature and updraft air velocity profiles are necessary.

Since the amount of drift mineral to be measured can be expected to be small, the results of field experiments will be sensitive to the accuracy and precision of the instruments and methods used. Nearly 100% of the drift mass is presumably deposited on

the ground within a few kilometers of the tower. Most sampling techniques take advantage of the cooling water miner minerals (e.g., salt) or cooling water additives (e.g., sulfuric acid and chromium). Chemical techniques, such as ion chromatography or atomic absorption spectroscopy, can be used to obtain quantitative measurements of various chemical species (e.g., sulfate, Ca^{++} , and Na^+) from bulk samples. The use of sensitized papers, filters and/or films as collecting surfaces for individual droplets, although requiring tedious analysis, provides a method of obtaining drift droplet size distributions.

Ideally, a series of sampling stations located downwind and along the centerline of the plume should be sufficient to define the drift deposition pattern. The reality of non-ideal wind patterns and the probable lack of suitable surface locations for samplers requiring a grid, most likely, will require a system of downwind arcs with three or more sampling locations per arc. Several upwind sampler sites are required to account for ambient background levels of drift chemicals. Sampling periods longer than one hour are necessary to average plume meandering, and most important, to assure sufficient collection of drift mineral for analysis.

Since meteorological observations are extremely important for data interpretation, profile determinations of wind speed and direction and dry- and wet-bulb temperatures are required. A monostatic acoustic sounding system and a high-quality tethered-balloon telemetry system satisfy these requirements. The acoustic sounder provides qualitative data of temperature variability to several hundred meters above ground level and provides a real time display of information related to atmospheric stability and the presence of convection from the surface. The tethered-balloon system can provide qualitative data to 500 m above ground level. Time-lapse photography can also supply valuable information on complex circulations within the plume.

A site survey and pre-experiment are planned to evaluate a number of factors, including the logistics of setting up suitable surface measurement arcs with respect to surface topography and prevailing meteorological conditions, available access to fan stack exits, and the amount of drift deposition which can be expected as a function of downwind distance. With these latter measurements, the bulk samples are to be used to check instrument sensitivity.

RAINFALL ENHANCEMENT DUE TO WASHOUT OF COOLING TOWER CONDENSATE

M. Terry Dana and M. A. Wolf^(a)

Theoretical calculations of the washout of cooling tower condensate droplets by frontal raindrops show that rainfall enhancement can be significant and is measurable under typical meteorological and cooling tower effluent source conditions. For the case of moderate rainfall rates and a wind speed of 5 m/sec, centerline rainfall enhancement was as much as 46%, cross-plume average enhancement as much as 7%, and distance to one-half depletion of the source 1 to 10 km.

The environmental effect of cooling tower effluents is currently being assessed by the Meteorological Effects of Thermal Energy Releases (METER) program. An interesting cooling tower-related problem, worthy of experimental and theoretical study, is the potentiality for rainfall to be enhanced downwind due to washout of effluent vapor and condensate. This report summarizes a more detailed report⁴³ which treated the possibilities of significant rainfall enhancement from scavenging of condensate (not vapor) theoretically.

The theoretical treatment of condensate washout drawn from Dana and Hales,⁴⁴ centers on the interaction between particulates (condensate droplets) and raindrops. Since significant condensate droplet sizes are in the micrometer range, the so-called inertial collection efficiencies for raindrop-droplet washout are appropriate in this study. Slinn's semi-empirical expression was used to calculate these collection efficiencies.⁴⁵ Given the efficiencies (a function of raindrop and droplet size predominantly), calculating washout deposition involves integration of the collection efficiencies over

particle- and raindrop-size spectra. For purposes of this study, a fixed droplet radius of 5 μm was used. Available data showed Gaussian droplet-size distributions for which use of the mean droplet size is a suitable approximation.^{43,46} A typical frontal-type raindrop size distribution was used. Since both the raindrops and the condensate droplets are composed of water, the concentration of scavenged water in the raindrop -- expressed in g/cm^3 units -- is exactly equal to the fractional rainfall enhancement because the concentration of water in the raindrop is 1 g/cm^3 .

The cooling tower plume used was a bivariate normal model⁴⁷ with a virtual source point. Because the rainfall enhancement near the source is of interest, the non-vertical trajectories of raindrops are considered, since, in a wind, many downwind-depositing raindrops will not encounter the elevated plume. (This approach to plume washout has been treated elsewhere.)⁴⁸ Figure 2.29 illustrates the plume and raindrop trajectory geometry; in the figure, V_t is the terminal velocity of fall of raindrops (a function of raindrop size), u is the wind

FIGURE 2.29. Plume and Raindrop Trajectory Geometry

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speed, V_0 is the plume loft velocity (a positive constant to a downwind distance X_1 , then zero), and W is the mouth diameter of the cooling tower. The diameter of the cooling tower's mouth was set to equal to six times the standard deviation of plume spread in order to locate the virtual source point.

Table 2.11 lists the input parameters used in the sample calculations. The cooling tower is a typical natural draft tower (e.g., Trojan power plant). Three wind speeds and two rainfall rates were employed in separate calculations of rainfall enhancement as a function of true distance downwind of the tower. Even though the washout concentrations (and hence rainfall enhancement) are not a function of the rainfall rate at a given point, an increased rainfall rate will result in a depletion of the condensate plume, leading to reduced concentrations downwind; this depletion was accounted for in the calculations by a reduction in the source term using an exponential decay expression. The effect of wind speed is primarily that lower wind speeds allow more

time for washout and thus greater depletion at a given downwind distance.

Table 2.12 shows partial results of the calculations: the plume centerline maximum rainfall enhancement and distance and the maximum cross-plume average rainfall enhancement. Maximum enhancement generally occurs at the point where most of the depositing raindrops have begun to encounter the plume, which is more concentrated near the real source point.

Figure 2.30 illustrates plume depletion as a function of downwind distance at a wind speed of 5 m/sec and for rainfall rates (J) of 1 and 5 mm/hr. The depletion half distance for $J = 1$ mm/hr is about 10 km. The corresponding distances for $u = 1$ and 10 m/sec are 2.5 and 20 km, respectively.

Despite the rather idealized nature of the model used here, it is apparent that under typical frontal rain conditions rainfall enhancement and condensate plume depletion can be significant. These should be measurable through suitably designed field experiments.

TABLE 2.11. Input Parameters Used in Calculations of Rainfall Enhancement

	Item	Value	Units
Source	Liquid water source	1.7×10^5	g/sec
	Cooling tower height	150	m
	Cooling tower mouth diameter	80	m
	Loft velocity (average)	2.5	m/sec
	Plume droplet radius (mass mean)	5	μm
Meteorology	Wind speed	1,5,10	m/sec
	Rainfall rate	1,5	mm/hr

TABLE 2.12. Maximum Rainfall Enhancement Values from Sample Calculations

Wind Speed m/sec	Distance to Maximum Enhancement, km	Maximum Rainfall Enhancement, %	
		Centerline	Cross-Plume Average
5	0.4	46	7
10	0.7	17	2

FIGURE 2.30. Cumulative Removal of Condensate by Scavenging: $u = 5$ m/sec

LABORATORY SIMULATIONS OF INTERACTIVE PLUMES
FROM MECHANICAL DRAFT COOLING TOWERS

L. D. Kannberg

In connection with studies being conducted under the Meteorological Effects of Thermal Energy Releases (METER) Program, Pacific Northwest Laboratory (PNL) has been simulating discharges from physical models of mechanical draft cooling towers to determine the impact of various conditions in cooling tower plume mixing and trajectory. Analysis of the data suggests that siting cooling towers should be based on ambient wind history, plume dynamics, and tower operating conditions, and possibly on site terrain.

Under the METER Program, experimental research is being conducted into the dynamics of plumes from large thermal energy releases. Part of that program involves the physical modeling of cooling tower plumes. Researchers in the Department of Water and Land Resources at PNL have been simulating the discharges from physical models of mechanical draft cooling towers in a PNL-operated hydraulic flume. Their goal is to determine the impact of various design, operational, and meteorological conditions on cooling tower plume mixing and trajectory. They are varying several parameters in the experiment, including wind speed, Froude number (initial relative buoyancy of the plume), number of towers, spacing between towers, orientation of towers with respect to wind direction, and terrain.

The simulations are being performed in a glass-walled hydraulic flume. An array of 105 thermistors are positioned at successive stations downwind to measure the temperatures of the plume. The data collected from these instruments are converted from analog to digital signals and stored on a computer file. The entire data management system is controlled by a PDP 11/10 computer subject to the operator's direction. The computer system includes graphical routines for displaying experimental results on a CRT terminal in the laboratory. A Laser Doppler Anemometer is being installed to aid in measuring wind velocity around the cooling towers.

During 1977, researchers analyzed experimental results from mechanical draft cooling towers and compared them with results from several computer and regression models. Analysis of the experimental results led to the following conclusions:

- Minimum mixing of the plume appears to occur for $K = 1.6$ (K is the ratio of

stack effluent velocity to wind speed), but the differences are very small for varying wind speeds.

- Wake effects increase by mixing locally, particularly for $K = 3.5$.
- Increasing wind speed makes the plume bend over more rapidly; trajectories are below the stack height for plumes in very high winds (~ 50 fps).
- Increasing Froude number (i.e., decreased initial relative buoyancy) decreases mixing.
- Recirculation of the plume back into the tower increases with wind speed, Froude number, and decreased spacing between successive towers positioned perpendicular to the wind.
- A tower aligned with its major axis in line with the wind mixes less than a tower aligned perpendicular to the wind and displays a higher trajectory, evidencing greater buoyancy.
- Downwind excess temperatures for two sequential towers aligned perpendicular to the wind are only affected by spacing to a distance of about 50 stack diameters downwind, for spacings varying from 6.25 to 20 stack diameters downwind.
- The trajectories of systems with two towers in series are higher than for a single tower; this is probably due to increased buoyancy.
- The excess temperatures of systems with two towers in a series are greater than those of single towers.
- For two tower systems, the downwind side of the first tower experiences the greatest plume fluid recirculation.

Comparing the limited models led to the following conclusions:

- The Briggs⁴⁹ formulas should be used with caution for predicting mechanical draft tower plumes. Use should be limited to the "active" portion of highly buoyant discharges in light winds.
- The Winiarski and Frick⁵⁰ model offers acceptable agreement in the near field of the tower with data if the cell discharge is used in model predictions.
- The Davis⁵² model provides good excess temperature agreement to data but poor trajectory comparison.
- The Kannberg and Davis^{52,53} regression curves give predictions which are warmer in temperature and higher in trajectory than data from this study.
- None of the models tested had the capability to reproduce wake effects or successive tower plume interaction.

Analyzing the data from the experimental program and comparing its results with model predictions suggest that optimal siting of cooling towers, particularly multiple towers, requires knowledge of ambient wind history, plume dynamics, and tower operating conditions. Based on the tower wake effects and on the results for interaction of plumes from two cooling towers, site terrain may also be a significant factor in plume dynamics and interaction.

THE FIRE ANALOG: A COMPARISON BETWEEN FIRE PLUMES AND ENERGY CENTER COOLING TOWER PLUMES

M. M. Orgill

Thermal plumes or convection columns associated with large fires have been compared to thermal plumes from cooling tower systems to evaluate the fire analog concept. Energy release rate of mass fires is generally larger than that of single or small groups of cooling towers but may be comparable to proposed large energy centers. However, significant physical differences exist between cooling tower and fire plumes. Cooling tower plumes are usually dominated by ambient wind and turbulence conditions. Fire plumes, depending on fire intensity and area, can transform into free convection energy systems resulting in convective columns, strong inflow and updrafts, turbulence and concentrated vortices. Since these characteristics have not been observed with cooling tower plumes to date, the fire analog concept is questionable at this time. Additional research is needed on fire and cooling tower plumes.

Past studies on the waste heat from cooling towers and proposed energy parks have suggested that the dissipation of this energy may have significant meteorological effects. Calculations have shown that atmospheric waste energy from groupings of cooling towers dissipates at approximately the same rate as energy from geophysical phenomena such as thunderstorms, volcanoes, and large fires. Cumulus clouds and convective vortices often result from these natural energy releases.

Literature on one of these geophysical analogs -- the large fire -- has been reviewed

to determine if fire thermal plume characteristics are typical of a thermal plume from cooling towers. Though the energy release rates and energy fluxes of fires have been found to be larger than those of cooling towers and energy centers, the range is so wide that a fire could possibly be matched with some cooling tower or group of cooling towers in terms of energy released.

Though current literature describes at least nine types of fires, the most relevant information for this study dealt with only three categories: the French Meteoron, the

mass fire experiments, and some wild fires. Information from three major mass fire experiments -- 1) Project Flambeau (1964-1967);⁵⁴ 2) Tumut Exploratory Mass Fire Experiment (1968);⁵⁵ and 3) Operation Euroka (1969)⁵⁶ -- and from the French Meteotron Experiments (1961, 1971-1973)^{57,58} were summarized. Literature on the characteristics of thermal plumes from cooling towers was also reviewed to determine how they were similar and how they differed from fire plumes.

The literature described fires as transient energy phenomena that depend upon fuel and atmospheric conditions for determining their size and intensity. After reaching a critical size and intensity, a fire may transform into a mass fire -- a fire that changes its behavior from nonviolent to violent. Mass fires burn at an intense rate, and are characterized by strong in-draft and updraft winds, turbulence, convection columns, particulates and noxious gases, and the formation of clouds and vortex motions. In mass fires wind-dominated smoke plumes transform to convection columns which can predominate over the ambient wind. Ambient wind vertical profiles and stability conditions can, however, influence the development of a mass fire.

In contrast, wet cooling towers emit visible (i.e., condensed or "wet") plumes over a wide range of atmospheric conditions. The dynamics of the wet plume are closely related to those of an isolated cumulus cloud, where condensation warms the core of the plume while evaporation and cooling occur near the edges. Entrainment of ambient air affects plume dynamics significantly. Environmental relative humidity is important in controlling evaporative cooling at the plume's edges and the rate of depletion of thermal buoyancy.

The energy released from these large fires and groups of cooling towers is similar. The energy release or power of larger fires varies from 10^2 MW to 10^7 MW, with an energy flux between 10^{-3} and 1 MW m^{-2} . The power from single to groups of cooling towers varies from 10^3 MW to 5×10^4 MW, with an energy flux between 5×10^{-4} and 1 MW m^{-2} . A comparison of energy releases from fires, cooling tower systems, geophysical phenomena, and man-made activities is shown in Figure 2.31.⁵⁹ Since fires release energy at different rates over different areas, they can possibly be matched up to some cooling tower grouping. However, the similarities between the two in energy dissipation to the atmosphere do not reflect the physical differences between the cooling tower system plume and fire system plume.

The similarities between the two energy systems show that both can release considerable amounts of sensible and latent heat to the atmosphere and can cause cumulus cloud formation. Plume temperatures and moisture content for both systems are generally higher than ambient levels. Fires differ from cooling tower systems, however, in that the fire plume can transform from a forced convection system (wind dominated) to a free convection system (convective column) with resulting violent fire behavior.

These differences in physical characteristics between cooling tower and fire plumes make the fire analog concept questionable even though the approximate energy requirements appear to be satisfied in case of large energy centers. Additional field research on cooling tower and fire upper-level plume characteristics and numerical simulations of fires and multiple cooling tower plumes is recommended.

- FM - FRENCH "METEOTRON"
- MF - MINIMUM CRITERIA FOR MASS FIRE
- TF - TUMUT EXPLORATORY MASS FIRE
- PE - PROJECT EUROKA
- PF - PROJECT FLAMBEAU (FIRE 460-7)
- PR - PACK RIVER FIRE STORM (SUNDANCE FIRE)
- SF - SUNDANCE FIRE (TOTAL AREA)
- BF - BUCKHEAD FIRE
- HF - HAMBURG FIRESTORM
- EC - 40,000 MWe POWER PARK ON 1 km^2 AREA

FIGURE 2.31. A Qualitative Plot of Power Dissipated by Mass Fires, the French Meteotron Fire Experiment, Forest Fires, and Anthropogenic Activities as a Function of Their Scale Size

PLUTONIUM CONCENTRATIONS IN FOREST FIRE SMOKE PLUMES

G. A. Sehmel and M. M. Orgill

Air filter samples from four forest logging slash burns were analyzed for plutonium. Although airborne concentrations were low, one fire definitely released fallout plutonium since the measured concentration in the smoke was one order of magnitude greater than ambient background measured upwind the same day.

Smoke plumes from forest fires were previously sampled with an aircraft-mounted air sampler to determine if fallout is released by fires.^{60,61,62} Although the total smoke plume was not sampled, concentrations in the sample did indicate radionuclide release and transport. The maximum radionuclide concentration ratio measured was for ¹³⁷Cs, a level 22 times the ambient background air concentration.⁶¹ This study measured plutonium concentrations in these same samples.⁶²

Plutonium concentrations in the sampled smoke and ambient air upwind are shown in Table 2.13 for four fires. The errors shown are only the 1 σ counting errors. Ratios of plutonium concentrations in the sample compared to ambient background are also shown. For three fires, too little pluto-

nium was collected to calculate a significant ratio due to the statistical counting errors. These ratios ranged from one to two. However, the plutonium concentration for the September 6, 1975 fire was one order of magnitude greater than ambient fallout level. For this fire, we feel a greater portion of the dense smoke was sampled.

This limited study has shown that some fallout plutonium is released and transported downwind by forest fires. However, though plutonium concentrations are greater in the smoke plume than in ambient background air, the total plutonium released by a fire cannot be accurately predicted. The entire forest fire smoke plume must be sampled and characterized to determine the plutonium release flux.

TABLE 2.13. Plutonium-239 Concentrations^(a) in Smoke from Forest Logging Slash Burns

Sample Site Near	Date	Airborne ²³⁹ Pu Concentration, dpm/KSCM			Concentration Ratio	
		Background Upwind	Smoke		(Background-Corrected Smoke Sample)/(b) (Background Sample)	(Total Smoke Sample)/(c) (Background Sample)
			Background Corrected Smoke Sample	Total Smoke Sample		
Mt. St. Helens	6/9/75	0.03 $\pm$ 33%	0.55	0.45 $\pm$ 9%	20	16 $\pm$ 34%
Slate Creek, ID	8/10/76	0.02 $\pm$ 50%	0.06	0.05 $\pm$ 26%	3	2 $\pm$ 56%
Kellogg, ID	12/10/76	0.02 $\pm$ 47%	0.02	0.02 $\pm$ 62%	2	1 $\pm$ 62%
Reed Mt.	15/10/76	0.04 $\pm$ 22%	0.07	0.06 $\pm$ 27%	2	2 $\pm$ 35%

(a) Only the 1 σ counting statistics are shown, %

(b) Corrected smoke sample has background collection when outside the smoke subtracted from total smoke plume.

(c) Total smoke sample includes radioactivity collected while outside the smoke plume.

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SOLAR

- **DOE/RL - Special Studies**
- **Ground-Based Insolation Measurements at Selected Field Sites**

Aside from clouds, solar radiation is attenuated largely through scattering and absorption by atmospheric particles. The first kilometer or so of the atmosphere above the ground is generally the most turbid, whether in urban or arid regions. Thus, characterization of the turbidity of this portion of the troposphere is an important consideration in utilizing solar energy.

Turbidity measurements at Hanford have identified the diurnal, day-to-day, weekly and seasonal variations in turbidity and their resultant effects on ground-based solar radiation. In addition, these measurements have shown the spatial and topographic variations in turbidity, and the feasibility of using the remote sensing capabilities from radio and optical astronomy to study tropospheric turbidity.

TURBIDITY VARIATIONS AT HANFORD SINCE JULY 1974

N. S. Laulainen

Spectral turbidity coefficients derived from multiwavelength sunphotometer measurements obtained from July 1974 to December 1976 have been analyzed for seasonal and weekly variations. Weak biannual variations in turbidity are apparent in the data. Day-to-day variations, however, can be much larger than the coefficients for the fitted biannual terms. Consequently, it now appears that observed variations in turbidity at Hanford are related to the synoptic meteorology with a smaller, superimposed seasonality of dust and smoke sources. Turbidity variations are also independent of the day of the week.

Seasonal variations were found in turbidity data derived from multiwavelength sunphotometer observations made at Hanford during 1976.¹ Similar seasonal effects were also noted in other observations made at Hanford since July 1974.^{2,3} Since 2 1/2 annual cycles of turbidity data were available, it was decided to combine them all and test for these seasonal variations once again, as well as for possible weekly variations.

Results of the study indicate that weak biannual variations in atmospheric turbidity are apparent in this data set, which comprised 170 observation days from July 1974 to December 1976. Day-to-day variations, however, can be much larger than the magnitude of the apparent biannual term and are related primarily to meteorological episodes, with a smaller, superimposed seasonal effect of dust and smoke.

Since weighted mean regression technique may be used to search for cyclic variations present in the turbidity data, the observations were fit to this equation by a generalized least-square matrix solution, where t is time as a fraction of a period (week, year), and $\tau(t)$ is the time-dependent total optical depth:

$$\begin{aligned}\tau(t) = & a_0 + a_2 \cos(2\pi t) + a_3 \sin(2\pi t) \\ & + a_4 \cos(4\pi t) + a_5 \sin(4\pi t)\end{aligned}\quad (1)$$

This procedure also calculates a regression correlation coefficient which approaches one for a perfect fit.

All data for the period July 1974 to December 1976 (170 daily averaged optical depths) were combined for these tests. There was no correlation between weekly variation results and the day of the week. Seasonal variation results for all the data are shown in Table 3.1. Also shown in Table 3.1 are the regression coefficients derived from the data for 1975-1976 and 1976 only. As noted earlier, none of the fits were particularly good with respect to the regression correlation coefficient, though the correlation coefficient improved somewhat as the number of observations increased. The biannual coefficients (a_4, a_5) may be significant since they were 3 to 4 times larger than the uncertainties of these coefficients ($\Delta a_4, \Delta a_5$) (i.e., the coefficients vary from zero by more than 3 or 4 standard deviations).

The coefficients in Table 3.1 may be interpreted as follows:

- a_0 gives a mean yearly optical depth due to Rayleigh scattering, ozone absorption, and aerosol extinction (the standard atmosphere values of Rayleigh plus ozone extinction⁴ are also shown in Table 3.1).
- (a_2, a_3) and (a_4, a_5) give the magnitudes of the annual and biannual variations in

TABLE 3.1. Regression Coefficients of a Least Squares Fit to Eq. 1 of Optical Depth Data at Hanford

Filter	Rayleigh, Ozone Optical Depths	No. of Days	Data Set	Regression Coefficients ^(a)					Regression Correlation Coefficient
				a_0	a_2	a_3	a_4	a_5	
1 (448.2nm)	0.223	170	1974-76	0.341(6)	-0.005(9)	-0.003(8)	-0.032(8)	-0.025(9)	0.14
		141	1975-76	0.340(7)	-0.010(10)	-0.001(9)	-0.029(9)	-0.023(10)	0.11
		83	1976	0.336(8)	-0.004(11)	-0.021(12)	-0.023(11)	-0.014(12)	0.09
2 (500.0nm)	0.153	170	1974-76	0.267(6)	+0.003(9)	+0.010(8)	-0.033(7)	-0.023(8)	0.17
		141	1975-76	0.267(6)	+0.001(10)	+0.008(9)	-0.031(8)	-0.021(10)	0.14
		83	1976	0.262(8)	+0.008(10)	-0.008(11)	-0.025(11)	-0.012(11)	0.09
3 (557.8nm)	0.125	170	1974-76	0.223(7)	-0.007(11)	+0.017(9)	-0.026(9)	-0.030(10)	0.13
		141	1975-76	0.222(8)	-0.009(12)	+0.013(11)	-0.022(10)	-0.030(12)	0.10
		83	1976	0.213(7)	+0.003(10)	+0.000(11)	-0.022(10)	-0.017(11)	0.08

^(a)A shorthand convention for expressing standard deviations is used, e.g., 0.341(6) = 0.341 ± 0.006.

particle extinction, respectively, although an annual variation in ozone may be evident in the data for filters 2 and 4.

If the biannual coefficients are indeed significant, then turbidity maxima are apparent in the spring and fall seasons.

It is not unreasonable to attribute the spring and autumn cyclic maxima to a combination of anthropogenic and meteorologic causes. In the spring months the timing of maximum land tillage coincides with wind speeds in excess of 11 mps⁵ and accompanying seasonal occurrences of dust or blowing dust. During the autumn months, the stagnant conditions associated with fewer windy days and weaker frontal storms, combined with the effects of burning locally and slash burning in Cascade forests to the west, cause many smoky-hazy days. Day-to-day variations, therefore, can be much larger than the coefficients of the periodic terms (see Figure 3.1). These variations are due largely to the meteorological effects noted above and appear to be responsible for the apparent poor fit of periodic terms to the data.

Other periodic effects may also be present in the data. The stratospheric ozone column,⁶ for example, has an annual cycle which can be approximated by

$$[O_3] = 0.360 + 0.030 \sin(2\pi t) \text{ (atm-cm)} \quad (2)$$

at a latitude of 46°N. The standard atmosphere value is 0.325 atm-cm. For filters 2 and 4, the standard atmosphere contribution to the total optical depth is 0.012 and 0.035, respectively. Using these values and the annual ozone cycle given by Equation 2, the annual variations of extinction in these two colors are

$$\tau_{z,2} = 0.013 + 0.001 \sin(2\pi t)$$

and

$$\tau_{z,4} = 0.039 + 0.003 \sin(2\pi t). \quad (3)$$

These periodic effects are smaller than the coefficients (a_2 , a_3) shown in Table 3.1 and, therefore, are not likely to be responsible for the observed seasonal variation.

FIGURE 3.1. Seasonal Variations in Total Atmospheric Optical Depth at Hanford from July 1974 to December 1976. Weighted Mean Regression Curves Fit to the Data are also Shown.

DIFFERENTIAL TURBIDITY MEASUREMENTS AT HANFORD

N. S. Laulainen, J. A. Bates, E. W. Kleckner,
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An experiment to examine differential turbidity effects on measured insolation between the Rattlesnake Observatory and the Hanford Meteorological Station was conducted during summer 1977. Several types of solar radiation instruments were used, including pyranometers, multi-wavelength sunphotometers, and an active cavity radiometer. Preliminary results show dramatic temporal variability of aerosol loading at HMS and significant insolation and turbidity differences between the Observatory and HMS.

Aside from clouds, solar radiation is attenuated largely through scattering and absorption by atmospheric particles. The first kilometer or so of the atmosphere above the ground is generally the most turbid, whether in urban or arid regions. Thus, characterization of the turbidity of this portion of the troposphere is an important consideration in utilizing solar energy and evaluating potential inadvertent climate modifications due to aerosols.

The Hanford area provides a unique opportunity for such a characterization because of the nearly 1 km altitude difference between the Rattlesnake Observatory and the Hanford Meteorological Station (HMS) and because of the large variability of atmospheric dust and haze in the Central Columbia Basin region.⁷ An experiment to examine turbidity effects on measured insolation was conducted during summer 1977 with several types of solar radiation instruments, including pyranometers, multiwavelength sunphotometers, and an active cavity radiometer.

Although maintaining the calibration of a few of the instruments was difficult at times, this study demonstrated the usefulness of operating several different types of solar radiation instruments for obtaining insolation and turbidity simultaneously. This dual measuring appears to be of significant importance for the August 1977 case study because of the wide range of atmospheric turbidity which occurred during that month. This study has also indicated the need for making measurements of total diffuse sky radiation. Provisions for incorporating this measurement along with the global (direct plus diffuse) radiation measurements are underway.

Global or total insolation on a horizontal surface was measured with the four types of pyranometers listed in Table 3.2. Direct solar measurements were made with two 10-wavelength sunphotometers and a 6-wavelength active cavity radiometer (ACR). One wavelength position on each instrument included the entire solar spectrum. (The sunphotometers employed silicon photo-voltaic cells as detectors and were sensitive to radiation in the range

TABLE 3.2. Solar Radiation Instrumentation Used in Differential Turbidity Study

Instrument	Model	Location	Type of Measurement
Pyranometer	Belfort, 5-3850	P _b	Total insolation
Pyranometer	Weather Measure, R401	P _{wm}	Total insolation
Precision Pyranometer	Eppley, PSP	P _{psp}	Total insolation, reference standard
Si-Cell Pyranometer	Lambda, L1200S	P _{si}	Total insolation, 0.4 - 1.2 μ m
10-Wavelength Sunphotometer	In-house prototype	S _{10λ}	Direct solar 9 Spectral bands
Active Cavity Radiometer	Radiometrics Corp.	A _{6λ}	Direct solar 5 Spectral bands Reference standard

0.4-1.2 μm , while the ACR was sensitive over the entire spectrum, 0.3-4 μm). The pyranometers were periodically (about once per month) brought back to HMS for comparison with the Eppley PSP. The sunphotometers were operated at the same location on several occasions and were also compared with the ACR at least once. Locations of the various instruments in the Hanford area are shown in Figure 3.2.

For the purposes of this study, only data taken on cloudless days since April 1977 were used in the analysis. As can be seen from the preliminary results shown in Table 3.3, the total insolation recorded at ALE-HQ and the Observatory on 6 July was lowered by orographic cumulus clouds forming over Rattlesnake Mountain during the late morning hours. Insolation values for the other two days increased with altitude, consistent with a reduction of radiation in the lower atmosphere due to haze. Insolation received at HMS was 9% lower on 8 August and 7% lower on 15 August than that received at the Observatory. Spectral turbidity differences between the Observatory and the ERC Reserve gate at the bottom of the mountain are shown in Figure 3.3. In computing the turbidities from the total optical depth, standard atmosphere values for molecular scattering and ozone absorption extinction have been used.

No corrections for water vapor and molecular oxygen absorption in the near infrared spectrum have been made. Consequently, the relative maximum near 700 nm may be an artifact arising from the computational procedure rather than a real aerosol effect. For low turbidity values, instrumental and measurement errors also become important.

Turbidity variations at HMS during the month of August were quite pronounced, as Figure 3.4 indicates. Minimum turbidity occurred around 1 August, 10 August and 25 August. Turbidity substantially increased on 4 August because of blowing dust and because of smoke on 8 August and between 15-18 August. Observations were not possible on all days because of cloudiness, especially towards the end of the month. Much of the variability can be explained in terms of the synoptic meteorology of the region.

A prolonged heat wave in the lower Columbia Basin weather for the first 22 days of August 1977 forced maximum temperatures higher than 95°F on all but two days. During this period the surface pressure featured a nearly continuous thermal trough from the south, which occasionally took the form of a small closed low over eastern Washington. Surface pressure gradients were consistently flat for this period. At the 850 mb level (~ 5000 ft msl),

FIGURE 3.2. Map of the Hanford Area Shows the Locations and Identity of Instrumentation Used in this Study. Instrument abbreviations are identified in Table 3.2.

TABLE 3.3. Total Insolation on a Horizontal Surface and Atmospheric Turbidity at Four Hanford Project Locations on 6 July and 3 August 1977

Station	Elevation	6 July 1977			3 August 1977			15 August 1977		
		Sky	Insolation, ly	Turbidity ^(a)	Sky	Insolation, ly	Turbidity ^(a)	Sky	Insolation, ly	Turbidity ^(a)
Mounds	122	Clear	740(7)	NA	Clear	NA	NA	Clear	NA	NA
HMS	223	Clear	744(7)	0.045(4)	Clear	608(7)	0.069(5)	Clear	548(7)	0.151(6)
ALE-HQ	372	Ptly Cldy (0900-1130)	727(7)	0.052(6) ^(b)	Clear	616(7)	0.063(6) ^(b)	Clear	569(7)	NA
Rattlesnake Observatory	1090	Ptly Cldy (0900-1130)	742(7)	0.028(6)	Clear	666(7)	0.039(6)	Clear	591(7)	NA

(a) Shorthand notation for standard deviation used, e.g., 0.045(4) = 0.045 ± 0.004 , $740(7) \pm 740 \pm 7$

(b) Observations taken near ALE Reserve gate No. 106, 220 m msl

a persistent offshore ridge from 1 August to about 11 August resulted in a northerly flow over eastern Washington. From 12 August to 22 August the thermal low or trough reached to this height with resulting light winds at the 850 mb level. The surface temperatures averaged 12°F above normal during this period of deep stagnation as compared with about 6.5°F above normal for the first 11 hot days with a more shallow layer of hot air.

After a sharp cold front on 23 August, the wind flow over the Columbia Basin was mostly moderate from the southwest at 850 mb before cold fronts swept the region on the 28th and 30th. Precipitation accumulation after the 22nd set an HMS record for August and occurred on all days except the 27th and 31st; cloud cover was considerable. Surface wind flow was mostly from the west after 22 August. Strong

gusty winds occurred the evenings of the 23rd, 24th, 26th, 28th and 29th. Temperatures were below normal after 22 August.

In brief, a hot, dry air mass covered eastern Washington for the first 11 days of August becoming hotter, deeper, and quite stagnant from the 12th to 22nd. A complete change occurred on 23 August with a cold frontal passage and the remainder of the month was unseasonably cool with frequent periods of precipitation and further air mass changes on the 28th and 30th.

Considerably more data remain to be analyzed, particularly the pyranometer measurements. Further comparisons of insolation with turbidity as the type in Table 3.3 will be reported later.

Neg. 77E431-2

FIGURE 3.3. Aerosol Optical Depth (Turbidity) Versus Wavelength on Two Different Days Measured at the Rattlesnake Observatory (RSO) and at the ERC Reserver Gate #106

Neg. 77E431-1

FIGURE 3.4. Spectral Turbidity Variations During the Month of August. The 597.5 nm values are sensitive to ozone variations while the 804.3 nm values are somewhat sensitive to water vapor variations; neither of these potential variations has been accounted for in the respective turbidity values.

ATMOSPHERIC REMOTE SENSING STUDIES PERFORMED
AT BATTELLE OBSERVATORY DURING AUGUST AND SEPTEMBER 1977

R. A. Stokes, J. J. Michalsky and G. J. Schuster

Preliminary remote sensing results presented here indicate that techniques drawn from radio and optical astronomy can be profitably applied to atmospheric problems. A study of tropospheric turbidity using a precision polarimeter in conjunction with a 31-in. optical telescope demonstrated the feasibility of remote measurement of aerosol size distribution. Stratospheric ozone spectra which can be used to infer the mixing ratio and vertical distribution of the molecule were obtained with a newly-developed millimeter wavelength spectrometer.

The staff members of the Battelle Observatory have been pursuing the development of state-of-the-art remote sensing instrumentation for use on the optical and radio telescopes for several years. Two devices, currently in the latter stages of development, are:

- a high-precision visible and near-infrared wavelength polarimeter that can detect very small linear and circular polarizations in the light emitted or reflected by some source, and
- a millimeter-wavelength spectrometer that has the sensitivity and bandwidth to permit measurement of molecular rotational spectra for studies of the upper atmosphere or the interstellar medium.

Two studies were performed to test the feasibility of using these devices for atmospheric work with the optical and the radio telescopes. The theoretical basis and preliminary results for each of these studies are described in the following sections.

USE OF THE OPTICAL TELESCOPE FOR TURBIDITY STUDIES

According to a recent theoretical study by Hitzfelder, Plass and Kattawar,⁸ both linear and circular polarization studies of the sunlit sky can provide useful information concerning aerosol loading and aerosol characteristics such as the index of refraction and size distribution.

From a theoretical viewpoint these studies are best performed in the so-called principal scattering plane. This plane is defined by the local zenith, the sun's position, and the perpendicular to the horizon. Several polarimetric measurements may be made with our instrument by scanning the sky in the principal plane and relating the results directly to aerosol effects. These measurements include:

- the position of neutral polarization as a function of wavelength,
- the magnitude of the maximum linear polarization in the plane as a function of wavelength, and
- the magnitude of the circular polarization as a function of wavelength.

The position of neutral polarization depends on both the index of refraction and the size distribution of the aerosols. The circular polarization is related in a unique way to changing aerosol characteristics and is not masked by Rayleigh scattering from atmospheric molecules, the major problem associated with linear polarization experiments.

In our short study, we concentrated on the most straightforward measurement, the determination of the size of the maximum linear polarization in the principal plane and its variation with wavelength. Two modifications to the polarimeter were required for this daytime linear polarization determination. Insertion of a neutral density filter and a fixed quarter-wave plate in front of the usual optics train reduced the sky brightness by a factor of 1000 and permitted simultaneous measurement of both of the linear polarization Stokes parameters. A detailed discussion of the polarimeter design and operation can be found in a previous article by Stokes, Ekstrom, and Swedlund.⁹

Two calculator programs were used (one had to be developed) to calculate the position in the principal plane 90° from the sun where the maximum linear polarization usually occurs. The telescope uses a celestial coordinate system and local measurements are, of course, made in a local reference system. The conversion between systems and the calculation of the sun's position in the local system were handled by the programs.

Two partial days were used to gather the data. We ran an active cavity radiometer equipped with filters to measure total aerosol loading (turbidity) as we measured the polarization maximum in four filters. Only one day's data were useful as clouds partially obscured the sky on the other, with the result that correlations at this point were not possible. The polarization data are, however, consistent with light turbidity as they indicate just slightly less polarization than a calculated Rayleigh pattern.

STRATOSPHERIC AND MESOSPHERIC REMOTE SENSING USING THE BATTELLE RADIO TELESCOPE

A technique which permits routine ground-based monitoring of middle atmospheric constituents could make great impact on our understanding of the photochemical and dynamical processes occurring there. Recently, attention has been directed to catalytic chemical processes resulting from man-made pollutants which increase the rate of ozone destruction in the stratosphere. Since stratospheric ozone is responsible for the absorption of potentially harmful near-ultraviolet solar radiation, a net reduction in the abundance of this minor constituent could have disastrous consequences on the earth's surface. We have concentrated our feasibility study on the remote detection of ozone using techniques drawn from radio astronomy and laboratory microwave spectroscopy.

The fundamental observation is the detection and measurement of a spectral line due to a rotational transition of a molecular constituent. The spectral lines considered lie in the frequency range of 10 to 300 GHz. At these frequencies the line shape is sensitive to the pressure and hence the altitude where the transition occurs. The spectral line observed at ground level looking up through the entire atmosphere is thus the composite of contributions from different levels of the atmosphere. The vertical distribution of molecular abundance is recovered by mathematically inverting the line shape detected by the spectrometer. For this work a broad-beam horn antenna was used to observe the ozone rotational transition in emission before the cold background of space. A variant of the technique involves use of the sun as a broad-band source of microwaves, and the ozone transition is observed in absorption at the same frequency.

The radio receiver available has a tunable range of 124-136 GHz. A 300 MHz filter bank consisting of 30 channels was used, giving a spectral resolution of 10 MHz. The radio telescope computer was used for on-line data handling. The receiver input was switched between the sky position and a standard laboratory calibration signal at a rate of 10 Hz. The difference was taken to remove long-term gain drifts in the receiver. The receiver was found to be sensitive to the chopper wheel movement producing a spurious signal many times the order of the line strength. The sensitive parts of the receiver were shielded from the chopper wheel greatly reducing this effect. However, the spurious signal remained several orders of magnitude greater than the expected signal.

To remove the remainder of this effect, the receiver frequency was switched by hand with a cycle as follows. The local oscillator was tuned such that the line center appeared in the upper half of the filter bank channels. Data was then taken in the load switching mode already described. The local oscillator was then tuned such that the line center appeared in the lower half of the filter bank. Again, load-switched data was taken. The two samples taken for each channel were then subtracted. One line-profile appeared in the upper channels and an inverted one in the lower. Such a frequency-switched ozone spectrum is shown in Figure 3.5. The effect of stratospheric ozone absorption in the continuum microwave emission from the sun is clearly evident even in these preliminary data.

The spectrum shown was taken on 30 September 1977, at 1:10 a.m. PST. The appearance is characteristic of a frequency-switched, pressure-broadened line. Since the graph in Figure 3.5 is actually the difference of two spectra, one with the line center in the upper range of the filter bank, the other in its lower range, the up and down excursion is expected. Since the quality of this data is not sufficient for detailed profile analysis, data-taking has been suspended while the requisite improvements are made in the hardware. In particular, the switching in frequency will be controlled by the computer at a rate to be determined by the nature of the load switching instabilities. Additional shielding is being designed. The facility should shortly be capable of monitoring stratospheric ozone on a continuous basis.

FIGURE 3.5. Frequency-Switched Spectrum of the Ozone Transition at $\nu = 124.389$ GHz. Abscissa is given in 10 MHz-wide channels. Channels 5 and 30 were not in service.

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OIL SHALE

- **Air Pollution Dry Deposition**
- **Atmospheric Boundary Layer Studies**
- **MAP3S Modeling Studies**

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

PNL has initiated a modest effort in the development of a plan-of-action for studying the transport and dispersion of pollutants in complex terrain and for setting up validation tests for numerical wind models. Other activities include the development of a tracer technique to measure dry deposition of pollutants over complex terrain and a new method for calculating energy fluxes over vegetation canopies.

DUAL TRACER DEPOSITION EXPERIMENT

G. A. Sehmel

A dual tracer of depositing lithium particles and nondepositing SF_6 gas is being used to measure dry deposition removal over complex terrain. Sampling equipment is also being tested.

Though many atmospheric transport and diffusion predictive models exist, little validation data are available to test those models. One problem is that available models are based on diffusion over flat, uniform terrain even though that type of terrain is unusual in most test situations. Another is that model validation of dry deposition processes has been limited. In the past, the deposition velocity has always been coupled to an assumed σ_z to interpret or predict downwind concentrations. To address these problems, an experimental technique is being developed to measure dry deposition over wide areas of complex terrain directly.

This dual tracer technique will permit measurement of fractional removal per unit length, mass transfer resistances, or deposition velocities for forest areas, valleys, canyons, cities, etc., so that the average removal rates for various types of regions can be predicted.

In addition, the use of 0.1 to 1 μm diameter particles will permit measurement of minimum deposition removal rates for these complex terrains. For other particle diameters, increased mass transfer, caused by either Brownian diffusion or gravity settling, raises deposition removal rates above the measured minimum deposition velocity.

This measurement technique is based on a dual tracer release of depositing lithium particles and nondepositing¹ SF_6 gas. Simultaneous real-time measurements of both tracers will permit calculation of plume depletion and hence deposition between sampling sites. The lithium particle diameters will be generated between 0.1 to 1 μm , the diameter range for which deposition removal is the least and for which deposition is nearly independent of particle diameter.

Sampling equipment is being built and tested for real-time measurement of both lithium and SF_6 .

REVISED METHOD OF ENERGY FLUX CALCULATIONS
OVER VEGETATION CANOPIES

J. G. Droppo

The energy budget method for determining sensible and latent heat fluxes has been widely applied in agricultural and forest studies to predict the dry deposition of pollutants. The requirement for accurate vertical gradients and the assumption relating sensible and latent eddy diffusivities, however, limit use of this method. To avoid these limitations, a computation procedure has been developed to equate the vertical flux ratios and storage term ratios of energy terms. The revised procedure is illustrated using data from a deciduous forest.

The dry deposition of air pollutants is closely related to concurrent fluxes of sensible and latent heat to and from vegetation canopies. A review of the energy flux models resulted in the formulation of a revised method aimed at reduction of experimental error in the energy budget approach.^{2,3} If proven, this method will significantly improve experimental accuracy in agricultural and forestry studies where the best alteration method, the eddy correlation method, is logistically limited to short duration, intensive studies.

The energy budget approach in the literature computes the sensible heat flux H and the latent heat flux (evaporation energy flux) LE through a horizontal layer of thickness Δz over the canopy. The relationships derived from an energy budget equation and Fickian flux equations are:

$$H = \frac{R_n - G_\ell - G_h - G_s - G_f - B}{\left(1 + \frac{L}{C_p} \frac{K_v}{K_h} \left(\frac{\Delta q}{\Delta z}\right) \left(\frac{\Delta z}{\Delta \theta}\right)\right)} \quad (1)$$

$$LE = R_n - H - G_\ell - G_h - G_s - G_f - B \quad (2)$$

where R_n = net radiation through the layer and rate of energy storage under the layer, G_ℓ = latent heat in the air, G_h = sensible heat in the air, G_s = sensible heat flux into soil, G_f = sensible heat flux into foliage, and B = other energy sources and sinks. K_v and K_h = eddy diffusivities of moisture and heat, L = latent heat of evaporation of water, C_p = sensible heat of air at constant pressure, q = specific humidity, and θ = absolute temperature. These relationships assume horizontal uniformity of the vertical sensible and latent heat fluxes over the layer.

The experimental accuracy of these equations is limited, however, when the vertical gradients of potential temperature or specific humidities either go to zero or become

poorly defined because of their small value. Assuming the ratio of the sensible and latent heat eddy diffusivities is also a source of error.

The revised computational model is aimed at reduction of these sources of experimental error. The approach is to use the rates of change of energy storage terms with time in the flux determination. The assumption is that the ratio of vertical sensible and latent heat fluxes is equal to the ratio of the sensible and latent heat storage terms in the flux layer. That is,

$$\frac{H}{LE} = \frac{G_h}{G_\ell} \quad (3)$$

Using the definition of the terms in terms of the rate of energy change over a finite time period (Δt), Equation 1 may be expressed in terms of the storage terms:

$$H = \frac{R_n - G_\ell - G_h - G_s - G_f - B}{\left(1 + \frac{L}{C_p} \left(\frac{\Delta q}{\Delta t}\right) \left(\frac{\Delta t}{\Delta \theta}\right)\right)} \quad (4)$$

Equation 4 requires the time rates of change of temperature and humidity in place of the vertical gradients. This is expected to considerably reduce the experimental error since the changes with time are usually larger than the vertical gradients. In addition the dropping of a requirement for an estimate relating the eddy diffusivities eliminates another source of error.

The experimental accuracies of applying Equations 1 (assuming $K_v = K_h$) and 4 are compared in Figure 4.1. The accuracy of energy budget ratios at one tower based on vertical gradients is shown by the standard error bars extending from the solid dot computed values. The accuracy and computed values of the energy budget ratios based on temporal changes are plotted as three solid lines. The experimental accuracy values are

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FIGURE 4.1. Comparison of Energy Ratios for Deciduous Forest Data

based on $\pm 0.03^\circ\text{C}$ and ± 0.05 mb for the gradient computation and $\pm 0.1^\circ\text{C}$ and ± 0.1 mb for the temporal computation. The computations for the other two towers demonstrated similar trends. The average three-tower energy budget is given in Figure 4.2. The budgets demonstrate consistent and reasonable trends that agree within their experimental accuracy.

The validity of the methods may be further checked by this comparison by computing an implied ratio of the eddy diffusivities, assuming that $K_v = K_h$ is a possible source of error. Table 4.1 contains the values from two towers along with several values from the literature for comparison.

The revised method considerably reduces the computed experimental error. The computed values obtained using the standard and revised methods are within experimental accuracy of each other, though this is largely a function of the large gradient inaccuracies. A better indication of the validity of the revised method is the reasonable ratios of eddy diffusivities implied by the results.

A definitive test of this method will require comparison with direct flux measurements to determine the validity of Equation 3. If proven valid, it will provide a relatively inexpensive method of accurately monitoring and studying evaporative losses and other fluxes over uniform vegetation canopies.

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FIGURE 4.2. Three-Tower Weighted Average Energy Budget (terms explained in text)

TABLE 4.1. Comparison of Derived Eddy Diffusivity Ratios (K_v/K_h) with Other Experiments

Eddy Diffusivity Ratios	K_v/K_h	
	Day Time	Night Time
Green Glow	0.52	0.88
O'Neill	0.73	0.58
Deciduous Forest	0.88 (A) ^(a)	-
	0.73 (B) ^(a)	-

^(a)Based on an average of five hours: A indicates data from Tower A, and B indicates data from Tower B

TRANSPORT AND DISPERSION IN COMPLEX TERRAIN: RESEARCH NEEDS

M. M. Orgill

Since future energy conversion activities in the western United States, such as oil shale production, may have a damaging effect on air quality, air quality assessment methods should supply reliable information about atmospheric transport and dispersion in complex terrain. Unfortunately, current methods are inadequate. This report gives an overall view of the problem of transport and dispersion in complex terrain and reviews and evaluates the research methods and needs required for realistic solutions to the problem.

Because of federal regulations preventing the significant deterioration of air quality in the Western states, the impact of energy conversion activities--oil shale production, fossil-fuel plants, nuclear plants, uranium mining, geothermal development, and strip mining--is of particular interest. In the future, even more stringent requirements may be required because subtle environmental effects are continually being identified.

If these air quality regulations are to be complied with, current air quality assessment methods must be improved to adequately describe atmospheric transport and dispersion over the complex terrain that is typical of the western United States. Terrain-induced airflows, often avoided in theory and modeling, represent a majority of environmental situations in this area. Studies of transport, diffusion, and air pollution in complex terrain in the past, however, have focused primarily on valleys rather than other landform types.

Atmospheric motion can be affected by terrain in three ways:

- 1) through boundary-layer effects as the result of surface friction and local pressure gradients,
- 2) by the barrier or orographic-dynamic effect caused by the blocking of airflow, spatial pressure differences, and gravity oscillations, and
- 3) through thermal effects due to vertical and spatial temperature gradients.

At an actual site in complex terrain, airflow may be influenced by all three of these at any time, though the importance of a given effect will change depending on the larger-scale airflow patterns, the diurnal heating and cooling cycles, and the complexity of landforms.

These three terrain effects produce a number of unique physical phenomena. A major problem with the present dispersion and transport models is their failure to incorporate many of these phenomena completely. A few of these phenomena are listed below:

- deformation of airflow
- diurnal airflows (slope-valley winds)
- separation of airflow
- high-level convective heat sources
- gravity oscillations (lee waves, etc.)
- mechanical and thermal turbulence
- plume impaction
- valley and basin stagnation
- orographic cloud systems
- vegetation canopy effects.

However, information about these phenomena is incomplete, and additional field and model studies are needed to understand their role in transport and diffusion.

The problem of atmospheric transport and dispersion of fine particulate matter and gaseous material is most easily approached by considering the four factors that influence the transport and spread of the material from the source: 1) source characteristics, 2) atmospheric air motions, 3) lower boundary (terrain) conditions, and 4) plume depletion and chemical transformation mechanisms.

The source characteristics vary with the type of energy production operation. The initial transport and spread of a plume depends on the local atmospheric air motions, which, in turn, depend on the upstream wind direction and speed, atmospheric stability, and weather. As the plume travels further from the source, the lower boundary modifies the plume's spreading and direction due to the existing terrain-induced airflow phenomena. Depletion of the plume takes place by a number of mechanisms, such as gravitational settling, plume impaction, chemical transformation, wet and dry scavenging, electrostatic attraction, absorption and coagulation. At much longer distances, the plume eventually is deformed, distorted, and spread out by the larger-scale motions of terrain and large-scale weather systems.

Though past field and modeling studies have contributed to the understanding of some of the factors influencing transport and dispersion in complex terrain, large gaps still exist in the development of quantitative formulations to describe certain terrain-induced phenomena. Basically, four methods can be used to study transport and dispersion in complex terrain: 1) theoretical, 2) field experimental, 3) physical modeling, and 4) numerical simulation.

The theoretical approach illuminates and provides comparative information for certain aspects of complex terrain problems, but will never be able to provide total solutions because of the mathematical restraints. Field experiments with tracers, sampling equipment, balloons, remote sensing, aircraft, etc. provide valuable data on transport and dispersion phenomena, but are not conducted for long periods or only in certain weather conditions because of the high cost of field operations. Field experiments are necessary, however, because they provide data on complex terrain airflow phenomena, for model input, and for testing and upgrading models, and because they suggest and test theoretical concepts.

A laboratory approach to the transport and dispersion problem has been of value because airflow and dispersion data can be obtained over simulated three-dimensional terrain, and the method is relatively inexpensive. The major difficulty with this approach is the physical restrictions imposed by a laboratory facility, an obvious problem when modeling a turbulent Ekman boundary layer. Yet, partial similarity between field and model may be adequate for certain problems since model results have shown some similarity to field phenomena.⁴

A satisfactory numerical model has not yet been developed to describe airflow and dispersion in complex terrain, thus numerical simulation is still being refined. A workable numerical model requires the specification of the wind field over simulated terrain, and, in turn, provides input data to advection and diffusion models. Some typical models⁵ are 1) Eulerian models with a closed-form solution to the steady-state advection-diffusion equation (bivariate Gaussian plume model), 2) Lagrangian models, 3) combined Eulerian-Lagrangian methods for solving the advection-diffusion equation wherein Lagrangian markers trace the movement of the pollutant through fixed cells, 4) "box models" based on a conservation-of-mass equation for pollutants, and 5) Monte Carlo methods.

These research methods need to be used to 1) describe the scales of turbulence in complex terrain, 2) evaluate short- and long-range transport and dispersion of plumes, 3) determine physical and chemical transformation in plumes, 4) remove pollutants from plumes, 5) determine the effect of multiple point, line and area sources, 6) predict long distance plume impaction, and 7) calculate 1-, 3-, 24-hr, and annual average pollutant concentrations.

A comprehensive research program can produce a more adequate data base for dealing with terrain problems, greater understanding of atmospheric transport and dispersion in complex terrain, and greater reliability in air quality prediction. A failure to plan ahead, on the other hand, can result in inadequate assessment techniques that rely on oversimplified concepts with poor credibility and that could sacrifice environmental quality or cause the imposition of unnecessary economic penalties on the developers of energy resources. Adequate information will be important in the development of various energy resources, particularly in the Western states and will make predicting environmental problems throughout the United States more efficient.

A VERIFICATION STATISTIC FOR NUMERICAL MODELS

T. C. Kerrigan

A generalized wind estimate based on a limited number of uncertain field measurements is computed at each point in a given geographical region. A point-by-point comparison with a numerical model prediction of the wind field is then described. This comparison results in numerical assessments of the probability that the model succeeded in predicting the actual wind field and that the field measurements contain sufficient information on which to base such a comparison.

Numerical models can be used to analyze airflow over complex terrain only if numerical predictions and field measurements of the wind agree. Settling on an adequate definition of agreement has proven to be a very thorny problem, however. The purpose of this project is to construct a statistic that can serve as the basis for this definition. Since this statistic quantifies the comparison between computed and measured values of the wind, it should prove useful

in assessing the performance of numerical models in predicting field measurements.

Specifically, the statistic pertains to the following situation. A geographical region R is considered over which the flow field is of interest. A modeler devises a numerical scheme for predicting the horizontal ground-level wind field $v(r)$ for r ranging in R . The model may require information about terrain features, atmospheric stability,

the wind at the boundary or at certain interior points of R , etc. On a particular occasion, we determine this information and in addition measure the wind velocity $v(r_i)$ at several locations $r_1, \dots, r_n$ in R . The modeler then uses the information he requires to compute u , the numerical estimate for v . Our purpose is to devise a scheme for comparing

$$\{u(r)\}_R \text{ and } \{v(r_i)\}_1, \dots, n$$

in order to accept or reject the model as a predictive tool.

We first list the intrinsic features of numerical modeling and field measurements which complicate any realistic comparison scheme. Accounting for these determines the form of our verification statistic.

Guided by these features, we next construct a generalized estimate of the horizontal ground-level wind field in R . This estimate is based on a limited number of uncertain field measurements which provide a basis for testing the predictions of the numerical wind model. In fact, we represent the wind at each point r in R by a formal Gaussian probability density $p(r, w)$. The mean velocity represents a best guess for the wind and the variance matrix embodies the uncertainty in this guess originating in uncertainty in the field measurements and deficiency in the extrapolation scheme.

Since we have at best a probabilistic description of the actual wind, namely p , we have to settle for assessing the perfor-

mance of the model in probabilistic terms. We derive an explicit expression for the probability that the model comes satisfactorily close to predicting the actual wind at each r in R . This function may be averaged over R to yield a succinct measure of the model's overall success.

Unfortunately this measure by itself does not carry sufficient information to serve as the verification statistic. We evidently need a second statistic to determine whether the field measurements carry sufficient information to form the basis for a decisive verification test. We accomplish this by deriving an explicit expression for the uncertainty in the generalized wind estimate at each r in R . This function may also be averaged over R to yield a succinct measure of the power of the field measurements to form the basis for a decisive verification test.

These overall measures of the model's probability of success and the field measurements' sufficiency form a potent verification statistic. They are sufficiently simple to be easily interpreted yet sufficiently complex to account for the essential aspects of the verification problem. In fact, this construction serves as an example of a general model verification technique.

This project offers two significant products besides the verification statistic itself: a numerical wind model in the form of the generalized wind estimate and a comprehensive discussion of the intrinsic difficulties in model verification.

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