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Part 1

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1969

PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1969
TO THE
USAEC DIVISION OF BIOLOGY AND MEDICINE
VOLUME II: PHYSICAL SCIENCES
PART 1. ATMOSPHERIC SCIENCES

June 1970

AEC RESEARCH &
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BNWL-1307
Part 1
UC-48, Biology
and Medicine

PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1969
to the
USAEC Division of Biology and Medicine
VOLUME II: PHYSICAL SCIENCES
Part 1. Atmospheric Sciences

By

C. L. Simpson, Manager
and
Staff Members

of the
Atmospheric Resources Department
Environmental and Life Sciences Division

June 1970

FIRST UNRESTRICTED
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PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

Volume I of this report to the ~~USAEC~~ Division of Biology and Medicine covering work in the Biological and Ecological Sciences, the latter with contributions from the Water and Land Resources Department, is issued as BNWL-1306, Parts 1 and 2, respectively. Volume II of this report covers work in the physical sciences and is issued in three parts: Atmospheric Sciences, BNWL-1307, Part 1; Radiological Sciences, BNWL-1307, Part 2; and Instrumentation, BNWL-1307, Part 3.

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

FOREWORD

The year, 1969, was one in which awareness of environmental problems increased and intensified. With the expansion that will occur in the 1970's to meet the needs of a growing population, a continuing assessment and understanding of the effects of change on the environment is required. For the atmosphere, such an assessment requires knowledge of the turbulent nature of the air over many scales of motion and the application of this knowledge to assure responsibly planned growth. The papers in this Annual Report are succinct summaries of the research supported by the Division of Biology and Medicine expressly directed to furthering the understanding of the atmospheric sciences and their application to atomic energy.

A very important contribution in this respect is the continuing research on the turbulent properties of the air as they relate to dilution, deposition and transport of matter and to the transfer and dissipation of energy. Experiments simulating various modes of environmental plume release utilizing particulate tracers provided new quantitative data on the relation between dilution and the height of release. Continued analysis of the experiments involving the release of the noble gas tracer ^{85}Kr identified the significance of the downwind stretching of the plume and its relative importance to other axial diffusion components, providing a more realistic basis for assessing the consequences of potential reactor releases. The ^{85}Kr tracer technique proved most valuable in producing reliable and detailed diffusion data; it offers great potential for further application in evaluating the role of particle and gaseous deposition.

In other studies, the effects of large scale transport were studied. The mesoscale analysis of the Hanford area is a natural extension of the diffusion studies conducted on the

Hanford Grid and is an example of the type of analysis that is required for a regional assessment pertaining to reactor siting. At the other end of the spectrum of atmospheric motions, three-dimensional measurements of the turbulent wind velocity and temperature provided basic information regarding the manner in which heat, moisture and matter is transferred in the environment. All are essential considerations in evaluating interchanges between the air and the earth surface, finding application in problems related to cooling towers, ponds, particle deposition and resuspension.

Microphysical to mesoscale studies continue of the below- and in-cloud precipitation scavenging of pollutants from the atmosphere. Measurements made at the experimental site at Quillayute, Washington, which characterize the size and electric-charge distributions of raindrops, are of fundamental importance to the interpretation of the experimental, rain-scavenging, results. Further, in-cloud scavenging is explored through measurements of the flux to the ground of cosmogenic radionuclides. Both these experimental programs are assisted by radiosonde data and computer models of the mesoscale air motions. Some theoretical analyses of the microphysical processes involved in scavenging are described later in this report. Further work is in progress toward the development of improved tracer dispersal and detection systems.

The most concentrated effort of the year was the Atmospheric Resources and Radiological Sciences Departments' joint participation in BOMEX (Barbados Oceanographic and Meteorological Experiment) which was conducted off the island of Barbados in the West Indies. The technology and capability inherent in tracer measurement, radiochemical analysis, meteorological interpretation over synoptic to micrometeorological scales, and precipitation scavenging were integrated to investigate the transfer of radioactivity between air and ocean. Evaluation

of the large scale circulation features required measurements taken over distances of hundreds of miles and up to several thousand feet altitude. Experiments to determine fluxes at the air-sea interface required the development of a sophisticated meteorological buoy system. The design of the experiment and a description of the development is given in this report. Analysis of data is continuing.

Scientists and program leaders responsible for the results reported herein represent many disciplines--meteorology, radiochemistry, engineering, physics and ecology directing their efforts to problems of great complexity and of considerable importance. We hope that this Annual Report will make a substantial contribution toward the solution of these problems.

C. L. Simpson, Manager
Atmospheric Resources Department

Reports previously issued in this series are as follows:

HW-63824	July 1, 1959 - December 31, 1959
HW-70050	Period Ending December 31, 1960
HW-73337	Period Ending December 31, 1961
HW-77609	Period Ending December 31, 1962
HW-81746	Period Ending December 31, 1963
BNWL-36	Period Ending December 31, 1964
BNWL-235 Part 1	Period Ending December 31, 1965
BNWL-481 Part 1	Period Ending December 31, 1966
BNWL-715 Part 3	Period Ending December 31, 1967
BNWL-1051 Part 1	Period Ending December 31, 1968

TABLE OF CONTENTS

ATMOSPHERIC DIFFUSION AND TRANSPORT	1
The Effects of Source Height and Stability on Plume Centerline Exposure - P. W. Nickola.	2
Real Time Histories of Diffusing Plumes and Puffs Determined from Use of an Inert Gas Tracer System - P. W. Nickola, J. V. Ramsdell, Jr., and J. D. Ludwick.	6
Measurements of the Transport Speed and Physical Dimensions of a Diffusing Puff of a Gaseous Tracer in the Atmosphere - P. W. Nickola .	10
Short Period Atmospheric Concentrations Resulting from near Ground Level Tracer Releases over Rough Terrain - P. W. Nickola .	15
Terrain Influence on Atmospheric Diffusion - W. T. Hinds	19
Modeling Peak-to-Mean Concentration Ratios in Diffusing Plumes - J. V. Ramsdell .	21
Peak-to-Mean Concentration Ratios in Building Wakes - W. T. Hinds	25
Concentration Fluctuations in Diffusing Plumes - J. V. Ramsdell	27
Mesoscale Studies at Hanford ~ H. G. Daubek and M. A. Wolf .	29
ATMOSPHERIC BOUNDARY LAYER STUDIES .	35
Turbulence Studies in the Atmospheric Boundary Layer ~ T. W. Horst and C. E. Elderkin .	36
Aircraft Instrumentation for Boundary Layer Meteorological Studies - F. O. Gladfelder and C. E. Elderkin .	42
BOMEX STUDIES	45
The Development and Testing of a Stable Buoy and Instrument System for Studies at the Sea-Air Interface - R. K. Woodruff and A. G. Dunbar .	46
Back Trajectories from the BOMEX Array - W. E. Davis	54

Preliminary Results from Radionuclide Measurements Made During BOMEX - J. A. Young, H. G. Rieck, Jr., D. E. Robertson, and W. B. Silker	60
FALLOUT STUDIES	61
RAFLUX - A Numerical Model for Calculating the Long Wave Radiative Transfer in the Troposphere - B. C. Scott	62
PRECIPITATION SCAVENGING STUDIES	69
Phoretic and Brownian Diffusion Processes in the Below-Cloud Scavenging of Aerosol Particles by Rain - W. G. N. Slinn and J. M. Hales	70
Field Experiments in Precipitation Scavenging - M. T. Dana, M. A. Wolf, and L. A. duPlessis	77
The Role of Anisotropic Brownian Diffusion in the Precipitation Scavenging of Submicron Particles - W. G. N. Slinn	82
Olympic Peninsula Rain: Characterization of Size and Electric Charge - M. T. Dana	86
Phoretic and Brownian Diffusion Processes in the In-Cloud Scavenging of Aerosol Particles - W. G. N. Slinn and J. M. Hales	92
A Numerical Model to Describe Orographic Precipitation in the Pacific Northwest - B. C. Scott	97
Cosmogenic Radionuclide Tracers of In-Cloud Scavenging Processes - R. W. Perkins, C. W. Thomas, J. A. Young, and H. G. Rieck, Jr.	105
An Interpretation of the In-Cloud Scavenging of Particulates - W. E. Davis	107
Anisotropic Brownian Diffusion in Free-Molecule Couette Flow - W. G. N. Slinn	111
A Kinetic Theory of Diffusely Reflecting Brownian Particles - W. G. N. Slinn	115
RADIOISOTOPES AS PARTICLES AND VOLATILES.	121
Wind Tunnel for Particle Deposition Measurements - G. A. Sehmel and L. C. Schwendiman	122
Particle Deposition from Turbulent Air Flow - G. A. Sehmel and L. C. Schwendiman	126

ECOLOGICAL MICROMETEOROLOGY AND CLIMATOLOGY .	. 131
Prerecorded Climatology of the Hanford Region - W. T. Hinds and J. M. Thorp .	. 132
Climate and Phenology on the Ale Reserve - W. T. Hinds and J. M. Thorp .	. 133
Foliar Interception of Precipitation in Sagebrush Communities - W. T. Hinds and J. M. Thorp	. 134
Biometeorological Studies of Contrasting Slope Exposures - W. T. Hinds .	. 136
ATMOSPHERIC RESOURCES DEPARTMENT STAFF .	, 139
PUBLICATIONS AND PRESENTATIONS	. 143

PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1969
to the
USAEC Division of Biology and Medicine
VOLUME II: PHYSICAL SCIENCES
Part 1. Atmospheric Sciences

ATMOSPHERIC DIFFUSION AND TRANSPORT

Atmospheric diffusion and transport studies are undertaken to relate the distribution and magnitude of atmospheric pollutant concentrations to pollutant source, terrain, and meteorological parameters. The technique generally employed in these investigations involves the release of tracers into the atmosphere to simulate industrial or other pollutants. Simultaneous measurements of meteorological parameters permit development of models which describe the measured tracer concentrations as a function of the measured meteorology. The observed and modeled tracer-meteorology relationships are then used in assessing or forecasting pollutant concentrations expected from routine, or accidental contaminant releases. The variety in meteorological conditions and in types of releases require data from many experiments to assure statistical significance.

Experiments of diffusion from elevated releases in stable atmospheric conditions (a situation of great concern, yet relatively unexplored) have recently been completed and are being assayed and studied. Additional Hanford data--resulting from continuous point source releases during a wide variety of meteorological conditions--will be examined to clarify remaining questionable aspects pertinent to mean concentrations. When mean concentration predictions of reasonable accuracy can be assured, emphasis will turn to the investigation, initiated in recent years, of the fluctuations in concentration which contribute to the mean.

Future emphasis will also be given to the investigation of pollution at greater distances (on the order of a few hundred miles), its dependence on meteorological conditions which can vary widely over time and space, and to the mechanisms of deposition and resuspension of pollutants. The relevance of these diffusion, deposition, and transport studies becomes clearer as man becomes increasingly concerned with the contamination of his environment.

THE EFFECTS OF SOURCE HEIGHT AND STABILITY
ON PLUME CENTERLINE EXPOSURE

P. W. Nickola

During 1969, nine field tracer releases were made at Hanford from an elevation of 365 ft. Plume centerline exposures resulting from these releases are compared with centerline exposures from previous releases from heights between 23 and 185 ft. The results are presented in the form of graphs of exposure versus travel time and exposure versus distance from source. Releases were primarily under stable atmospheric conditions.

During the Summer and Fall 1969, a series of nine tracer releases was made from an elevation of 365 ft on the Hanford 400 ft Meteorology Tower. The intent was to add to the volume of experimental evidence on the effects of source height on downwind exposure in relatively stable atmospheres. To enhance the probability of finding a stable atmosphere, releases were scheduled during nighttime hours. During each tracer release, up to a total of 374 filters was exposed along arcs (or roads approximating arcs) concentric about the source point. The sampling arcs were located at distances of 0.4, 0.8, 1.6, 3.2, 5.0, 7.0, and 12.8 km from the source tower. A known flow rate was initiated through each of the filter samples prior to the initiation of tracer generation. Generation of the fluorescent powder tracer lasted 30 min during each experiment. Vacuum on the filters was maintained until well past the time that calculations based on surface level wind speed revealed that all tracer should have passed.

The results presented here involve the relationships between plume centerline exposure (E_{CL}) and travel time or distance from source. Exposures were measured at breathing level, where the latter is defined as an elevation of 5 ft. Travel time is defined as the time required for a given element of tracer to travel from source to sampling location.

Results of aloft tracer release field experiments were reported earlier⁽¹⁾ and were summarized in the form of a series of curves relating travel time to normalized exposure. Release heights varied from 23 ft to 185 ft, and the atmospheric thermal stability ranged from "unstable" to "very stable." These thermal stabilities were based on temperature differences between the 200-ft elevation and the 3-ft elevation, and were defined as follows:

Very Stable	$\Delta T > 7.5 \text{ }^{\circ}\text{F}$
Moderately Stable	$7.5 \geq \Delta T > 2.5$
Slightly Stable	$2.5 \geq \Delta T > 0.5$
Neutral	$0.5 \geq \Delta T > -0.5$
Unstable	$-0.5 \geq \Delta T$

The curves presented on Figure 1 for release heights (H) between 23 ft and 185 ft are those presented in the

summary mentioned. The E_{CL} values have been normalized to unit mass of emitted tracer (Q) and mean wind

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FIGURE 1. Normalized Plume Centerline Exposure at Ground Level Versus Travel Time

speed ($\bar{U}$). The wind speed used was that measured at the geometric mean between the heights of release and sampling.

The data resulting from the nine Summer-Fall 1969 releases are presented as the shaded area labelled "H = 365 ft, Neutral to Moderately Stable." Seven of the nine tests fell into the moderately stable category; the remaining tests were one "neutral" test and a "slightly stable" test. Most of these nine field experiments, for reasons not well understood, gave more erratic exposure measurements from side-by-side and from arc-to-arc samplers than had most previous field tests. Because of the relatively low quality of the experimental data, and because of the lack of variety in thermal stability, these experiments were not classified by stability. Instead, the shaded area of Figure 1 gives only the range of the 365-ft release measurements.

Although there are good theoretical grounds and experimental evidence for presenting exposure as a function of travel time,^(2,3) there are practical considerations which make it advantageous to present exposure as a function of distance. One such practical reason is that use of the travel time concept requires the use of a transport wind speed, which is effective in carrying a pollutant to a receptor. One is faced with selection of an effective transport level as well as measuring or estimating the mean wind speed at that level. In deference to these practical con-

siderations, Figure 2 presents the data for releases from 85 ft and from 365 ft in the more conventional exposure versus distance format. The shaded areas give the range of measurements for both release heights.

Additionally, the 25 field experiments carried out from an elevation of 85 ft have been categorized on the basis of Richardson Number (Ri), a measure of atmospheric stability involving both thermal stratification and wind speed shear. The atmospheric layer for which Ri was computed was the layer 3 to 50 ft. Larger Ri values indicate greater stability. The curves presented on Figure 2 for the various Ri values are mean values for the tests falling within each stability category. These curves separate logically with respect to maximum exposure, with lower maxima resulting from the more stable releases. However, it would be expected that the more stable conditions would result in higher exposure values at the greater distances. This latter aspect is not suggested by the curves.

Considering only the ranges of measurements for the 85 ft and 365 ft elevation releases, the zones appear as would be expected from theory. The upper level releases show a maximum exposure, which is smaller in magnitude, but is found at a greater distance from the source. At a distance of about 10 km, the source height effect is no longer significant.

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FIGURE 2. Normalized Plume Centerline Exposure at Ground Level Versus Distance from Source

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REAL TIME HISTORIES OF DIFFUSING PLUMES AND PUFFS DETERMINED FROM USE OF AN INERT GAS TRACER SYSTEM

P. W. Nickola, J. V. Ramsdell, Jr., and J. D. Ludwick*

A brief description is given of the Hanford inert gas tracer system employed in atmospheric diffusion studies. Some example results of computer processed field data are presented. The example concentration versus time measurements for a number of field locations result from a 20-min continuous tracer release (plume) and an instantaneous point release (puff).

The development of an inert gas tracer system at Hanford has previously been reported.⁽¹⁻³⁾ During the developmental stage of this system, thirteen releases of krypton-85 were made. Although these releases were made primarily to test the feasibility and response of the prototype system, the data generated were of such high quality and in such

unusual detail that some quantitative analysis of the data has been made.⁺⁽⁴⁻⁶⁾ In view of the paucity of such real time data (especially for puffs), the measured concentrations and associated meteorological measurements have been assembled into a data volume.⁽⁷⁾

* Radiological Sciences Department

t P. W. Nickola. "Measurements of the Transport Speed and Physical Dimensions of a Diffusing Puff of a Gaseous Tracer in the Atmosphere," see the following section.

The field grid upon which the tracer was released is shown schematically on Figure 1. A plume or puff of krypton-85 was released at the center of the sampling grid. Vertical profiles of wind and temperature were monitored on two nearby towers. In the case of plume releases, the source was a pressurized gas cylinder from which krypton was released at a controlled rate of about 1 Ci/min for periods of 10 to 20 min. The effective source height was approximately 1 m. Puffs were generated by crushing quartz ampules containing the tracer by means of a guillotine-like device. The gas in each ampule had been sealed at near

atmospheric pressure to minimize the initial volume of the "instantaneous point source." Vials were crushed at ground level.

As the plume or puff was carried downwind, it was detected by a series of 64 halogen-quenched Geiger-Muller tubes located on concentric arcs 200 and 800 m from the source point. Twenty of these detectors, 1.5 m above the ground, were spaced at 2 degree intervals on each of the sampling arcs. The remaining 24 samplers were mounted on 6 towers, 3 towers on each arc.

Information from these detectors was relayed by coaxial cables to a 4096 address memory which was programmed to accept data simultaneously from the 64 detectors for 64 time increments. A time interval selector permitted automatic advance of the stepping process at a slow or fast rate through the 64 time increments. The time increment selector permitted counting intervals of 1.2, 2.4, 4.8, or 38.4 sec. When accumulated information completely filled the memory, the memory was read out to a unit which provided a photographic reproduction of the counts accumulated in each channel. Data from the photographs were punched onto cards for electronic data processing.

The body of Figure 2 is a photograph of the printer output resulting from electronic data processing of raw Geiger tube counts. Isopleths were added by hand. The numbers plotted are counts per second generated by each 1.5 m level detector and are directly proportional to krypton

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FIGURE 1. Schematic Representation of Krypton-85 Field Diffusion Grid

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FIGURE 2. Ground-Level Distribution of Relative Concentration with Respect to Time and Direction from Source. Tracer released continuously for 20 min.

concentration. Figure 3 presents similar concentrations at 200 m resulting from a puff release. The data at the left apply to the 1.5 m level samplers. At the right is a vertical cross section observed at the tower at 114 degrees azimuth.

The field system is being altered to avoid the previous limitation of 64 time increments for data storage. Continuous concentration monitoring will be possible. Further, the in-

corporation of a computer-compatible magnetic tape system will eliminate the manual processing of data now required with the prototype system.

The described detailed concentration measurements (and future such measurements) provide opportunities to develop deeper insight into atmospheric diffusion processes and offer direct measurements for testing of existing or future atmospheric diffusion models.

HEIGHT ABOVE SURFACE

(All data from azimuth of 114 degrees)

azimuth	0.0M	1.0M	3.0M	6.0M	10.0M
-	-	-	-	-	-
-	-	-	-	0.5	2.0
-	-	0.1	-	6.3	100.0
137.7	137.7	236.0	-	399.7	220.5
130.7	290.7	619.9	-	467.6	134.9
179.0	329.2	607.4	-	399.2	769.5
263.2	354.7	833.2	-	1205.3	1138.4
250.1	433.8	897.4	-	2246.8	1147.4
427.2	564.4	707.6	-	3473.4	1128.6
408.2	604.0	526.3	-	778.6	740.3
380.7	716.3	728.8	-	807.6	1209.1
363.6	554.4	1075.2	-	12976.8	646.1
604.0	726.4	1306.2	-	1260.9	499.0
617.8	693.5	976.1	-	2014.6	433.3
666.5	704.4	636.1	-	1515.3	261.8
580.2	626.8	1312.6	-	1113.7	57.4
1006.2	1012.2	1047.0	-	490.5	67.4
786.5	653.7	907.6	-	437.4	74.8
534.5	662.3	555.5	-	567.4	36.5
479.0	439.6	461.5	-	87.4	5.3
363.6	386.5	197.4	-	49.7	5.3
385.1	375.7	222.2	-	65.9	15.3
306.3	362.4	335.1	-	64.2	1.7
299.5	220.5	77.0	-	25.9	2.6
10.1	41.5	13.8	-	1.3	1.7
83.0	60.3	11.1	-	10.1	0.1
50.7	44.2	30.9	-	16.1	0.1
44.4	30.3	5.3	-	0.1	0.5
47.4	45.3	12.1	-	1.3	-
21.2	31.1	5.7	-	0.9	-
17.8	4.9	1.7	-	0.5	-
20.1	9.5	2.0	-	-	0.5
12.1	9.5	1.5	-	-	-
5.3	4.5	-	-	-	-
4.0	3.0	0.7	-	-	-
5.1	2.2	-	-	-	0.5
5.7	1.7	-	-	-	-
4.7	1.1	0.9	-	-	-
4.0	2.4	0.9	-	0.1	0.1
2.0	0.7	2.6	-	0.1	-
3.5	0.5	0.7	-	-	-
1.7	-	-	-	-	-
0.9	-	-	-	-	0.3
-	-	-	-	-	-
2.0	1.3	1.1	-	1.3	-
1.1	0.9	-	-	-	0.1
2.4	0.9	0.5	-	-	-
0.7	0.5	-	-	-	-
0.9	-	0.1	-	-	-
0.1	0.9	-	-	-	-

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MEASUREMENTS OF THE TRANSPORT SPEED AND PHYSICAL DIMENSIONS OF A DIFFUSING PUFF OF A GASEOUS TRACER IN THE ATMOSPHERE

P. W. Nickola

Measurements of puff dimensions and transport speed were obtained from five inert gas tracer releases. The speed of transport was found to increase with distance from the source, and the speed was found (in all but one case) to correspond to wind speeds measured at heights between tracer release and sampling heights.

Puff dimensions were greater in the downwind direction (length) than in the crosswind direction (width) in all cases. In unstable atmospheres, the ratio of puff length to puff width decreased as the puff moved from 200 to 800 m from the source. However, in more stable atmospheres, the ratio of puff length to width increased during the 200 to 800 m transport period.

The real-time nature of data generated during field testing of the prototype of the Hanford inert gaseous tracer system⁽¹⁾ affords an opportunity to measure diffusion parameters which are generally not easily obtained. The ability to generate

instantaneous puffs, combined with the real-time capability, permits an unusually detailed examination of diffusion from an instantaneous point source. The parameters herein examined are standard deviations of krypton distribution which resulted

from a puff release and the effective height and wind speed with which the diffusing puff moved downwind.

During the testing of the prototype krypton system, a total of eight puffs was released. The release technique and the field grid are described elsewhere.* Of the eight puffs released, five moved within the limited crosswind extent of the field grid. A portion of the concentration data generated during one such puff release is given in Figure 3 of the immediately preceding section. The left half of that figure shows relative concentration of krypton for the 200-m arc at an elevation of 1.5 m above the surface.

Summation by columns in the figure provides the input for determining the standard deviation of the distribution of krypton dosage in a crosswind direction. This crosswind distribution for the puff (test P8) is given in Figure 1. Summation by the rows of data (i.e., horizontally) provides a distribution for determining the mean time of passage T , and the standard deviation. This distribution is given in Figure 2. If T and its associated standard deviation are multiplied by an appropriate wind speed, these time parameters convert to length parameters and the resulting standard

deviation, σ_{xI} , is a measure of the length of the puff in a direction parallel to the mean wind direction.

The problem in this computation is to choose a wind speed which is representative of the puff movement to the specific distances (200 and 800 m) and height (1.5 m) where the measurements took place. After resisting the first inclination to use the 1.5-m wind as appropriate for computing σ_{xI} values, the speed employed was x/T , where x is the distance of the sampling location from the source. This speed and the associated height

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FIGURE 1. Crosswind Distribution of Exposure (concentration Time Integral) at an Elevation of 1.5 m During Puff Release P8

* P. W. Nickola, J. V. Ramsdell, Jr., and J. D. Ludwick. "Real-Time Histories of Diffusing Plumes and Puffs Determined from Use of an Inert Gas Tracer System," see the preceding section.

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FIGURE 2. Crosswind Sum of Relative Concentration Versus Time at an Elevation of 1.5 m During Puff Release P8

of measurement are termed the "effective transport speed," u_e , and "effective transport height," z_e , respectively.

Wind profiles obtained from an instrumented 25-m tower near the source provide the means for determining the effective height value. Anemometers were mounted at elevations of 0.8, 1.5, 3.0, 6.1, 12.2, and 24.4 m.

Values of u_e and z_e are given in Table 1. At 200 m, the z_e values are between the release height ($z = 0$ m) and sampling height ($z = 1.5$ m) in every instance. At 800 m, however, the effective transport

height is at 1.7 m during the stable atmosphere of test P8. Note also that the test P3 release into a thermally neutral atmosphere resulted in an effective transport height of 1.4 m--nearly the same as the sampling height of 1.5 m. The z_e values for the thermally unstable tests average 1.0 m. These results suggest that z_e increases with increasing stability.

A word concerning the z_e values of 0.5 m for the 200-m arc is in order. The fact that all the extrapolations result in the same value (0.5 m) is no doubt coincidental, but it is not the result of large

TABLE 1. Transport and Diffusion Parameters Determined from ^{85}Kr Puff Releases

Test No.	Qualitative Thermal Stability	Wind Speed at 1.5 m Elevation, m/sec	200 m from Source						800 m from Source				
			u_e , m/sec	z_e , m	σ_{xI} , m	σ_{yI} , m	σ_{zI} , m	$\frac{\sigma_{xI}}{\sigma_{yI}}$	u_e , m/sec	z_e , m	σ_{xI} , m	σ_{yI} , m	$\frac{\sigma_{xI}}{\sigma_{yI}}$
P3	Neutral	4.2	2.5	0.5*	33	12.6	-	2.6	4.1	1.4	123	28	4.4
P5	Unstable	8.0	5.1	0.5*	40	9.0	5.6	4.4	7.1	1.0	95	50	1.9
P6	Unstable	7.3	4.7	0.5*	39	11.0	-	3.5	6.6	1.1	76	44	1.7
P7	Unstable	4.6	3.0	0.5*	32	9.9	5.9	3.2	4.0	0.9	75	54	1.4
P8	Stable	1.5	1.1	0.9	47	12.1	-	3.9	1.6	1.7	154	37	4.2

* Extrapolation

smoothing procedures. The extrapolation of the measured wind profiles from the lowest anemometer level, 0.8 m, to the determined 0.5 m is not a very subjective procedure.

An examination of the columns headed σ_{xI}/σ_{yI} shows that the puffs are all elongated in the x direction. However, for the three unstable tests, comparison of the σ_{xI}/σ_{yI} ratios for 200 and 800 m reveals that, between these two distances, the puffs were becoming more symmetrical in shape. For the neutral and stable tests, the puffs were continuing to elongate in a downwind direction. The data suggest that for ground level releases: 1) Regardless of stability, diffusion in a downwind direction is more effective than crosswind diffusion for the first moments of a puff's existence; 2) In stable atmospheres, the downwind elongation continues; and 3) In unstable atmospheres, the downwind and cross wind growth rates approach each other as diffusion continues.

At 200 m, puffs traversed the field grid in such a manner that the 1.5-m elevation puff centerlines were at 112 and 114 degrees, respectively, during tests P5 and P7. Since vertical samples were taken at 114 degrees, these puffs provided an opportunity to compute σ_{zI} values. The summed counts observed during these tests are shown in Figure 3. Admittedly, the lack of adjacent tower samples, the assumption of perfect reflection from the surface, and the modest extrapolation of dosage values beyond the top of the uppermost samples leave the computed σ_{zI} values on somewhat shaky ground, but they are presented in Table 1. The vertical dimensions of the puff are smaller than either the crosswind or the downwind dimensions.

Another method of checking the x dimension of the plume is possible. Since the krypton concentration data were collected in real time, it is possible to specify that the x dimension of a puff was at least

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FIGURE 3. Vertical Distribution of Exposure (Concentration Time Integral) at 200 m Distance and 114° Azimuth During Puff Release P5 and P7.

600 m if the puff was simultaneously monitored on both the 200 and 800-m sampling arcs. Of the five puff releases listed in Table 1, only puff P7 failed to stretch the 600 m between the two sampling arcs. During test P8, the puff was monitored simultaneously on both arcs for more than 2 min. On one of the less successful puff releases (from the standpoint of horizontal containment), a puff release into a very stable

atmosphere with a 1.3 mps wind speed resulted in more than 8 min of simultaneous detection at 200 and 800 m.

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SHORT PERIOD ATMOSPHERIC CONCENTRATIONS RESULTING FROM NEAR GROUND
LEVEL TRACER RELEASES OVER ROUGH TERRAIN

P. W. Nickola

Real-time measurements were made of atmospheric concentration of a tracer released over a marine terrace in southern California. Tracer was released continuously over periods of thirty minutes. The maximum short period (5 sec) concentration observed near plume centerline was as much as 14 times the value of the 30-min mean concentration observed at the plume centerline. The maximum short-period-to-mean-centerline concentration ratio was found to decrease with increasing distance from the centerline. Distributions of short period concentrations contributing to mean concentration are given for several different locations within a diffusing plume.

Air concentrations of a pollutant to which an individual may safely be exposed are dependent upon the duration of his exposure to the pollutant. Therefore, it is pertinent to investigate not only the average concentration of a pollutant over a relatively long period of time, but also the distribution of shorter term concentrations within the long period. A Hanford-developed device⁽¹⁾ capable of monitoring atmospheric concentrations of zinc sulfide tracer on a real-time scale was employed during a series of field experiments conducted along the extreme southern coast line of Vandenberg Air Force Base in southern California.⁽²⁾ Use of the sampling device permitted some investigation into the character and magnitude of short-period concentrations. The selected short period, 5 sec, approximates the breathing period of man.

On five occasions, a real-time sampler (RTS) was operated at loca-

tions within the area swept by a plume resulting from a 30-min tracer release. The tracer release height was about 2 m, and the sampling height was approximately the same. Terrain was rather rugged (contours from sea level to 400 ft) on the marine terrace over which these experiments were carried out.

Table 1 lists information pertinent to the tracer releases and the associated RTS measurements, and defines symbols used in the table and elsewhere in this report. The atmosphere was thermally unstable during all releases. However, a variety of wind speed and wind direction standard deviations were in evidence. Column E is a ratio of the mean tracer concentration at the RTS location compared to the mean at the plume centerline. (Exposure of a series of filter samplers permitted determination of the mean plume centerline.) The releases have been ordered with respect to the magnitude of column E. Note that the

TABLE 1. Location, Meteorology, and Concentration Data

Tracer Release	A X , km	B σ_{θ} , degrees	C ΔT , C°	D $\bar{u}$, m/sec	E $\bar{x}_{RTS}/\bar{x}_{CL}$	F $x_{p5}/\bar{x}_{CL}$
1	0.8	23.7	-0.6	1.6	0.20	1.5
2	2.1	4.0	-1.0	8.6	0.54	3.3
3	2.1	3.6	-0.4	4.2	0.55	4.5
4	1.9	10.0	-3.6	3.1	0.85	7.0
5	1.0	9.5	-1.0	4.9	0.95	14

X is distance of sampler from source.

σ_{θ} is wind direction standard deviation.

ΔT is temperature at height of 54 ft
minus temperature at 6 ft

$\bar{u}$ is mean wind speed

$\bar{x}_{RTS}$ is 30-min mean tracer concentration at RTS
location

$\bar{x}_{CL}$ is 30-min mean tracer concentration at
plume centerline

x_5 is 5-sec mean tracer concentration

x_{p5} is maximum 5-sec mean tracer concentration

ratio $x_{p5}/\bar{x}_{CL}$, as given in column F, also falls into order.

All else being equal, the column F ordering could be expected to order with column E proximity to the plume centerline. Since the mean plume centerline is the location where the instantaneous plume would be expected to appear most frequently, it is also the point where there is the highest probability of observing high x_{p5} values.

Figure 1 gives the histories of concentration observed during releases 4 and 5, the tests during which the RTS was operated closest to the plume centerline. The mean 5-sec concentrations are plotted as points

and these points are joined for graphical clarity. Despite the common near-centerline exposure, there are considerable differences in the character of the distributions. During test 4, the plume was detectable at the RTS location for about 21 consecutive minutes, or 70% of the time. During test 5, the plume was in evidence only 43% of the time and on a much more sporadic basis. The graphs point out that even near the mean plume centerline, the tracer concentration is far from a constant value.

Although the 5-sec concentration of greatest interest is the peak value, x_{p5} , it is also of interest to know the distribution of 5-sec dura-

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FIGURE 1. Concentration Versus Time for Releases 4 and 5.

tion concentrations, x_5 , of lesser magnitude. Figures 2 and 3 give cumulative frequency distributions of $x_5/\bar{x}_{CL}$ for the five tracer releases. It cannot be stated that these distributions are "typical," but they are at least rough guides for locations near to and well removed from the plume centerline.

Some example reading from these graphs may be in order. Consider the curve for test 5 in Figure 3. What percentage of the x_5 values was equal to or greater than the centerline mean contribution? To answer, read the ordinate of 24% above the value $x_5/\bar{x}_{CL}$ of 1.0. What percentage was at least five times the mean? Above $x_5/\bar{x}_{CL} = 5.0$, the answer is read as 4.4%.

The discontinuous appearance of the x_{p5} values plotted as circles on Figures 1, 2, and 3 deserves comment. The curves in Figure 1 and the data for the other two figures resulted from the computing of x_5 values for a series of end-to-end, 5-sec time increments from strip charts associated with the RTS sensors. The selection of the first increment, and hence all succeeding increments, was dictated merely by a timing division on the strip chart. It would have been quite unlikely to find that the x_{p5} value for any 5-sec time increment would have coincided with one of the specified end-to-end increments. The plotted x_{p5} values on all three figures result from use of a sliding time in order to assure the maximum value.

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FIGURE 2. Percent of Time 5-sec-to-Centerline-Mean Concentration Ratio Equals or Exceeds the Indicated Ratio. (Curves are for locations well removed from plume centerline.)

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FIGURE 3. Percent of Time 5-sec-to-Centerline-Mean Concentration Ratio Equals or Exceeds the Indicated Ratio. (Curves are for locations near plume centerline.)

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TERRAIN INFLUENCE ON ATMOSPHERIC DIFFUSION

W. T. Hinds*

Continued analysis of diffusion data obtained during Operation Mountain Iron led to significant conclusions. The relation between time and distance regression was shown to lead to a prediction of relations between meteorological variabilities that was verified, thus assuring the reality of the two regressions. Evidence that rough terrain decouples diffusion from source point meteorological conditions is shown. The feasibility of using Lagrangian techniques in operational analyses was shown.

For many years now, most diffusion research (whether supported by the AEC or others) concentrated on diffusion over relatively flat terrain. With the significant progress attained in this field by the early 1960's, intensive research into the influence of mountainous terrain began. An earlier annual report in this series contained a description of a study entitled Operation Mountain Iron. The rather straightforward analysis presented in that annual report has been supplemented by continuing work, leading to some rather novel conclusions.

By a comparison of two regression equations of concentration on meteorological factors, one developed using time as the downwind variable, another using distance, it was possible to show that the two were related, leading to a combination of parameters equivalent to the Richardson's Number and verified by the data. Thus, the two regressions were not only statistically signifi-

cant, they were physically realistic. Furthermore, since the time-dependent analysis' was developed on the basis of Lagrangian principles (i.e., using a coordinate system attached to the diffusing particles), it was possible to show that Lagrangian principles can be applied to operational problems. This had never been clearly established before; Lagrangian techniques were known to be theoretically correct, but thought to be impractical.

A comparison of the results from Mountain Iron with results from earlier tests over flatter terrain brought to light a suggestive interaction between roughness and concentration variability. A summary of results is given in Table 1. Here diffusion programs using similar techniques are related to the roughness of the experimental site and the dependence of concentration on distance (X) alone, ignoring variations in meteorological factors during the test programs. The apparent relation (between predictive success using only distance and macroscale

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TABLE 1. Regression Equations as Related to Site Roughness

<u>Program</u>	<u>Best Fit Relation</u>	<u>% Data Predicted Within ± Factor of 4</u>	<u>Elevational Difference Over Test Site</u>
Dry Gulch and Ocean Breeze and Prairie Grass	$39.8 X^{-2.14}$	72	~20 m
Green Glow +30 Series	$6.85 X^{-1.85}$	85	~50 m
Mountain Iron	$0.39 X^{-1.66}$	92	~400 m

site roughness) points to a decoupling of diffusion from meteorological variations as terrain roughness increases. This curious hypothesis could mean, for example, that site evaluation in flat terrain would be much more expensive in terms of meteorological effort than site evaluation in mountainous regions. Because of the importance of this possibility, the data leading to Table 1 and other data available need to be thoroughly analyzed from this viewpoint.

Another conclusion not previously reported was that crosswind and vertical dispersion appear to be very approximately commensurate over rough terrain during daytime conditions, if vertical dispersion is determined from mass continuity, even in the presence of confining subsidence inversions at about the same elevation as the terrain relief. This is perhaps more indicative of tracer deposition than dispersion characteristics, but nothing more definite can be said without experimentation.

MODELING PEAK-TO-MEAN CONCENTRATION RATIOS IN DIFFUSING PLUMES

J. V. Ramsdell

A mathematical model for estimating peak-to-mean concentration ratios in diffusing plumes has been developed using linear least squares regression techniques.

During 1968 an analysis of average peak-to-mean concentration ratios ($\bar{P}/\bar{M}$) was made using data collected with the Hanford ^{85}Kr diffusion system.^(1,2) This analysis showed relationships between $\bar{P}/\bar{M}$ and (1) the normalized crosswind position within the mean plume (y/σ_y), and (2) the ratio of the time of passage of the plume to the interval over which the peak concentration was smoothed (T/t). During 1969 these relationships have been refined by a more detailed analysis. The refined relationship between $\bar{P}/\bar{M}$ and y/σ_y is shown in Figure 1, and that between $\bar{P}/\bar{M}$ and T/t is shown in Figure 2. These figures have been used as a guide in developing a $\bar{P}/\bar{M}$ model.

The double curvature in the relationship between $\bar{P}/\bar{M}$ and y/σ_y indicated that the modeling attempts should include $\tanh^2(y/\sigma_y)$ as a factor. Similarly, the relationship between $\bar{P}/\bar{M}$ and T/t indicated that the $\ln(T/t)$ might also be a factor. These functions, then, provide one input to the model. A second input was obtained from the following theoretical considerations:

- 1) At $T/t = 1$, PM ratios must equal 1 regardless of the crosswind position,

- 2) The maximum PM ratios should not exceed the value of T/t , and
- 3) Near the edge of the mean plume, PM ratios should approach the value of T/t .

In the modeling attempts the hyperbolic tangent and logarithmic functions were combined in various ways, each designed to meet the theoretical considerations. Regression coefficients were determined for each model using the data from which Figures 1 and 2 were derived and linear least squares techniques. Each model was judged by comparison of lateral and transverse cross-sections of the model with Figure 1 and 2. Quantitative estimates of the goodness of fit of each model to the data were not made because the variance of the individual P/M about the $\bar{P}/\bar{M}$ is not homoscedastic.

At the present time the best $\bar{P}/\bar{M}$ model, as judged by comparison with Figures 1 and 2, is:

$$P/M = 1.9 Y^2 X^4 + 0.90 X^2 + 1.0$$

where

$$Y^2 = \tanh^2 [(y/\sigma_y)/(X + 1)]$$

and

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FIGURE 1. Peak-to-Mean Concentration Ratios as a Function of Normalized Crosswind Position

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FIGURE 2. Peak-to-Mean Concentration Ratios as a Function of the Time Ratio

$$x^2 = \ln (T/t).$$

The behavior represented by this model is shown in an isometric view in Figure 3. The increase of $\overline{P}/\overline{M}$ with both y/σ_y and T/t is clearly seen in this figure. Qualitatively, this model can be evaluated by comparison of Figures 4 and 5 (which show projections of the model on the $\overline{P}/\overline{M}$ - y/σ_y and $\overline{P}/\overline{M}$ - T/t planes respectively) with Figures 1 and 2. This comparison shows that the behavior of the modeled $\overline{P}/\overline{M}$ is approximately the

same as the behavior of the measured $\overline{P}/\overline{M}$ ratios.

The present model is not entirely satisfactory, however, as the modeled $\overline{P}/\overline{M}$ ratios exceed the value of T/t near the edge of the mean plume for small values of T/t . It would also be desirable to be able to estimate the goodness of fit of models quantitatively. Modeling of peak-to-mean concentration ratios is, therefore, continuing in an attempt to rectify the deficiencies of the present model and to provide a quantitative evaluation of the model.

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FIGURE 3. Modeled Relationship Between Normalized Crosswind Peak-to-Mean Ratios

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FIGURE 4. Modeled Relationship Between Peak- to- Mean Ratios and Normalized crosswind Postion

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FIGURE 5. Modeled Relationship Between Peak- to- Mean Ratios and the Time Ratio

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PEAK-TO-MEAN CONCENTRATION RATIOS IN BUILDING WAKES

W. T. Hinds*

Results from a short but unique series of tests of diffusion from a point source located just upward of an obstacle are described. It was shown that diffusion in a wake is quite similar to bimodal plumes from an unobstructed point source, and that there was no important difference in peak-to-mean ratios between point source plumes and building wakes, if both sources are near ground level.

Up until the early 1960's, most atmospheric diffusion research, whether theoretical or experimental, was restricted to point or line sources in air flow with essentially parallel streamlines. However, it was clear that sources located near buildings must diffuse in more complex flow conditions. By the mid-1960's, several workers had elucidated many of the differences between mean concentration patterns in wakes and ordinary undisturbed patterns.

A parallel development in diffusion was the investigation of short term concentrations within a plume. Unsteadiness in atmospheric flow conditions leads to quite a variable concentration history within a diffusing plume, so that for brief intervals, concentration may be many times higher (i.e., peak concentrations) than the mean concentration. Some fair idea of the behavior of these peak-to-mean ratios had also been garnered by the mid-1960's.

* *Ecosystems Department*

But, there was essentially no information about the effects of building wakes on peak-to-mean ratios.

A short series of tests in building wakes was carried out with the objective of determining whether building wakes induce first-order differences in peak-to-mean ratios from an upwind point source at ground level in comparison with simple point-source diffusion. Several conclusions were important.

First, by comparing histories of concentration in point-source and building wake plumes, it was possible to show that building wakes increase the intermittency with which the plume intersects a given point. Further, the bilobate wake pattern is nearly indistinguishable from a bimodal pattern from a point source in undisturbed flow. Hence, the principal effect of a wake on a point source located just upwind of an obstacle is to alter diffusion to a bimodal character, increasing intermittency.

A second conclusion followed from a comparison of peak-to-mean ratios at sites within point source plumes with comparable data taken within wakes. In this comparison, data and analyses from ^{85}Kr experiments were invaluable in defining the relation between peak-to-mean ratios and position within a point-source plume. Direct comparison of point-source and building wake results showed there was no important difference in peak-to-mean ratios, a significant conclusion.

However, it is clear from previous work that diffusion within a building wake is dependent upon the manner in which the tracer or contaminant enters the wake. In particular, the results from this study do not apply at all to diffusion from a short chimney on top of a building, a common configuration. Therefore, a considerable amount remains to be done before building wakes are as well understood as point-source diffusion.

CONCENTRATION FLUCTUATIONS IN DIFFUSING PLUMES

J. V. Ramsdell

Concentration fluctuations in diffusing plumes are examined through cumulative frequency distributions of short-term concentration measurements and the intensity of the concentration fluctuations. Concentration intermittency and the intensity of fluctuations are larger than expected. Both cumulative frequency distributions and the intensity of fluctuations are a function of crosswind position in the mean plume.

In an earlier report⁽¹⁾ a particular aspect of concentration fluctuations in diffusing plumes was discussed, namely, peak-to-mean concentration ratios. The following presents a more general discussion of the concentration data used in that study and subsequently published by Nickola, Ramsdell, and Ludwick.⁽²⁾

Each concentration measurement was normalized to the local mean concentration and assigned a relative crosswind position in the mean plume. The normalized concentrations range in value from 0 to 5 at positions near the center of the plume and from 0 to more than 25 near the edge of the mean plume. This variation of the range of normalized concentrations with relative crosswind position suggested that the data be combined by crosswind position for analysis. Five crosswind groupings were therefore chosen arbitrarily and maintained throughout the analysis.

A cumulative frequency distribution for the normalized short-term concentrations in each crosswind

group is shown in Figure 1. These distributions indicate that, regardless of location in the mean plume, a large portion of the time the concentration will be considerably less than the mean concentration. If it is assumed that for all practical purposes the plume is absent from a position when the short-term concentration is less than 1/10 the mean concentration at that position, then at the mean plume axis the real meandering plume is absent more than 35% of the time and at the edges of the mean plume it is absent almost 80% of the time. Further up the cumulative distributions, it is interesting to note the frequency with which the mean concentration exceeds the short-term concentrations. This frequency varies from 65% near the mean plume centerline to about 85% at the edge. As the normalized concentrations become relatively large they may be considered as peak-to-mean concentration ratios. Thus, the cumulative frequency distributions in Figure 1 may also be used to estimate the frequency of occurrence of a

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FIGURE 1. Cumulative Frequency Distributions of Normalized 38.4 sec Concentrations Grouped by Crosswind Position

given peak-to-mean ratio. For example, at the centerline of the mean plume a peak-to-mean ratio of 5 can be expected to occur less than 0.5% of the time, while near the edge of the mean plume, peak-to-mean ratios of 5 occur more than 6% of the time.

It is obvious from Figure 1 and the above examples that, while the meandering plume frequents the center of the mean plume more often than the edge, high normalized concentrations are more frequent at the edge. This behavior can be more closely examined in Figure 2, where the intensity of concentration fluctuations (analogous to the intensity of

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FIGURE 2. Intensity of Concentration Fluctuations as a Function of Crosswind Position

turbulence) is shown as a function of normalized crosswind position. Near the mean plume axis the standard deviation of the short-term concentrations is only slightly larger than the mean concentration, but near the edge of the mean plume the standard deviation is more than triple the mean. This is almost identical to results reported by Becker, Rosensweig, and Gwozda(3) for turbulent diffusion in pipe flow.

Qualitatively the behavior of plumes shown in Figures 1 and 2 is much as expected; quantitatively, however, the intermittency and intensity are larger than anticipated. The magnitude of the intermittency factor and the intensity of fluctuations serve to emphasize the need for

caution when attempting to extrapolate diffusion data from one time scale to another.

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MESOSCALE STUDIES AT HANFORD

H. G. Daubek and M. A. Wolf

Wind velocity summaries at several sites on the Hanford reservation, streamline analysis and the tracking of tetroons and pibals were used to identify mesoscale anomalies in the airflow which preclude the use of single-site data in the accurate definition of the trajectory and diffusion of pollutants on the reservation.

It is common in atmospheric diffusion work to evaluate the first few miles of plume travel and growth with pertinent parameters measured near the source. At Hanford, wind velocity climatology from the 200 ft level of the Meteorology Tower (622) has been used without qualification at other sites. Both of these practices assume spatial homogeneity. Unpublished summaries by Scoggins and Engelmann have shown significant

spatial variability of wind velocity near the surface which cast doubt on the validity of this assumption. Requirements for detailed trajectory information for potential releases at sites remote from the Meteorology Tower led to an investigation of mesoscale variability.

The investigation, thus far, has emphasized the analysis of a two-year record of wind velocity data acquired from a network of seven wind

sensors including one at the 200 ft level of a second tower (100N). Data from this latter source also have been compared with data from the Meteorology Tower. Computer programs were utilized to provide summaries and comparisons of the data for the several sites.

One technique undertaken to evaluate the airflow patterns was stream-

line analysis. The dates to be analyzed were randomly selected for preparation of 159 streamline analyses. Figure 1 is an example of a vortex pattern which occurred in approximately 30% of the prepared streamline analyses. Such vortex circulations which have been observed with tetrons and drifting smoke from Hanford power plant plumes

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FIGURE 1. Streamline Analysis March 13, 1969 0200 hr

indicate that these surface patterns are present in the overlying air as well.

The complexity of airflow is also demonstrated in the comparison of wind velocity summaries at individual sites. Wind shear between the 100N and 622 towers is implied by differences in the tower data summaries. Figure 2 shows the percent frequency of wind direction at each site. It may be noted that 100N has a higher percent frequency of easterly winds, a lower percent frequency of southwest winds, and a much lower percent frequency of northwest winds. A comparison of wind direction versus speed at 622

and 100N indicates that the highest wind speeds are from the southwest at 622 while at 100N the higher wind speeds are from the northwest. Also, there are fewer observations in the higher wind speed ranges at 100N than at 622. It appears that the basin in which the 100N tower is located provides shelter from strong southwest winds.

At individual sensor sites the summarizations of wind directions as a function of time of day give evidence of the diurnal effects and channeling which result from local terrain influences. Diurnal wind direction changes appear at all

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FIGURE 2. Percent Frequency Distribution of Wind Direction, 622 and 100N, 200 ft levels

sites but one. Figure 3, for example, shows a prevailing wind of 70 degrees during night and a wind direction of 250 degrees during the day at site 101. There was little diurnal effect noted at site 104, shown on Figure 4, where the flow is channeled along the river.

A limited investigation of the overlying airflow was conducted with tetrooms and pibals. Tetrooms released in the vicinity of the 100N tower were tracked at low elevations to observe the horizontal and vertical motions of the air which would transport and diffuse potential effluent. The vertical projection of one of these tetroom flights, included as Figure 5, shows a high

altitude portion which has been observed on all flights over the Columbia River. Since the tetroom follows a constant density surface, this indicates that the air was more dense over the river than over the land on these days. Horizontal projections from the tetroom flights appear to be consistent with surface streamlines.

Simultaneous release of pibals from two or more sites is another useful technique in mesoscale analysis. Figure 6 shows one such release from the Meteorology Tower and the FFTF site which is approximately 15 miles to the southeast. Most striking is the abrupt wind shift above 3500 ft which is a

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FIGURE 3. Percent Frequency of Occurrence, Wind Direction Versus Time of Day at 101

	TIME OF DAY																							
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24
010	.01	.00	.01	.00	.01	.01	.02	.06	.07	.07	.11	.09	.06	.07	.07	.06	.02	.05	.01	.01	.01	.00	.01	.01
020	.01	.01	.02	.01	.01	.02	.02	.04	.06	.12	.13	.12	.14	.10	.10	.12	.03	.05	.02	.01	.00	.01	.00	.00
030	.01	.01	.01	.00	.01	.02	.04	.07	.09	.15	.16	.21	.18	.14	.16	.12	.11	.05	.01	.03	.00	.01	.01	.01
040	.01	.01	.01	.01	.01	.03	.04	.06	.09	.14	.14	.12	.17	.15	.18	.18	.11	.06	.04	.03	.01	.01	.01	.01
050	.01	.02	.02	.00	.03	.01	.07	.07	.12	.04	.06	.09	.11	.12	.10	.11	.06	.04	.06	.03	.03	.01	.02	.02
060	.02	.01	.02	.01	.02	.05	.07	.04	.05	.05	.08	.06	.07	.09	.10	.07	.08	.04	.07	.04	.01	.02	.02	.02
070	.02	.01	.01	.04	.02	.02	.04	.05	.05	.03	.06	.06	.06	.12	.06	.02	.08	.09	.09	.08	.07	.03	.01	.01
080	.04	.02	.01	.04	.04	.04	.07	.07	.06	.04	.04	.10	.05	.05	.06	.08	.07	.06	.09	.04	.04	.01	.01	.01
090	.04	.04	.06	.05	.06	.07	.08	.07	.07	.04	.09	.04	.08	.09	.07	.07	.07	.11	.10	.04	.05	.02	.02	.02
100	.04	.07	.05	.03	.07	.04	.06	.07	.05	.07	.06	.07	.04	.04	.06	.09	.09	.11	.12	.08	.06	.05	.06	.04
110	.02	.05	.05	.06	.09	.07	.06	.07	.10	.08	.07	.02	.02	.06	.03	.03	.05	.07	.06	.06	.02	.02	.04	.02
120	.04	.04	.08	.06	.05	.06	.06	.06	.06	.06	.04	.07	.07	.05	.04	.06	.07	.09	.11	.04	.04	.02	.01	.04
130	.05	.04	.07	.02	.04	.04	.04	.05	.04	.04	.02	.06	.01	.02	.04	.07	.11	.06	.08	.05	.04	.01	.01	.01
140	.03	.08	.07	.11	.11	.09	.05	.03	.06	.05	.05	.04	.05	.03	.03	.05	.06	.05	.07	.04	.04	.05	.06	.04
150	.03	.07	.04	.08	.09	.03	.07	.06	.03	.02	.03	.05	.03	.04	.02	.09	.04	.05	.04	.06	.06	.04	.03	.05
160	.06	.04	.02	.04	.07	.07	.04	.06	.02	.04	.03	.04	.02	.04	.05	.03	.06	.03	.03	.05	.06	.03	.04	.04
170	.01	.01	.06	.08	.06	.07	.01	.06	.02	.02	.02	.00	.02	.03	.03	.01	.05	.03	.04	.01	.01	.04	.05	.02
180	.01	.02	.03	.04	.06	.06	.04	.03	.02	.02	.01	.04	.03	.02	.01	.01	.06	.06	.07	.02	.02	.03	.00	.03
190	.04	.04	.02	.04	.02	.04	.03	.02	.02	.02	.02	.01	.01	.01	.05	.04	.02	.03	.03	.04	.02	.00	.02	.01
200	.02	.05	.05	.04	.07	.04	.01	.03	.02	.02	.01	.03	.02	.05	.00	.02	.04	.02	.02	.02	.04	.02	.01	.01
210	.02	.05	.04	.02	.02	.04	.02	.01	.04	.01	.03	.02	.04	.04	.03	.02	.04	.07	.02	.06	.01	.03	.02	.02
220	.06	.06	.07	.06	.04	.06	.04	.04	.02	.01	.02	.04	.05	.03	.05	.06	.05	.04	.07	.07	.07	.03	.04	.09
230	.07	.06	.05	.07	.09	.05	.08	.06	.06	.03	.04	.02	.07	.06	.04	.07	.06	.06	.09	.05	.07	.06	.10	.07
240	.09	.06	.09	.06	.07	.07	.03	.04	.06	.01	.08	.07	.09	.11	.07	.05	.12	.11	.07	.13	.08	.14	.10	.09
250	.17	.19	.12	.16	.14	.09	.11	.14	.11	.12	.12	.12	.20	.19	.17	.18	.17	.12	.12	.15	.15	.19	.16	.20
260	.22	.20	.19	.16	.17	.19	.21	.19	.23	.22	.21	.27	.24	.19	.29	.20	.26	.21	.22	.24	.35	.30	.28	.25
270	.45	.36	.29	.41	.25	.32	.36	.35	.37	.40	.39	.41	.27	.30	.33	.33	.24	.25	.22	.27	.34	.44	.48	.38
280	.36	.42	.38	.26	.37	.38	.30	.39	.39	.39	.33	.40	.35	.29	.34	.33	.28	.30	.23	.35	.37	.41	.45	.40
290	.49	.42	.46	.45	.34	.35	.40	.36	.35	.35	.28	.32	.28	.22	.22	.22	.21	.28	.38	.48	.54	.57	.45	.53
300	.53	.58	.61	.61	.55	.53	.51	.45	.40	.39	.33	.24	.28	.35	.28	.33	.45	.40	.56	.51	.48	.59	.66	.65
310	.40	.40	.40	.39	.38	.31	.35	.33	.23	.20	.30	.24	.24	.24	.24	.30	.41	.31	.29	.32	.32	.36	.36	.36
320	.26	.25	.24	.24	.23	.24	.27	.20	.21	.19	.17	.14	.12	.14	.15	.24	.19	.15	.19	.17	.15	.23	.20	.20
330	.08	.04	.08	.06	.06	.07	.07	.11	.08	.09	.10	.10	.04	.12	.11	.09	.08	.11	.10	.08	.10	.05	.09	.09
340	.07	.06	.06	.04	.04	.06	.05	.11	.04	.04	.05	.06	.05	.06	.05	.06	.04	.06	.06	.06	.04	.05	.04	.04
350	.03	.03	.01	.03	.04	.02	.04	.02	.06	.04	.06	.04					.04	.02	.02	.02	.04	.03	.01	.02
360	.03	.06	.05	.04	.06	.06	.07	.10	.11	.09	.12					.13	.12	.07	.08	.06	.07	.05	.04	.02
VAR	.21	.23	.25	.24	.28	.2	.27	.19	.21	.22	.32	.30	.30			.33	.26	.20	.19	.14	.22	.29	.17	.20
CALV	.13	.06	.08	.10	.14	.1	.1	.11	.10	.09	.06	.05	.06	.04	.04	.08	.07	.07	.12	.07	.10	.09	.11	.09

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FIGURE 4. Percent Frequency of Occurrence, Wind Direction Versus Time of Day at 104

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FIGURE 5. Tetraodon Flight No. 15, Vertical Positions

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FIGURE 6. Simultaneously Released Pibals from the Meteorology Tower (622) and the FFTF Site

clear indication of the influence of surrounding terrain on winds at lower levels. The differences in speed and direction at the two sites in the lower levels show the unequal effect of the orographic influence. Further definition of the mesoscale meteorology will be possible with the AEC aircraft which will be instrumented for precise measurement of temperature and humidity.

Comparisons of wind velocity summaries at several sites, streamline analyses, and the tracking of

tetroons and pibals all indicate that complex mesoscale flow at Hanford precludes the use of data from a single instrumented site for the definition of pollutant trajectory and diffusion. The several techniques which were used and the additional employment of an instrumented aircraft can contribute significantly to the identification and understanding of mesoscale anomalies. Such understanding will lead ultimately to improved predictability of mesoscale meteorology.

ATMOSPHERIC BOUNDARY LAYER STUDIES

The planetary boundary layer comprises approximately the lowest kilometer of the atmosphere, that portion of the atmosphere within which man lives and works and which is the prime source for atmospheric pollution, the pollutant sink of most concern, and an important medium for short and medium-range transport of contaminants. Studies of the boundary layer are presently aimed at describing the turbulent motions which cause atmospheric diffusion and the organized convective motions which lead to vertical transport of contamination. Knowledge of the evolution, maintenance, structure, and decay of turbulence and convective elements is needed to parameterize adequately these aspects of the atmosphere and relate them to diffusion and transport studies. A detailed understanding of the dynamics and structure of turbulence and of organized convective motions is also essential to studies of the distribution and consequences of thermal pollution of the atmosphere.

Earlier single-point studies of turbulence structure and turbulent transport as a function of height and stability have been extended recently by simultaneous, multiple-level, turbulence measurements of both wind and temperature. Initial analysis of these experiments has demonstrated the vertical gradient of momentum transport above the constant flux layer. Present measurements from towers will be extended and supplemented in the future by aircraft measurements throughout the boundary layer. The development of aircraft instrumentation for the measurement and recording of ambient atmospheric conditions is reported in what follows.

TURBULENCE STUDIES IN THE ATMOSPHERIC BOUNDARY LAYER

T. W. Horst and C. E. Elderkin

Five, 35-min periods were selected for analysis from 18 hr of micrometeorological data recorded during 1969. Necessary steps are outlined for obtaining the surface Reynolds stress from cup anemometer wind profiles taken over terrain requiring a zero-plane displacement correction. Directly measured values of the Reynolds stress at 16 m and 58 m were combined with the profile derived stress at 4 m to obtain a vertical gradient of momentum transport. This gradient was found to agree with estimates obtained from the averaged equation of motion in the direction of the mean wind except for the case of maximum thermal instability. Here the 35-min period was found to sample inadequately the lower frequency contributions to the Reynolds stress.

During the month of July, 1969, 15 hr of three-level turbulence data were recorded during a wide range of atmospheric stability conditions. The data include turbulent fluctuations of the three components of the wind at heights of 4 m, 16 m, and 58 m, turbulent temperature fluctuations at 4 m and 58 m, and mean profiles of temperature, wind speed, and wind direction from 2 m to 46 m. Further description of the experimental site and equipment can be found in the Annual Report for 1968.⁽¹⁾ In order to provide a basis for comparison of the three-level turbulence data, the three fast-response anemometers were then set up adjacent to each other at a height of 4 m, and 3 hr of instrument comparison data were recorded.

During 1969 the capability has been developed at Hanford for a wholly digital computer analysis of the turbulence data, including

a fast-Fourier-transform spectral analysis computer program; this method of data reduction will be used for the majority of our future work. At present the data from the two sonic anemometers, which were at 16 m and 58 m during the three-level experiments, have been analyzed. The Wind Component Meter data, taken at the 4 m level, will be analyzed after further calibration studies are performed with this anemometer. Four, 35-min periods from the three-level data, as well as 35 min of the anemometer comparison data, were selected for instrument performance evaluation and micrometeorological analysis.

The purpose of these experiments was to investigate the height dependence of turbulence statistics and spectra, the turbulent transport of momentum and heat in the atmospheric boundary layer, and the turbulent energy budget in the boundary layer. In this report we will discuss the

reliability of the experimental measurements and the vertical gradient of the turbulent momentum transport.

Analysis of the data is continuing.

Table 1 is a statistical summary of the experimental runs. Calibration corrections of the order of 10% were made to the data obtained from the horizontal axes of the 16 m sonic anemometer. This correction was based on the statistics and power spectra from the anemometer comparison experiment. Calibration changes

between experimental runs are less than 1%. Measurements of the sonic anemometer transducer arrays indicate that alignment and leveling errors in the Reynolds stress⁽²⁾ are less than 5%, and no corrections have been made for these errors.

Comparison can also be made between the sonic anemometers and the profile measurements. A mean vector wind, $\bar{u}$, was calculated from the mean wind speed measured by the cup

TABLE 1. Statistical Summary of Experiments (a)

Period	Instrument	$\bar{u}$	$\frac{2}{u}$	$\frac{2}{v}$	$\frac{2}{w}$	u_*^2
3A	58 m Sonic	8.90	0.602	0.497	0.184	0.061
	16 m Sonic	6.70	0.915	0.473	0.251	0.178
	Profile Data	6.65 ^(b)				0.15 ^(c)
3E	58 m Sonic	7.80	0.297	0.120	0.041	0.0115
	16 m Sonic	5.24	0.491	0.187	0.099	0.062
	Profile Data	5.0 ^(b)				0.089 ^(c)
4A	58 m Sonic	9.54	1.54	0.700	0.450	0.189
	16 m Sonic	8.69	2.24	0.824	0.394	0.32
	Profile Data	8.2 ^(b)				0.29 ^(c)
4B	58 m Sonic	8.62	1.95	1.86	0.876	0.54
	16 m Sonic	7.62	2.91	1.87	0.471	0.43
	Profile Data	7.65 ^(b)				0.25 ^(c)
Comparison Experiment	58 m Sonic ^(c)	5.43	1.88	0.919	0.289	0.26
	16 m Sonic ^(c)	5.49	1.84	0.846	0.299	0.23
	Profile Data	5.7 ^(c)				0.27 ^(c)

a. all wind speeds in m/sec

b. at 16 m

c. at 4m

anemometer, $\bar{v}_h$, by assuming a normal distribution of θ , the instantaneous wind direction, (3)

$$\bar{u} = \bar{v}_h \exp \{-1/2 \sigma_\theta^2\}. \quad (1)$$

Here σ_θ^2 is the variance of θ , measured by wind vanes. The measured wind profile was also modified by the use of a zero-plane displacement, d , to give a logarithmic profile under neutral conditions,

$$\bar{u} = \frac{u_*}{k} \ln \left(\frac{z - d}{z_0} \right). \quad (2)$$

Here u_* is the friction velocity, k is the Karman constant, and z_0 is the roughness length. The zero-plane displacement is a necessary addition to the usual logarithmic profile for surfaces covered with high vegetation, such as the sagebrush at Hanford, and has been found to be valid where the roughness elements are sufficiently uniform in height and distribution. (4) The best fit of the Hanford data is given with values of d and z_0 equal to 140 cm (a value close to the average height of the roughness elements) and 3 cm, respectively (Figure 1).

The values of $\bar{u}$ and u_*^2 given by these modified profiles for the anemometer comparison experiment agree well with the data from the sonic anemometers, within 5% for the mean wind and within 8% for the friction velocity. In addition to verifying the sonic anemometer data, the modified profiles exemplify the care which must be exercised in deriving values

for the surface stress from cup anemometer data taken over nonideal surfaces. Values of u_*^2 derived from uncorrected profile data differed by 50 to 100% from those measured by the sonic anemometers. Finally, it is noted that Bernstein (3) was unable to directly check Equation 1 due to the nonavailability of simultaneous profiles of $\bar{v}_h$ and θ . The present use of Hanford profile data along with directly measured values for u , is evidence for the validity of Equation 1.

Since in all cases the profile below 4 m is found to be close to that expected for neutral thermal stratification, the values for u_*^2 at a height of 4 m can be derived from the profile data. These are listed in Table 1 along with the values of u_*^2 from the sonic anemometers at heights of 16 and 58 m. In the first three periods there is a decrease in the Reynolds stress from 16 to 58 m and in the fourth period there is an increase. In the second and fourth periods, where the 4 and 16 m data differ by large percentages, the 4 m Reynolds stress values follow the stress gradient indicated by the sonic anemometer data. However, in the first and third periods the 4 to 16 m change is opposite in sign to the 16 to 58 m change, but the 4 and 16 m values differ by less than 20%, a reasonable error limit for an experimental measurement of u_*^2 . It is concluded that the Reynolds stress gradients reflected in the three values are true measures for the time periods analyzed.

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FIGURE 1. Wind Speed Profile from Anemometer Comparison
Experiment [(Ri)_{4m} = 0.01]

These Reynolds stress gradients must be interpreted in terms of the equation of motion in the direction of the mean wind,

$$\frac{\partial \bar{u}}{\partial t} = -\frac{1}{\rho} \frac{\partial p}{\partial x} + \frac{\partial}{\partial z} u_*^2. \quad (3)$$

Here p is the pressure, ρ is the density of air, x is the coordinate in the direction of the mean wind, and we assume horizontal homogeneity for the wind field and neglect molecu-

lar momentum transport. Table 2 lists the values for the three terms. The acceleration was calculated from 10 min averages of the 32 m wind speed at the beginning and end of the 35 min period, the pressure gradient was calculated from the equations of the Ekman spiral and an estimate of the boundary layer thickness, which is strictly valid only for adiabatic conditions, (5)

TABLE 2. Evaluation of Equation 3

Period	$(R_f)16 \text{ m}$	$\frac{a\bar{u}}{\partial t} \text{ m/sec}^2$	$-\frac{1}{\rho} \frac{\partial p}{\partial x} \text{ m/sec}^2$	$\frac{a}{\rho} u_*^2 \text{ m/sec}^2$
3A	0.034	-3.8×10^{-4}	2.7×10^{-5}	-2.41×10^{-5}
3E	0.134	-1.5×10^{-4}	1.6×10^{-3}	-1.07×10^{-3}
4A	-0.078	-1.2×10^{-4}	4.5×10^{-3}	-2.41×10^{-3}
4B	-0.346	1.17×10^{-3}	3.8×10^{-3}	3.62×10^{-3}

$$H = 0.2 u_o^*/f, \quad (4)$$

where f is the coriolis parameter, u_o^* is the surface friction velocity estimated by the 4 m value, and the stress gradient was calculated from the sonic anemometer data. The acceleration should be accurate to 10%, the pressure gradient to 50%, and the stress gradient to 20%.

In the first three cases the acceleration term is quite small, and the residual from adding the pressure gradient and stress gradient terms is smaller than the estimated errors, indicating that the individual terms have been correctly evaluated. As expected, the horizontal pressure gradient accounts for the large observed stress gradients. In the fourth case, however, the terms do not balance. The error is found in the Reynolds stress measurement. Inspection of the cospectra between the vertical and longitudinal wind fluctuations for the two anemometers (Figure 2) shows that there is a large contribution to the measured Reynolds stress from frequencies below $3 \times 10^{-3} \text{ Hz}$ (the frequency at which a marked cospectral gap occurs at both heights) and that the

contribution from the higher frequencies is nearly the same for the two heights. This indicates that the measured stress gradient is primarily due to the low frequency contributions, probably from organized convective elements, which were inadequately sampled during the 35-min period that was analyzed. This is in agreement with Kaimal's contention that "averaging periods of the order of an hour are needed to provide meaningful profiles of momentum flux during unstable conditions."⁽⁶⁾

As can be seen from an evaluation of Equation (3) and the measurements presented here, the constant flux layer is quite shallow compared to the depths of the atmospheric boundary layer within which surface diffusion and transport take place. Existing similarity theory for the constant flux layer must be extended or supplemented in order to develop valid models for the turbulence at greater heights. Continued study will be made of the flux gradients, and the turbulent spectra and turbulent energy budget will be analyzed in order to better parameterize the atmospheric processes occurring above the constant flux layer.

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FIGURE 2. uw Cospectra from Period 4B

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AIRCRAFT INSTRUMENTATION FOR BOUNDARY LAYER
METEOROLOGICAL STUDIES

F. O. Gladfelder and C. E. Elderkin

A single engine (Cessna 305A) aircraft has been instrumented to record temperature, humidity, altitude, and air speed in support of boundary layer studies at Hanford.

Aircraft have been used in meteorological studies at Hanford in the past, and plans for the future require an even greater measurement capability with airborne instrumentation. Presently, a Cessna 305A (military designation: L-19) is available for meteorological programs. The Cessna 305A is a single engine, high wing aircraft, accommodating a pilot and technician in tandem. It can operate safely at speeds as low as 60 miles per hour and, while it has serious space and weight limitations, it is especially useful for low level meteorological measurements. By removing the rear seat, and installing a rack specially designed and fabricated for quick removal and reinstallation, it is readily equipped with data recorders and instrument electronics. Sensing equipment is suspended from the aircraft's right wing strut.

The aircraft is equipped with a 50 ampere generator supplying a 28 volt dc battery. The 28 volts dc provided by the aircraft electrical system is converted to 120 volts ac by a Topaz static inverter, supplying power up to 500 volt-amperes for operation of the instrumentation.

A Metrodata DL 620A recorder has been included in the aircraft measurement and recording system. It provides for digital recording of 18 channels of data plus 2 time channels on magnetic tape at a rate of 48 channels per second. It is especially well suited for use in aircraft because of its light weight (18 lb) and low power consumption (35 watts).

Presently, the airborne system measures and records temperature and humidity from thermistor and carbon strip hygistor probes along with aircraft altitude and air speed. While this extends the capability for determining mean variables above existing tower-based measurements in connection with diffusion studies through greater depths in the boundary layer, it is also planned to add turbulence instrumentation to the system.

A Metrodata DL 622A tape reader for the recorder has been interfaced with the SEL 840A computer which is presently being used for preparation of meteorological data from other measurement and field recording systems. The data can be treated in the SEL 840A or transferred to

computer compatible digital tape for more extensive analysis on the UNIVAC 1108 computer available at Hanford.

Data have been measured and recorded with the system, and reason-

able results were obtained. A comprehensive comparison of inflight measurements from the system with tower-based measurements is planned for the near future.

BOMEX STUDIES

The relative importance of sea-air interaction processes to an understanding of basic environmental processes and to improvements in weather prediction has been more fully recognized in recent years. The marine environment has been identified as an area for intensified research. The processes of heat exchange, evaporation, and frictional coupling interact to drive the oceans and atmosphere. These fluxes may be studied at their source by observations in the near-surface boundary layers at the sea-air interface by means of buoys. Understanding radionuclide transport and deposition is inextricably part of understanding these fluxes as well as atmospheric processes on larger scales. At the same time, the presence of radionuclides in the atmosphere provides a source of tracers for studying atmospheric processes. The fluxes of heat, momentum, and moisture are major factors to be considered in dealing with hurricanes and marine pollution. Both are problems of economic and social importance requiring immediate attention. The location of our major population centers along seaboards and the Great Lakes, as well as the value of the marine resources, emphasizes the importance of sea-air interaction studies. The location of nuclear power plants in coastal areas requires careful evaluation of the marine environment as a potential dispersant of heat and radioactive pollutants.

THE DEVELOPMENT AND TESTING OF A STABLE BUOY AND
INSTRUMENT SYSTEM FOR STUDIES AT THE SEA-AIR INTERFACE

R. K. Woodruff and A. G. Dunbar

The fluxes of momentum, heat, and moisture at the sea-air interface affect atmospheric processes on many scales. These processes control the transport and deposition of radionuclides. In 1969, Battelle-Northwest joined Florida State University in preparation and operation of the FSU TRITON buoy system during BOMEX for measuring mean meteorological and oceanographic parameters and the turbulent eddy fluxes of momentum, heat, and moisture. These measurements can be used with profiles of radionuclide concentration to estimate deposition, and for the investigation of other basic sea-air interaction processes. The power requirement of the turbulence system resulted in the development of a unique thermoelectric generator power supply. During the BOMEX, the power supply was demonstrated to be applicable and functional. Difficulties, related to the initial incompatibility of the added turbulence data acquisition system, resulted in a limited amount of turbulence data. After some modification, the data system operated reliably as an independent system during a subsequent shore-based study. Although the TRITON system was not fully integrated with success during BOMEX, it is anticipated that this can be achieved in the near future. Refinements to the buoy vehicle, data acquisition systems, turbulence sensor array, and buoy motion sensors are planned with additional bench and field testing in 1970.

The importance of sea-air interaction processes in the near-surface boundary layers to long-range weather prediction, the transport and deposition of radionuclides and pollutants, hurricanes, and to an understanding of basic environmental processes guided the development of a research capability within the Atmospheric Resources Department focused on the marine environment. An initial step was taken in this direction by joining the Department of Oceanography, Florida State University, Tallahassee, Florida, in the further development, testing, and operation of the FSU TRITON buoy system.

A description of the initial development and the field operation

during the Barbados Oceanographic and Meteorological Experiment (BOMEX) is presented. The results to date give reason for an optimistic view of possible future success of the system as a necessary and valuable tool for studies of the sea-air surface boundary layers.

The TRITON buoy was initially developed by Florida State University to provide a stable platform for measuring mean atmospheric and oceanographic parameters and wave spectra in the surface boundary layers. The data, when related to other data defining larger scale features, can be used to investigate the energy and moisture transfer at the sea surface and the processes by which the

surface boundary layer, the planetary boundary layer, and the synoptic scale process interact, feed upon, and distribute this energy and moisture for the maintenance of a global energy balance.

The accuracy of the necessary measurements required a stable platform to support the sensors and a data acquisition and storage system. The entire system was to be anchored in the open (deep) ocean, where conditions are unaffected by continental influences, and allowed to operate unattended for days at a time.

The resultant buoy, TRITON, was of the spar buoy configuration, sufficiently large to provide a relatively stable platform, yet small enough to induce a minimum of contamination into the measurements. The buoy is portable for overland and at-sea transportation. For overland transportation, it can be disassembled into components which can be readily transported by rail, truck, or air freight. For deployment at sea, the main body of the buoy is assembled ashore, lowered into the water, and towed to its anchorage where the air-filled cylinder at the bottom end can be flooded. This rights the buoy such that 80 ft of its 115 ft length is submerged, giving it relative stability against waves. Under rough sea conditions, the buoy will pitch and roll less than 3 to 5 degrees. With wave heights of 6 to 8 ft, the heave is about 15 cm. The lowest resonant period is 10 to 13 sec. Upon completion of the mooring, the buoy can be manned for installation of the sensor and data

recording systems. The reverse operation retrieves the buoy.

Two levels of above water sensors monitor mean wind speed, direction, humidity, and temperature. A rain gage at the mast top can provide rainfall measurements. At the waterline, a multidimensional wave staff array was used for determination of directional wave spectra. Just below the water surface, wave pressure, water temperature, and current velocity and direction sensors are located. Another current meter and thermistor is located at a depth of 10 m. Within the buoy, a compass provided directional reference; accelerometers and inclinometers sensed vertical and horizontal accelerations, pitch and roll. The data acquisition system was made up of three parallel commutators, digitizers, and recorders run by a common digitizing oscillator. Additional details will be given below. The power supply was made up of batteries.

This buoy configuration was successfully tested during the FSU Barbados Experiment in 1968 although the desirability of refinements was apparent.

In 1968, Battelle-Northwest joined FSU in efforts to improve and extend the capabilities of the TRITON buoy for participation in the 1969 BOMEX. The goal of the Atmospheric Resources Department was to instrument the buoy with turbulence instrumentation for direct eddy flux measurements of momentum, sensible and latent heat, and moisture. It was anticipated that direct eddy flux measurements, in conjunction with the other buoy

observations, would provide several opportunities.

- 1) Direct measurements of the eddy fluxes would provide a point reference value for comparison with aircraft observations and estimates from large scale budget calculations to be made during BOMEX. In addition, the system could be left in operation between experiment phases and provide data continuity.
- 2) Estimates of dry radionuclide deposition could be made utilizing the direct eddy flux measurements, profile measurements from the buoy, and the radionuclide profile data to be collected from the buoy tender and the U.S. Navy Floating Instrument Platform. In addition to the radionuclide concentration profiles, the Radiological Sciences Department of BNW would measure concentration profiles in the ocean, and rainfall rate and nuclide concentration; the rainfall measurements provide an estimate of the deposition by rain. With these estimates, it is possible to evaluate to what extent a radionuclide mass budget model of the form:

$$\frac{dC_w}{dt} = F_r + F_{rw} - \alpha C_w$$

can be satisfied,
where

C_w = concentration of radionuclide in the column of water reaching to a depth where the concentration is nil,

F_r = dry turbulent radionuclide flux entering the water from the air,

F_{rw} = radionuclide flux entering the water in rainwater,

$-\alpha C_w$ = is the rate at which the radionuclide is being depleted in the water column as the result of natural decay, where α is a constant characteristic of a particular nuclide, and

t = time.

Such a model requires horizontal homogeneity, or zero relative horizontal advection between the local source and sink, and that the profiles are in equilibrium. These requirements would be most closely approximated at a remote open ocean site.

- 3) One of the prime requirements for improved environmental modeling and prediction is the incorporation of fluxes of energy into the atmosphere at the sea-air interface. However, great numbers of these measurements are required. Instrumentation to measure eddy fluxes directly is sophisticated and expensive and requires continual maintenance. Therefore, eddy flux measurements would be impracticable. Eddy flux measurements when observed simultaneously with other meteorological and oceanographic parameters, including profiles and wave characteristics, can be used to attempt to relate these other parameters to the desired fluxes. The ultimate goal is some relatively simple and

universal relationship of parameters that can be economically measured with sufficient density and accuracy to satisfy the requirements of long-range prediction models. Although progress has been made by several investigators, very few have had the opportunity to analyze data representative of the situation for which the analysis was intended. The variety, quantity, and quality of data which are considered reasonable to expect from a system of the configuration being discussed could result in meaningful progress.

- 4) Direct eddy flux measurements, when simultaneous with measurements of processes of the various larger scales can provide data for studying how the energy transfer at the sea-air interface feeds the various scales of motion and ultimately the global scale atmosphere.
- 5) Ocean surface currents and waves are produced by frictional coupling with the wind. The relationships between the eddy flux of momentum and the generation and maintenance of these appear to be complex. Again, this is an area where accurate measurements of the processes in their natural equilibrium will be of great value.

The basic requirements for eddy flux measurements at sea, after providing a stabilized platform, are a quality data recording system and high frequency response sensors. As FSU had successfully used a commercially available digital recording

system, the same system was selected for recording turbulence data in order to maintain system compatibility. Sensors for turbulence measurements had been in use in other work at Hanford and their application for buoy use appeared to be reasonable.

The power necessary for the additional recording device, turbulence sensors, and a gyro required the redesign and development of a buoy power supply. This resulted in a unique system utilizing 12 catalytic thermal electric burners. The 12 burners were arranged symmetrically in a spoke-like configuration inside a normally submerged (25 ft) section of the main buoy tube (Figure 1) with the thermopiles in contact with the inside surface of the buoy tube. The high thermal conductivity of the metal tube and the high thermal capacity of the water outside provided an excellent heat sink and maintained a temperature differential across the thermopile sufficient to generate 250 watts of electrical power. The maximum power capacity of this system has been computed to be 400 watts, but has not been tested at that power load. The subsurface location required that air intake and exhaust lines be run to a point well above the water surface. The thermoelectric generator (TEG) fuel source was 300 lb of propane stored at 75 psi in a buoy tube section located 40 ft below the waterline (Figure 1). Normal fuel consumption at a power load of 250 watts is 100 lb

FIGURE 1. TRITON Buoy Configuration
During BOMEX

per week. The TEG is used in conjunction with a battery pack which is charged as necessary by the TEG. Voltages are stepped and regulated ahead of the equipment loads as necessary.

The data acquisition system is diagrammed in Figure 2 showing the four parallel systems sharing a common timer/programmer unit for time correlation of all measurements and for controlling a common on-off sampling sequence. The telemetry link shown has not been developed for applications to date. The on-off program was initially set for 20 min on and 100 min off; however, this is programmable.

The digitizing rate is controlled by a common oscillator at 24 words per second, each word containing 16 information bits. With the sampling format used, this limits aliased energy in power spectra to that energy above 2 cycles/sec for sonic wind components and temperature, and 1 cycle/sec for the propeller anemometer components and wet bulb temperature (humidity).

The digital tape decks use a continuous loop, two-track cassette tape and record on each track, one at a time, requiring that the tape cycle twice to fill both tape tracks. Data are recorded at a bit density of 525 bits per inch. The tapes, being 1200 ft in length, provide 8 hr of continuous operation, or 3 days of unattended operation when used in the 20/100-min on-off mode (at 24 words per second).

The sensor configuration for the buoy used for BOMEX was essentially

FIGURE 2. TRITON Digital Data Acquisition System.

that used by FSU in 1968, as described above, with the addition of the turbulence instrumentation and a gyro. The turbulence sensors consisted of a three-component sonic anemometer, a three-component propeller anemometer and wet and dry bulb thermistors.

The sonic anemometer has a frequency response greater than the sampling rate used, accuracy of $\pm 3\%$, and resolution of ± 2 cm/sec. The propeller anemometers have a distance constant of 0.82 m, a threshold of 0.22 to 0.36 m/sec, about 1% accuracy and resolution of ± 5 cm/sec. The cosine response is approximated to within $\pm 3\%$ for attack angles between 60 and 120 degrees, and is correctable when the data are processed. The thermistors for the wet and dry bulbs

were calibrated to within ± 0.1 °C and had a response time of 0.1 sec. The response time of the wet bulb with the wick wetted has not been determined. The thermistors are housed in naturally ventilated radiation shields which are oriented into the wind by vanes.

Measurements of wave and air turbulence spectra require a knowledge of any buoy motions. The buoy motion sensors used in 1968 required improvement. To improve this monitoring capability, a gyro was installed to sense pitch and roll.

Preparation schedules for the completion of the buoy system were scaled to meet the BOMEX schedule, which was the most immediate objective for the system. Buoy components and subsystems were tested at Hanford

and at FSU prior to departure for Barbados, and after arrival at Barbados. Late delivery of the fourth data acquisition system, however, precluded the desired checks of the total integrated data acquisition system.

Preparatory to BOMEX Phase I, TRITON was towed to a station at the center of the BOMEX array and anchored in 5500 m of water. It was instrumented and placed in operation for testing. The TEG was found to be fully functional and developing the necessary power. Shortly after the buoy was put in operation a propane fuel line developed a leak and by the end of Phase I, the shipboard fuel supply had been depleted. This problem, coupled with other problems related to the initial incompatibility of the added data system with the original system, limited the amount of turbulence data collected during the first period.

Attempts at turbulence measurements were to be suspended during Phase II until a larger shipboard fuel capacity could be provided. However, adverse weather led to a damaged TEG air-intake valve, which resulted in the flooding of the TEG. The high power consumption of the sonic anemometer and the gyro precluded their further operation.

During Phase III, sufficient battery power was obtained to record several hours of turbulence data with the 3-component propeller anemometer. For Phase III, the added data recording system was made independently of the others in the hope of circumventing the interaction problems that

appeared to be generated when the systems were integrated. Unfortunately, a tape monitoring capability was not available during sea operations and allowed only general system checks. With the completion of Phase III, the sea-air interaction experiment of BOMEX was complete and the turbulence system was retrieved.

Prior to Phase IV, the BNW turbulence system was converted and tested for shore operation at East Point as a part of a shore-based study of the island meteorology. Turbulence observations were obtained at a height of 16 m at four sites along a line approaching the windward beach line. The first site was beside a 16 m FSU tower supporting temperature and wind profile sensors and a vertical wind turbulence sensor. This intercomparison provided for the use of the FSU tower as a reference point while the BNW system was observing the horizontal change of turbulence characteristics approaching the beach line. Additionally, FSU was observing profiles 1 km with a tethered balloon carrying data telemetry instrumentation.

A data tape monitoring capability, though limited, was available for the island data. The results indicated that the BNW data system was operating properly. Recent "quick-look" samples also confirm the data, although a thorough analysis will be necessary for its complete validation.

Although the first test of the fully integrated TRITON system met with a number of problems, several achievements can be identified:

- 1) The successful deployment and mooring of the TRITON platform in deep ocean water (5500 m) for an extended period of time (3 months).
- 2) The operation of a prototype thermal electric generator power supply with a load capacity of 250 watts for two weeks, at which time it was stopped only because of a fuel shortage.
- 3) Ultimately, the BNW turbulence system, when deployed independently, operated properly to the extent that this can be determined from a cursory review of the data. Provisions are being made for detailed data quality assessment and data analysis.
- 4) The successful servicing of a stable platform by personnel on a scheduled basis and at times in moderately rough seas without a mishap or loss of operating time for the 3 month period.

The successful testing of a fully integrated TRITON buoy system remains to be accomplished. The results and knowledge gained during the BOMEX, and developments since that time, indicate that the initial goals are fully realizable in the near future.

System refinements and open environment tests are being planned for 1970. Specific refinements being pursued are:

- 1) Complete integration of the four parallel data systems with common timing and data sampling program control.
- 2) Improved capability for higher sampling rate to allow spectral estimates for higher frequencies of

turbulence by increasing the commutation rate to 64 words (sensors) per second, and by providing for a variable sensor sampling format such that a few sensors could be sampled at a higher rate at the expense of not sampling others for certain periods.

- 3) Minor redesign of buoy vehicle features for additional utility, serviceability, convenience, safety, and integrity.
- 4) Redesign of wave staff array for safer servicing and minimizing problems of accounting for buoy motion in wave data.
- 5) Improvement of the turbulence sensor system by using a faster responding and physically smaller humidimeter and a small unshielded, radiation-compensated, air temperature thermistor, all mounted within the frame of the sonic anemometer to improve cross-correlation accuracy.
- 6) The testing and development of a gyro stabilized platform for buoy motion sensors such that adequate compensation can be made to data sensitive to buoy motion.

Integrated tests of the total TRITON system, excluding the buoy sections acting only as flotation spaces, will be made on the bench and in the environment for an extended time at a shore facility. Subsequent tests of the total buoy system at sea for at least 1 month are anticipated hopefully at a site where reference measurements can be made from a fixed platform.

BACK TRAJECTORIES FROM THE BOMEX ARRAY

W. E. Davis

A series of back trajectories for selected levels was used to find maximum latitude displacement of air parcels sampled over the BOMEX array. The displacement compared with ^{95}Zr measurements over the array indicated high values correlated with displacements from the mid-latitudes of the Northern Hemisphere while low values were correlated with low latitudes or the Southern Hemispheric air. A series of back trajectories from sampling positions around the array indicated in one case the contact of parcels coming from the mid-latitudes with air parcels possibly originating in the Southern Hemisphere.

Aircraft measurements of radionuclides were taken over the BOMEX array between elevations of 1,000 to 60,000 ft during the first and third experimental periods. The measurements were made in order to describe the spatial and temporal variations of the concentration distributions of radionuclides throughout the atmospheric volume over the array. These concentrations and their variations were pertinent to the removal by rain and the eventual dry deposition at the air-sea interface. To understand why changes occurred in the concentrations of the radionuclides, a series of back trajectories was started. It was hoped that these trajectories would indicate short term latitudinal sources for the radionuclides and, hence, provide some explanation of changes in observed concentration.

A rather gross approach was used in that the trajectories were calculated on constant pressure surfaces in areas where data, in some in-

stances, were lacking. In areas away from the equator, winds were estimated through the use of a gradient wind assumption. The temperature of the air parcel was noted in order to determine if any large scale vertical motions had affected the path of parcels.

In cooperation with BNW radiochemists, ^{95}Zr was selected as a tracer in this study, since ^{95}Zr has a maximum in the mid-latitudes of the northern hemisphere. Hence, mid-latitudinal northern hemispheric air would tend to have higher values of ^{95}Zr than low latitudinal or southern hemispheric air. These air concentrations of ^{95}Zr were measured over the array for Periods 1 and 3. The mean concentrations over the array are shown in Figure 1. It is quite evident from the plot that a change in variability as well as mean concentration value occurred between the first and third period.

Trajectories were taken on the 200, 300, 500 and 700 mb levels,

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FIGURE 1. Plot of Mean Air Concentrations of ^{95}Zr over BOMEX Array for First and Third Periods for 20, 30 and 40 Thousand Feet

which have been assumed to be associated with the 40,000, 30,000, 20,000, and 10,000 ft aircraft samplings, respectively, for both the first and third period.

The results of back trajectories at 40,000 ft originating at 15°N 57°W longitude, indicated as an air mass source the latitudinal range between the equator and 30°N in the

first period. For the most part, detailed concentrations variations within the period were not explained except for the lowest mean concentration value, observed on May 7, 1969. A back trajectory from May 7 to 12 indicated a possible southern hemispheric source with the trajectory being dropped at the equator due to lack of data.

First and second period maximum latitudinal displacements from 15° N are shown in Figure 2. The bars

indicate a range of maximum latitudinal source. In general, the higher the latitudinal source the higher the concentration. Also, the higher degree of variability can be explained by the greater latitudinal range of sources for the air over the BOMEX array.

The same approach was taken for the 30,000 ft samples. The results indicated in Figure 3 are similar to those of Figure 2.

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FIGURE 2. Plot of Maximum Displacements from 15° N as Indicated from Back Trajectories Versus Levels of Mean Concentrations over the Arrays for 40,000 Feet

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FIGURE 3. Plot of Maximum Displacements from 15° N as Indicated from Back Trajectories Versus Levels of Mean Concentrations over the Arrays for 30,000 Feet

In the first period very low values of ^{95}Zr ($<50 \text{ dpm}/10^3 \text{ m}^3$) on the 9th, 10th and 11th of May were identified with trajectories traced back near the equator. Also, the higher latitudinal sources indicate high values for ^{95}Zr . The same approach was used in estimating the 20,000 ft trajectories using the 500 mb chart with less definitive results, but in general agreement with observations of higher concentration at higher latitudes found for other levels (see Figure 4).

Trajectories taken on the 700 mb surface were less successful in explaining variations observed in the ^{95}Zr concentrations. Generally, tra-

jectories during the third period indicated a latitudinal range from 10 to 30° N for their sources. No equatorial or southern hemispheric sources were noted for these trajectories. Further analysis may reveal more information on the variations in concentration. However, the latitudinal gradient of the concentrations at these elevations is considerably less than at higher altitudes. This diminishes the possibility of explaining variations at altitudes at and below 10,000 ft. Because of the lessening of the gradient as well as the tendency of the

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FIGURE 4. Plot of Maximum Displacements from 15° N as Indicated from Back Trajectories Versus Levels of Mean Concentrations over the Arrays for 20,000 Feet

back trajectory to move 'from areas of low data at altitudes less than 10,000 ft, back trajectories were not traced below 10,000 ft.

In the third measurement period for one set of aircraft measurements at 30,000 ft on June 30, 1969, a comparison of the four sampling areas around the BOMEX array was of particular interest. The two southern samples were less than $200 \text{ dpm}/10^3 \text{ m}^3$ while the northern samples were an order of magnitude higher. Back trajectories were initiated for the center of each of the four sampling points. The northern trajectories were traced back to a latitudinal range of 19 to 35° N while both of the southern points were traced back to the equator. The reason these trajectories were dropped at the equator, as noted previously, was the lack of data. Even with the data limitations, the series of trajectories indicate some of the complexity in air mass motions over the BOMEX array, with possible southern hemispheric air being brought together with mid-latitudinal northern hemispheric air.

The use of radionuclide analysis for studies of atmospheric transport for short-term trajectory has indicated a considerable degree of success. Since these trajectories were of only short term (less than 1 week) and secondly because of inherent errors in the meteorological analysis, the success of these trajectories exceeded expectations. This statement is made since origins of the back trajectories in the low latitudes could have their roots in the

higher latitudes. But the short term over which the trajectories were taken or the inadequacy of the meteorological data may prevent determination of origin for the concentrations observed. Further work using other radionuclides may give added confirmation to the indicated sources of the air through the back trajectory analysis and thus reveal more on the reliability of these short-term trajectories in explaining the concentration variations over the BOMEX array.

The findings may be summarized as in the following.

A series of back trajectories from the BOMEX array was found to explain some of the variability observed in mean aircraft measurements of ^{95}Zr . During the first period, low variability along with low mean concentration values were correlated with low latitudinal sources from the short-term trajectories. During the third period, high latitudinal sources of air mass were correlated with high concentration values of ^{95}Zr , which, in turn, explained the high variability during this period.

Southern hemispheric sources of air were indicated for at least 3 days at 30,000 ft and for 1 day at 40,000 ft during the first period.

A series of back trajectories for the 30,000 ft level from four measuring positions around the BOMEX array on June 30, 1969, indicated the possibility of an air mass from the southern hemisphere coming into contact with mid-latitudinal northern hemisphere air over the BOMEX array.

PRELIMINARY RESULTS FROM RADIONUCLIDE MEASUREMENTS MADE DURING BOMEX*

J. A. Young, H. G. Rieck, Jr., D. E. Robertson, and W. B. Silker**

During the experimental phase of the Barbados Oceanographic and Meteorological Experiment, the concentrations of ^7Be and several nuclear-weapons-produced radionuclides were measured in air, seawater, and rain samples. Because of its relatively short half-life, ^7Be is confined primarily to the top 50 m of the ocean. Observed changes were not due to rain since the largest amount of ^7Be contributed by any one rainfall was less than 1% of the total amount in the sea. During the experimental periods, the rate of addition of ^7Be to the sea by rainfall was only 10 to 15% of its rate of decay in the sea. It does not seem likely that turbulent transport across the sea surface could cause rapid changes in concentrations which generally occurred simultaneously throughout the top 30 m. The decreases in concentration throughout some vertical profiles could not have been caused by vertical transport; therefore, the changes in concentration were probably due to the horizontal transport of water of different activities through the array. Fresh water was present during the second two weeks of June. The high activities observed during

this period were probably caused by Amazon Basin water since it is produced primarily by rain, with concentrations averaging approximately 1000-fold that of seawater.

Although the activities of ^7Be and the nuclear-weapons-produced radionuclides in the atmosphere rapidly increased above 9.1 km, they remained fairly constant from 0.3 to 9.1 km. Rather large changes in concentrations with time and location were observed. On the average there was a minimum in the activities of all of the radionuclides at 1.5 km, probably due to rainout. At a specific altitude, some vertical profiles showed activity minimums for the bomb-produced radionuclides but not for ^7Be . These minimums probably resulted from intrusions of southern hemispheric air since the activities of the bomb-produced radionuclides are considerably lower in the southern hemisphere than in the northern, while the ^7Be activities are about the same. Some profiles showed large activity minimums at 9 km which were not caused solely by intrusions of southern hemispheric air since ^7Be showed the same activity minimum as the bomb-produced radionuclides. These minimums were probably due to rainout. Some of the air samples collected near the sea surface showed increasing radionuclide activities from 0.3 to 21 m.

* This report appears also in
BNWL-1307 Part 2, p. 21.

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FALLOUT STUDIES

Fallout studies are concerned with describing the behavior of radioactive debris and industrial pollutants as they are transported over large distances from release points, diffused, and subsequently delivered to the surface. Models of transport and diffusion must be integrated with removal models describing delivery to the boundary layer where materials become involved in dry deposition processes or delivery to cloud systems where wet removal and rainout to the surface can occur.

In various environs, the movement of toxic materials can be adequately described with the aid of adiabatic trajectories. More frequently, however, energy imbalances in the earth-atmosphere system restrict the effectiveness of adiabatic trajectories and require more sophisticated analysis techniques. Diabatic conditions such as those produced by precipitation, surface heating, terrain effects, and radiational cooling are presently being evaluated in order to predict the behavior of atmospheric debris as it is transported through these diabatic zones by the prevailing winds.

RAFLUX - A NUMERICAL MODEL FOR CALCULATING THE LONG WAVE
RADIATIVE TRANSFER IN THE TROPOSPHERE

B. C. Scott

The neglect of radiative cooling in the construction of energy conserving isentropic trajectories can produce serious errors in calculated trajectory end points. A numerical model has therefore been developed to utilize reported pressure, temperature, and moisture profiles for radiative cooling calculations. The theory necessary for the development of the numerical model is briefly discussed, and results of radiation calculations are presented. All calculated radiative flux profiles are in close agreement with those of existing models, thereby suggesting that the assumptions made in the present numerical model are neither too restrictive nor unrealistic.

The long wave radiative flux divergence in the atmosphere is the result of varying degrees of absorption and emittance by the earth's surface, clouds, and the atmosphere. Generally, the net effect is to produce cooling rates on the order of 1 to 2 °C/day in the troposphere. Hence, the radiative transfer of heat at times becomes an important diabatic effect which if neglected can produce serious errors in the construction of energy-conserving isentropic trajectories.

The procedure for including this diabatic effect in trajectory computations has been discussed in detail by Davis and Scott,⁽¹⁾ but the calculation of the magnitude of the diabatic effect has been neglected primarily because past estimates involved lengthy and tedious operations with existing tables and charts. If radiative flux divergence is to be computed at several grid points and

included in isentropic trajectory computations, a rapid and accurate procedure is necessary. Thus, RAFLUX, a numerical model for calculating the long wave radiative transfer in the atmosphere, has been developed as a subroutine to the existing Montgomery stream function program. RAFLUX will utilize the observed pressure, temperature, and moisture profiles at radiosonde sites for calculating the net cooling rate in selected regions.

A detailed description of the theory and assumptions utilized in constructing radiation charts and models can be found in papers by Elsasser and Culbertson,⁽²⁾ Yamamoto,⁽³⁾ and Sasamori.⁽⁴⁾ In this report, only the fundamental aspects of the theory are presented.

Essentially, the radiative flux through a unit horizontal surface can be defined in terms of a mean absorptivity R

$$R(u_i, T) = \int_0^{\infty} [1 - \tau_f(u_i)] \frac{dB_\gamma}{dT} d\gamma, \quad (1)$$

where u_i is the effective optical path length of CO_2 , O_3 , or H_2O ; T is the temperature; B_γ is Planck's radiation function expressed in flux at frequency γ ; and τ_f represents the mean transmissivity in a small frequency interval where B_γ is nearly constant.

The summation of R over the radiatively significant part of the atmosphere determines the net flux through a level z . In a cloudless atmosphere, the individual upward and downward components of the net flux are described by

$$F_{\downarrow}(z) = \sum_i \int_0^{T(z)} R\{u_i(T') - u_i[T(z)], T'\} dT', \quad (2a)$$

and

$$F_{\uparrow}(z) = \sigma T^4(o) + \sum_i \int_{T(1)}^{T(z)} R\{u_i[T(z)] - u_i(T'), T'\} dT'. \quad (2b)$$

The summation sign indicates that the integration is carried out for each absorber; i.e., H_2O , CO_2 , and O_3^* . The additional notation, $\sigma T^4(o)$,

is the well known expression for the blackbody flux at the surface. Here a has been assigned the value $8.14 \times 10^{-11} \text{ cal cm}^{-2} (\text{°K})^{-4} \text{ min}^{-1}$.

In those portions of the atmosphere where CO_2 and H_2O absorb in the same spectral region, the net absorption is reduced. Following the procedures outlined by Martin and Tupaz,⁽⁵⁾ it can be shown that the actual net flux through a level z is equal to

$$F = F_w + \int_{T=0}^{T(z)} \tau_f(u_{\text{H}_2\text{O}}) \int_{540 \text{ cm}^{-1}}^{820 \text{ cm}^{-1}} \left[1 - \tau_f(u_{\text{CO}_2}) \right] \frac{dB_\gamma}{dT} d\gamma dT. \quad (3)$$

The term F_w represents the net flux contribution from water vapor. Equation 3 states that the net flux is the sum of the water vapor contribution and the corrected CO_2 contribution.

In RAFLUX, the last term in Equation 3 has been approximated by

$$\begin{aligned} & \int_{T=0}^{T(z)} \tau_f(u_{\text{H}_2\text{O}}) \int_{540 \text{ cm}^{-1}}^{820 \text{ cm}^{-1}} \left[1 - \tau_f(u_{\text{CO}_2}) \right] \frac{dB_\gamma}{dT} d\gamma dT \\ &= \int_{T=0}^{T(z)} \tau_f(u_{\text{H}_2\text{O}}) R(u_{\text{CO}_2}, T) dT \end{aligned} \quad (4)$$

* The flux contribution by ozone in the troposphere is generally quite small and has been neglected in the present numerical model.

The magnitude of the error introduced by the approximation of Equation 4 is unknown at the present time, but comparisons with total flux calculations obtained from other numerical models indicate that the error is small.

The transmissivity function, $\tau_f(u_{H_2O})$, is obtained from Sasamori,⁽⁴⁾ and applies in the overlap region (540 to 820 cm^{-1}). As the optical path length of water decreases to zero, τ_f becomes larger and approaches the value one. Thus Equation 4 simply states that in regions where the total u_{H_2O} is large (τ is small), the CO_2 flux contribution is reduced by a large amount. Conversely, in the upper reaches of the atmosphere where the total u_{H_2O} is small ($\tau \approx 1.0$), the CO_2 flux is correct as calculated.

In RAFLUX, Equation 2 is expanded in order to use the empirical expressions of the absorption functions derived by Sasamori.⁽⁴⁾ Hence,

$$F(z)_i = \tau_i \left[\int_{T(\text{constant } u_i)}^{T(z)} R_i(u_i, T) dT + \int_{T=0}^{T(\text{constant } u_i)} R_i(u_i, T) dT \right] \quad (5)$$

where the subscripts represent evaluation with respect to the water and CO_2 expressions; τ_f equals $\tau_f(u_{H_2O})$ when dealing with CO_2 and 1.0 when dealing with water contributions.

In the numerical model, the temperature and the mixing ratio of water are held constant above 16 km.

The volume percent of CO_2 throughout the atmosphere is assumed to be constant with the surface partial pressure equal to 0.32 mb.

The second integral in Equation 5 is analogous to the off-chart area described by Elsasser and Culbertson.⁽²⁾ Hence, following Elsasser, the temperature where u_{CO_2} becomes constant is taken as 193°A. The temperature where u_{H_2O} becomes constant is calculated directly by the model.

The capability of calculating the radiative flux divergence in a cloudy atmosphere is presently being added. The procedure being used has been described by Manabe and Strickler⁽⁶⁾ and has the following characteristics:

- 1) The upper and lower surfaces of a cloud deck are assumed to radiate as a blackbody surface. Cirrus decks are treated as exceptions and are assumed to radiate at one-half the blackbody radiation.
- 2) The amount of sky covered by the overlapping of cloud layers is equal to the product of the cloudiness of each layer.

For example, the upflux through level z in Figure 1 is equal to the reduced upflux (corrected for overlap and reduced by cloud layers below level z) from the atmosphere ($\vec{F}_g$), plus the contribution from each cloud layer below level z ($\vec{F}_1, \vec{F}_2, \vec{F}_3$).

Upward, downward, and net flux calculations from RAFLUX in a cloudless atmosphere have been compared with the calculated results of several existing radiation models.

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FIGURE 1. The Upward Flux Component Through Level Z. In the figure C_1 is the cloud cover of the first cloud layer; C_2 is the cloud cover of the second cloud layer; f_1 is the blackbody flux from the top of the first cloud layer; f_2 is the blackbody flux from the top of the second cloud layer; and f_g is the total upflux from the atmosphere.

Among the most recent and detailed models are those developed by Rodgers and Walshaw⁽⁷⁾ and Manabe-Strickler.⁽⁶⁾ The comparison of RAFLUX with these two models is shown in Figure 2. The agreement is good in the troposphere. The slight deviation from the net flux curve by RAFLUX at pressures less than 600 mb is the result of an underestimate of the downflux at these altitudes. The neglect of O_3 is the most likely reason for this underestimate. However, the error is slight and amounts to a 0.1 to 0.2 °C/day overestimate in the cooling rates.

Table 1 compares observed down-flux contributions with those calculated by Sasamori,⁽⁴⁾ RAFLUX, and the atmospheric radiation tables (ART) from Elsasser and Culbertson.⁽²⁾ Figure 3 compares RAFLUX with results computed by London⁽⁹⁾ and Yamamoto.⁽³⁾ In Figure 3, the slope of the curves is the important feature, for in all radiation models, the cooling rates are proportional to the change of net flux with altitude. In all cases, the agreement with existing models is extremely good in the lowest 8 km of the troposphere.

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FIGURE 2. Net Infrared Flux in the Troposphere for March, Northern Hemisphere, Clear Skies. The temperature, moisture data used have been compiled by London. (9) The calculations for the London and Yamamoto models are from Yamamoto. (3)

TABLE 1. Comparison of Observed and Calculated Downflux Contributions

Time and Date (Local Times) 1953-54	Observed Downward Flux, $\text{cal cm}^{-2} \text{min}^{-1}$	Calculated Downward Flux		
		RAFLUX	Sasamori	ART
Sept 27 0000h	0.454	0.472	0.469	0.397
Oct 27 0012h	0.378	0.381	0.387	0.325
Oct 28 0000h	0.395	0.404	0.399	0.333
Nov 4 0025h	0.375	0.379	0.311	0.368
Dec 1 0000h	0.382	0.392	0.390	0.326
Dec 2 0040h	0.407	0.421	0.408	0.343
Dec 13 0000h	0.343	0.350	0.342	0.293
Dec 24 0000h	0.316	0.322	0.316	0.268
Jan 7 0000h	0.314	0.320	0.334	0.283
Jan 9 0000h	0.334	0.333	0.333	0.281
Jan 19 0000h	0.322	0.337	0.324	0.275
Feb 2 0010h	0.290	0.303	0.302	0.261
Mar 18 0000h	0.301	0.323	0.301	0.259

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FIGURE 3. Comparison of Upward, Downward, and Net Long Wave Radiation Flux as Calculated by the Manabe-Strickler (M-S), Rodgers-Walshaw (R-W)₂, and RAFLUX Models. The calculations for the (M-S) and (R-W)₂ models are from Stone and Manabe.(8)

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PRECIPITATION SCAVENGING STUDIES

The objectives of the precipitation scavenging studies are to determine experimentally the magnitude of the effects, and to understand the mechanism and develop predictive capabilities for the removal of pollutants from the atmosphere by precipitation. The scope of this work extends to all types of pollutants, including particulates and gases of both natural and anthropogenic origin, and to all types of precipitation. To accomplish the objectives, extensive field and laboratory experiments, and intensive theoretical analyses have been performed. A brief description follows of some of our accomplishments during 1969, for both the in-cloud and below-cloud studies. Further details are given in the following reports and in the more complete articles which are referenced therein.

Below-Cloud Scavenging

- In the report by Slinn and Hales, it is shown that thermophoresis can be a significant mechanism for the rain-savenging of aerosol particles of radii between 0.01 and 1.0 μ .
- Dana, Wolf and du Plessis report on experimental measurements of the rain-savenging of rhodamine dye particles and SO₂ gas. Particular attention has been given to the influence of dry deposition on the measurements.
- It is shown that the previously reported anisotropy of Brownian diffusion does not significantly influence the scavenging of sub-micron particles. In a companion paper by Slinn, Brownian diffusion in free-molecule Couette flow is described.
- Characteristics of Olympic Peninsula rain, including raindrop-size spectra and electric charges, are described in a report by Dana.
- Other work in progress in this study includes the development by du Plessis of improved methods for generating and detecting particulate tracers.

In-Cloud Scavenging

- Some rather startling and perhaps controversial results concerned with phoretic processes within non- and subfreezing clouds are described in the second report by Slinn and Hales.
- Scott's contribution illustrates a part of our response to the realization that cloud-dynamics form an integral part of the in-cloud scavenging problem.
- Perkins, Thomas, Young and Rieck present an interpretation of their recently reported measurements of the in-cloud scavenging of cosmogenic radionuclides.
- In the report by Davis, a model is presented to describe the in-cloud scavenging of natural aerosol particles which are tagged with the cosmic-ray-produced radionuclide, ²⁴Na.
- Presented in the review by Slinn is a new and complete theory of the molecular diffusion of small aerosol particles which can diffusely reflect the gas molecules.

PHORETIC AND BROWNIAN DIFFUSION PROCESSES IN
THE BELOW-CLOUD SCAVENGING OF AEROSOL PARTICLES BY RAIN*

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

An analysis is made of the relative importance of thermophoresis, Brownian diffusion and diffusiophoresis in the below-cloud rain scavenging of aerosol particles. It is shown that for particles of radii less than about 0.01 μ , Brownian diffusion is most important. For particles of radii between 0.01 and 1.0 μ , thermophoresis can be dominant. A graph of the resultant washout coefficient is presented which includes an estimate of the enhancement of the scavenging by thermophoretic effects.

Many physical processes must be considered when an attempt is made to analyze the below-cloud scavenging of aerosol particles by rain. For example, it is known that the inertia of the particle is of dominant importance for particles of radii larger than about 1 micron (μ), whereas for particles smaller than 0.01 μ , Brownian diffusion is significant. For all particle sizes, electrical interactions can be important if the electrical charges are large enough but unfortunately these are still not known with sufficient accuracy to complete the estimate of electrical influences in scavenging.⁽¹⁾ In this report a review will be made of our recent assessment of the relative importance of thermophoresis, Brownian diffusion and diffusiophoresis

The objectives of precipitation scavenging studies are to understand the mechanisms involved in, to develop the capabilities to predict and, not of least importance, to measure, the removal by precipitation of pollutants from the atmosphere. To this end, we seek the washout coefficient, A .

If the precipitating element is not saturated with the pollutant, then the reduction, dN_p , in the number of particles per unit volume, N_p , during a time interval dt should be proportional to the time interval and the number of particles which are present:

$$dN_p = -A N_p dt. \quad (1)$$

In general, the proportionality factor, A , (the washout coefficient) depends on the nature of both the pollutant and the precipitation. That A can depend also on N_p , especially in the scavenging of

* This report is based on a portion of an article by the authors which has been submitted for publication in the Proceedings of the 1970 Precipitation Scavenging Meeting.

gases, is a subject of current investigations. This effect should not be of major importance in the scavenging of particles. From Equation 1 we obtain that if the initial density of particles is N_{po} , then

$$N_p = N_{po} \exp (-At) \quad (2)$$

describes the density at a later time, t .

The phenomena of diffusio- and thermophoresis (phoresis = force) are some influences on the motion of aerosol particles that result from nonuniformities in the host gas. Recent reviews are given in References (2) and (3). As a result of temperature nonuniformities in the host gas, the particles drift in the direction of the heat flux. As a result of concentration gradients of components of the host environment, the particles may drift in the direction of one of the diffusing species. In fact, Brownian diffusion also depends on nonuniformities in the host gas⁽⁴⁾ but the resultant anisotropy of the diffusion is insignificant in the scavenging problems.⁽⁵⁾

Thermo- and diffusiophoresis might be expected to be important in the below-cloud scavenging problem since usually there is heat flux to and evaporation from the falling precipitation element. However, the role of these phoretic effects in below-cloud scavenging has apparently not been studied previously. The role of diffusiophoresis in the in-cloud problem was discussed by Facy⁽⁶⁾ and studied more recently

by Goldsmith et al.⁽⁷⁾ and by Vittori and Prodi.⁽⁸⁾ A number of Soviet scientists (e.g., Ziman,⁽⁹⁾ Borovikov et al.⁽¹⁰⁾) have commented on the lack of quantitative estimates of the phoretic influences on both in- and below-cloud processes.

The portion of the total flux of particles to a falling raindrop, which arises from Brownian diffusion, can be estimated from the semiempirical data for the heat transfer to an isothermal sphere.^(1,5) The magnitude of this number flux is given by

$$\Gamma_B = \frac{D_p N_p}{R} (1 + \beta \text{Re}^{1/2} \text{Sc}^{1/3}) \quad (3)$$

where D_p is the diffusivity of the particles; R is the radius of the raindrop; β is a numerical factor⁽¹⁾ near 0.4; $\text{Re} = VR/\nu$ is the Reynolds number for the raindrop falling at velocity V through air of kinematic viscosity, ν ; and $\text{Sc} = \nu/D_p$ is the Schmidt number for the particles. Equation 3 adequately describes the Brownian diffusional flux for particles of arbitrary size⁽⁵⁾ but it is to be emphasized that for particles larger than about 1μ , inertial contributions to the flux must be incorporated into the analysis.

The thermophoretic, number flux of particles, Γ_{tho} , is in the direction of the heat flux, $\vec{q}$. For an evaporating raindrop, the heat flux is toward the drop and its magnitude is given by⁽¹⁾

$$q = \frac{k(T_a - T_d)}{R} (1 + \beta \text{Re}^{1/2} \text{Pr}^{1/3}) \quad (4)$$

where $(T_a - T_d)$ is the temperature difference between the air and the drop, and the air is characterized by its: Prandtl number, $Pr = \nu/\alpha$; thermal diffusivity, $a = k/\rho c_p$, thermal conductivity, k ; density, ρ and specific heat at constant pressure, c_p .

In terms of the heat flux, the thermophoretic flux of aerosol particles is given by (2,11)

$$\vec{\Gamma}_{tho} = N_p f(\lambda, a, k, k_p) \vec{q}/p \quad (5)$$

where p is the pressure of the gas and where f is a function of the Knudsen number, K , of the particle (the ratio of the mean free path of the gas, A , to the radius of the particle, a) and of the thermal conductivities (k, k_p) of the gas and particle. Specifically, Brock's theory gives (11)

$$f = \frac{2}{5} \frac{[1 + AK + BK \exp(-C/K)] (k + C_t K k_p)}{(1 + 3C_m K) (2k + k_p + 2C_t K k_p)} \quad (6)$$

Equation 6 is plotted in Figure 7 of Reference 2. There, the values used for the constants are: $A = 1.45$, $B = 0.40$, $C = 1.0$, $C_t = 2.5$, $C_m = 1.0$. In the range of interest

of the parameters, the values of f are between 0.16 and 0.01.

The diffusiophoretic, number flux of particles, $\vec{\Gamma}_{do}$, which is the direction of the number flux of water molecules, $\vec{\Gamma}_w$ is given by (2,3)

$$\vec{\Gamma}_{do} = N_p g(a, \lambda) \left\{ \frac{n_w(m_w)^{1/2}}{n_w(m_w)^{1/2} + n_a(m_a)^{1/2}} \right\} \frac{\vec{\Gamma}_w}{n_w} \quad (7)$$

where n and m are number densities and masses of the molecules of the specified species (water and "air") and g is a weak function of the Knudsen number of the particles. Its numerical value is not known

well but based upon the experimental data of Figure 9 of Reference 2, we will take

$$1.0 > g > 0.85 \quad (8)$$

As will be seen below, determining g more accurately would be of fundamental importance to our results.

For applications to the scavenging problem, Equation 7 can be simplified. Since the mass density of water vapor in a saturated system at between 10 to 20 °C is only, approximately, 10^{-5} g/cm³ whereas the density of "air" molecules is approximately 10^{-3} g/cm³, then the term $n_w(m_w)^{1/2}$ in the denominator of Equation 7 can be ignored. Further, obviously there is a relation between the flux of water from, and the flux of heat to, the raindrop. When the temperature of the raindrop has reached a steady value, close to the wet-bulb temperature,⁽¹²⁾ then the role of the heat flux is solely to evaporate water from the drop. Since $L = 600$ calories evaporates one gram of water, then a heat flow of $4\pi R^2 q$ calories per second evaporates $(4\pi R^2 q/L)$ grams of water per second. Thus $\Gamma_w = q/(Lm_w)$. Consequently, for the scavenging problem, the diffusiophoretic number flux can be written as

$$\vec{\Gamma}_{do} = N_p g \frac{(m_w/m_a)^{1/2}}{L m_w n_a} (-\vec{q}). \quad (9)$$

The ratio of Equations 5 and 9 enables us to determine the relative importance of the two counteracting fluxes, arising from thermo- and diffusiophoresis:

$$\frac{\Gamma_{tho}}{\Gamma_{do}} = \frac{f}{g} \left(\frac{m_a}{m_w} \right)^{1/2} \frac{L m_w}{b T_a} \quad (10)$$

in which b is Boltzmann's constant. Notice that not only is this ratio, Equation 10, independent of the size of the raindrop, it is also independent of the relative humidity of the air. Using the values of f and g which are given in Figures 7 and 9 of Reference 2, Equation 10 has been plotted in Figure 1. From this figure it is seen that, regardless of the thermal conductivity of the particle, thermophoresis dominates diffusiophoresis for particles of radii less than about 1μ .

From Equations 3 and 9, we can obtain an estimate of the relative importance of Brownian diffusion and diffusiophoresis. Taking the ratio of these two equations results in

$$\frac{\Gamma_{do}}{\Gamma_B} = g \frac{m_w/m_a^{1/2}}{L m_w n_a} \frac{k(T_a - T_d)}{D_p} \frac{(1 + \beta Re^{1/2} Pr^{1/3})}{(1 + \beta Re^{1/2} Sc^{1/3})} \quad (11)$$

Since the convective terms in the fluxes ($\beta Re^{1/2} Pr^{1/3}$, etc.) usually dominate in these equations for the fluxes, then Equation 11 can be simplified to

$$\frac{\Gamma_{do}}{\Gamma_B} = g \left(\frac{m_w}{m_a} \right) \frac{\gamma}{\gamma - 1} \frac{b(T_a - T_d)}{L m_w} \left(\frac{\alpha}{D_p} \right)^{2/3}$$

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FIGURE 1. Ratio of Thermo- to Diffusiophoretic Fluxes According to Equation 10 Using Data of Reference 2

where $\gamma = c_p/c_v$ is the usual ratio of specific heats. Substituting into Equation 12 the numerical values for the parameters, it is found that diffusiophoresis dominates Brownian diffusion for particles of radii satisfying

$$a > 0.013 (\Delta T)^{-3/4} (\mu) \quad (13)$$

where ΔT is the temperature difference between the air and the raindrop, measured in $^{\circ}\text{C}$. In turn, since thermophoresis dominates diffusiophoresis for all particles of radii less than 1μ , we see that, unless the relative humidity of the air is very close to 100%, then thermophoretic effects result in an enhancement of the below-cloud, rain sca-

vening of particles with radii between (approximately) 0.01 and 1μ .

Using these results, we can revise our previous estimate⁽¹⁾ of the wash-out coefficient. From Equation 5 and the ratio, Equation 10, the phoretic contribution to the flux of particles to the raindrop is found to be

$$\Gamma_{ph} = N_p (1 - \kappa) f \frac{\alpha}{R} \frac{\gamma}{\gamma - 1} \left(\frac{T_a - T_d}{T_a} \right) (1 + g \text{Re}^{1/2} \text{Pr}^{1/3}) \quad (14)$$

where κ is the inverse of the ratio given by Equation 10. The collection efficiency, E_c , is obtained from the flux by nondimensionalizing the total flow of particles to the raindrop by the flow in the undisturbed fluid to the cross-sectional area of the drop, πR^2 . Thus

$$E_c = \frac{4(1 - \kappa) f}{Re Pr} \left(\frac{\gamma}{\gamma - 1} \right) \left(\frac{T_a - T_d}{T_a} \right) \left(1 + \beta Re^{1/2} Pr^{1/3} \right). \quad (15)$$

To obtain an estimate of the washout coefficient, we propose to use, for convenience, the formula given by Ziman⁽⁹⁾ for the raindrop size

distribution and raindrop terminal velocity:

$$N(R) dR = \frac{10^{-4} J_o}{6\pi R_m^7} R^2 \exp \left(\frac{-2R}{R_m} \right) dR, \text{ cm}^{-3} \quad (16)$$

and

$$V = 8000 R \text{ sec}^{-1} \quad (17)$$

where J_o is the rainfall intensity in cm/sec and R_m is the radius of the drops in the maximum of the spectrum. Since the number of aerosol particles collected by the drop during dt is $4\pi R^2 \Gamma dt$, and since the number of drops per unit volume that have radius R is given by Equation 16, then, using the definition of the washout coefficient as given by Equation 1, we get for the phoretic contribution to Λ the result

$$\Lambda_{ph} = \frac{(1 - \kappa) f}{4} \frac{\gamma}{\gamma - 1} \left(\frac{T_a - T_d}{T_a} \right) \frac{10^{-4} J_o}{R_m^3} \left[1 + 2\beta Re^{1/2} Pr^{1/3} \right] \quad (18)$$

where $Re = 8000 R_m^2 / \nu$. If J_o is measured in cm/hr, then Λ is given in hr^{-1} . Equation 18, our previous estimate of the effects of Brownian diffusion, and contributions from inertial effects⁽⁹⁾ are plotted in Figure 2.

It is seen in Figure 2 that the thermophoretic flux of particles to an evaporating raindrop can significantly increase the washout coefficient for aerosol particles in the size range 0.01 to 1 μ . Such parti-

cles are described by Ziman⁽⁹⁾ as "illusive." Although diffusio-phoresis deters the below-cloud scavenging of particles by an evaporating, precipitation element, the large value of the heat of vaporization of water and the resultant thermophoretic effect overwhelms diffusio-phoresis. In the apt phraseology of Professor Dingle, it might be said that thermophoresis helps to "bridge the Greenfield gap."⁽¹³⁾

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FIGURE 2. The Washout Coefficient as a Function of Particle Size, Resulting from Brownian Diffusion, Thermophoresis and Inertial Effects

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FIELD EXPERIMENTS IN PRECIPITATION SCAVENGING*

M. T. Dana, M. A. Wolf, and L. A. duPlessis

Field experiments conducted at Quillayute Air Base on the Olympic Peninsula utilized soluble rhodamine dye particles and sulfur dioxide gas as tracers. Consistency was found in the dry deposition characteristics of the dye particles though exact deposition velocities could not be assigned. In one experiment, the experimental washout coefficient agreed well with that computed from raindrop size spectra and Mason's collection efficiencies. Experimental washout coefficients for sulfur dioxide generally fell below theoretical estimates.

Precipitation scavenging field experiments utilizing soluble rhodamine dye particles, atomized in methanol solution, were conducted at the field site at Quillayute Air Base on the Olympic Peninsula. As reported previously,⁽¹⁾ modifications were made to the tracer dispenser which is a mechanical insecticide atomizer. The purposes of these modifications were to reduce the undesirable effects of delayed evaporation of the methanol solvent and to prevent intake of ambient water into the air-supply blower.

Examinations of previous experiments⁽¹⁾ and additional experimental work have shown that the delayed evaporation effects can be significant. In particular, high washout coefficients were obtained at high liquid flow rates. These agree with

washout coefficients for the scavenging of liquid droplets which are considerably larger than the desired particles.

Experimental details are shown in Table 1 for two successful experiments in which the liquid feed-rate was held to low values. The particle size was determined from a previously obtained calibration curve of particle size versus solution concentration. Multiple precipitation collector arcs were used at distances of 50, 100 and 150 ft from the central source point which is a 26 ft portable tower.

In Table 2 are indicated the experimentally determined "apparent" washout coefficients. These include contributions from both wet and dry removal. The increase in the apparent washout with distance from the source demonstrates that dry-deposition contributes to the amount of recovered mass. If an estimate of expected wet deposition is made, then a dry deposition velocity can

* The work on sulfur dioxide scavenging was supported in part by Battelle Memorial Institute

TABLE 1. Soluble Dye Experimental Details

Experiment	Mass Median Particle Diameter, μ	Mass of Tracer Released, g	Wind Speed, cm sec^{-1}	Rainfall Intensity, mm hr^{-1}
UIL-20	2.1	0.83	404	0.8
UIL-22	4.4	4.34	385	1.3

TABLE 2. Soluble Dye Washout Coefficients and Deposition Velocities

Experiment	$\Lambda_T^{(a)}$ sec^{-1}	Arc $^{(b)}$	$\Lambda_A^{(c)}$ sec^{-1}	$V_d^{(d)}$ cm sec^{-1}
UIL-20	---	A	1.2×10^{-4}	---
		B	2.4×10^{-4}	6.4
		C	5.2×10^{-4}	5.4
UIL-22	2.0×10^{-4}	A	2.5×10^{-4}	---
		B	3.6×10^{-4}	5.0
		C	4.4×10^{-4}	6.8

a. Calculated from Equation 2 using experiment raindrop size spectra and the collection efficiencies of Mason⁽³⁾ for 4 μ diameter particles.

b. Arc A - 50 ft from source; B - 100 ft; C - 150 ft.

c. Apparent washout coefficient including dry deposition, if any.

d. Calculated from Equation 3 with $\Lambda_w = \Lambda_A$ for Arc A.

be calculated using measurements of tracer mass, sampled by filters located on the array at the height of the precipitation collectors. This deposition velocity is given by

$$V_d = \frac{F}{a} \frac{m_s}{m_f} \left[1 - \frac{\Lambda_w}{\Lambda_A} \right], \quad (1)$$

where F is the air sampling flow rate of the filters; a is the area of a precipitation collector; m_s is the sum of the tracer masses recovered from the precipitation collectors which are adjacent to the filters; m_f is the sum of the masses of tracer found on the filters; Λ_w

is the assumed removal coefficient due to wet deposition (washout coefficient); and Λ_A is the apparent washout coefficient.

In general, previous work^(1,2) has indicated that there is little or no dry deposition at the first arc. Thus, using the assumptions that at Arc A, the apparent washout coefficient is equal to the true washout coefficient and using Equation 1, then the deposition velocities can be calculated. The results are shown in Table 2. The consistency of these velocities at the outer arcs is apparent but care must be taken in the interpretation of the magnitudes which are given in the table. The reason for this equivocation is that in the experiments the filters were facing downwards to avoid wetting whereas Equation 1 does not account for this orientation. Further experiments are planned to determine the relative efficiency of the filters at various orientations.

For experiment UIL-22, a calculation of the theoretical washout coefficient was made using collection efficiencies predicted by Mason.⁽³⁾ The integration in the expression for the washout coefficient,

$$\Lambda_T = \int_0^{\infty} FEA \, dD \quad (2)$$

was approximated by a summation. In Equation 2, F is the raindrop flux density; E is the collection efficiency; and A is the cross sectional area for drops of diameter D. Raindrop size spectra data which were

collected during the experiment were used in the calculation. The result is given in Table 2. The value agrees well with the experimental determination of the washout coefficient for Arc A.

An initial series of experiments was conducted at Quillayute to determine the below-cloud washout of the common pollutant sulfur dioxide. The nature of this tracer required several modifications of the basic procedure which has been reported elsewhere.^(2,4) The sulfur dioxide was dispensed from small commercial cylinders which were heated in a controlled manner to vaporize the liquid and allow a steady release rate.

Although SO₂ is quite soluble in water, the solutions have a considerable partial pressure which demanded special care in collecting and handling samples. Immediately after the sampling, the bottles were capped to retain the SO₂, and it was demonstrated that little loss of SO₂ would occur during the time required for sampling and transporting the samples to the field laboratory.

The analytical method used was a modified West and Gaeke⁽⁵⁾ procedure, in which the sulfur dioxide in the sample is absorbed by a tetrachloro-mercurate solution, forming a non-volatile ion. This ion is subsequently measured by spectrophotometry, after color development by pararosaniline-formaldehyde reagents.

The results of three field experiments are shown in Table 3. In all the experiments, the mass which was

TABLE 3. Sulfur Dioxide Washout Results

Experiment	Mass Released, g	Wind Speed, cm sec ⁻¹	Rainfall Intensity, mm hr ⁻¹	Λ_A , sec ⁻¹ × 10 ⁵		Λ_T , sec ⁻¹ × 10 ⁵
				Arc A	Arc B	
UIL-16	680	788	2.8	2.3 ± 0.2	5 ± 0.5	---
UIL-17	645	264	3.7	3.1 ± 0.4	12.5 ± 1.3	11.0
UIL-21	1260	425	1.6	---	---	8.0

recovered was not sufficient for the desired accuracy in analysis. The errors that are shown indicate the extent of this deficiency. In particular, UIL-21 did not result in any recoverable masses, despite the higher quantity which was released.

Theoretical washout coefficients were also computed and are shown in Table 3. The theoretical washout coefficient for perfectly soluble gases is given by⁽⁶⁾

$$\Lambda_T = \int_0^{\infty} \pi Y \text{ Sh } D \frac{F}{u} dD, \quad (3)$$

where Y is the diffusivity of the gas (≈ 0.1 cm²/sec for SO₂); Sh is the Sherwood number; F is the flux of raindrops of diameter D ; and u is the terminal velocity of the raindrops.

For spheres, the Sherwood number is given by

$$\text{Sh} = 2 + 0.6 (\text{Re})^{1/2} \left(\frac{\nu}{Y} \right)^{1/3}, \quad (4)$$

where Re is the Reynolds number and ν is the kinematic viscosity.

The calculations were made using measured raindrop spectra and by approximating the integral by a summation over raindrop-size intervals. No raindrop spectra were available for UIL-16. The experimental results are well below both the theoretical calculations and the theoretical results of Engelmann, et al.⁽⁶⁾ which are shown as a solid line in Figure 1. The only exception is the value for UIL-17, Arc B. No deposition measurements were made in these experiments.

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FIGURE 1. Sulfur Dioxide Washout Coefficients

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THE ROLE OF ANISOTROPIC BROWNIAN DIFFUSION IN
THE PRECIPITATION SCAVENGING OF SUBMICRON PARTICLES*

W. G. N. Slinn

The concept of anisotropic Brownian diffusion is discussed within the context of precipitation scavenging of submicron particles. The analysis starts from a generalized, convective diffusion equation which contains the influence on the motion of Brownian particles of nonuniformities in the host gas. These effects include tensor diffusivity and "stressphoresis." A boundary layer approximation to the new equation reveals that the corrections to the use of Einstein's diffusion coefficient are insignificant for below-cloud scavenging. Finally, it is indicated that the new effects could be important in flows with large Mach or Knudsen numbers.

Of the many physical processes that could be significant in the precipitation scavenging of submicron particles from the atmosphere, one of the most important appears to be Brownian diffusion of the particles to the cloud-droplet or raindrop surface. Recently a heat transfer analogy to the convective diffusion equation was used to calculate the washout coefficient for particles of radii less than about 0.1μ .⁽¹⁾ This analogy was shown to be capable of predicting, reasonably well, the difficult-to-obtain experimental measurements of the washout coefficient.⁽²⁾

One of the uncertainties in the comparison of theoretical to experimental results arose from our use of Einstein's diffusion coefficient in

the convective diffusion equation. Although this is the traditional procedure, it is prima facie invalid. Einstein's diffusion coefficient is derived under the restrictive assumption that the host environment, in which the Brownian particles are diffusing, is in thermodynamic equilibrium. For example see Chandrasekhar's classic review article⁽³⁾ and references therein. On the other hand, in the neighborhood of a falling raindrop (and, for that matter, in most engineering applications of the convective diffusion equation) the environment is not in equilibrium.

We have recently developed a method for determining the influence of nonuniformities in the host gas on Brownian diffusion of particles whose Knudsen number is large compared to unity.^(4,5) One of the predictions of the resultant, generalized, convective diffusion equation is that in a nonuniform gas

* *This report is a condensation of a paper which has been accepted for publication in The Journal of Geophysical Research.*

flow, Brownian diffusion is anisotropic. In this report, a boundary layer approximation to the new convective diffusion equation will be explored to determine the order-of-magnitude of the effect of anisotropic Brownian diffusion on the precipitation scavenging of submicron particles.

Since the objective here is to determine only the order-of-magnitude of the effect of anisotropic Brownian diffusion, a number of simplifications of the general result¹⁵) will be made. For incompressible, isothermal flow of the host gas, the new convective diffusion equation (see Reference 6, Equation 1) simplifies to

$$\frac{\partial N}{\partial t} + \vec{v}_f \cdot \nabla N = \nabla \cdot \left[\tilde{D} \cdot \nabla N - N \vec{v}_e \cdot \left(\tilde{I} + \frac{\tilde{\tau}}{5p} \right) \right], \quad (1)$$

in which N is the number density of aerosol particles, $\vec{v}_f$ is the velocity of the host fluid, and $\vec{v}_e$ is any "external" drift velocity of the particles which might arise, for example, from electrical effects. In Equation 1, the diffusion coefficient appears as the second order tensor

$$\tilde{D} = D \left(\tilde{I} - \frac{\tilde{\tau}}{5p} \right) \quad (2)$$

where D is Einstein's (scalar) diffusion coefficient, $\tilde{I}$ is the identity or metric tensor, $\tilde{\tau}$ is the viscous tensor and p is the pressure in the gas.

When there is no "external" drift velocity, the steady-state version of Equation 1 is

$$u_i \frac{\partial N}{\partial x_i} = D \frac{\partial}{\partial x_i} \left[\left(\delta_{ij} - \frac{\tau_{ij}}{5p} \right) \frac{\partial N}{\partial x_j} \right] \quad (3)$$

To obtain a boundary layer approximation to Equation 3, consider the two-dimensional flow over a flat plate lying in the xz -plane with the x -axis (direction i) in the direction of the main-stream flow and the y -axis (unit vector j) perpendicular to the plate. Using the boundary layer approximation for the viscous stress tensor,

$$\tau = \rho \nu \frac{\partial}{\partial y} (\hat{i}\hat{j} + \hat{j}\hat{i}), \quad (4)$$

where ρ is the density and ν is the kinematic viscosity of the gas, then Equation 3 becomes:

$$u \frac{\partial N}{\partial x} + v \frac{\partial N}{\partial y} = D \left[\frac{\partial^2 N}{\partial x^2} - \frac{\partial}{\partial x} \left(\frac{\rho \nu}{5p} \frac{\partial N}{\partial y} \right) - \frac{\partial}{\partial y} \left(\frac{\rho \nu}{5p} \frac{\partial N}{\partial x} \right) + \frac{\partial^2 N}{\partial y^2} \right] \quad (5)$$

in which the fluid's velocity has been written as $\vec{v}_f = u\hat{i} + v\hat{j}$. Now, each term in Equation 5 will be nondimensionalized so that its order-of-magnitude is apparent. Let L be a characteristic length in x -direction and let U_0 be the speed of the main-stream. It is assumed that the Reynold's number, $Re = L U_0 / \nu$, is large compared to unity so that the boundary layer approximation will be adequate.

Actually, there is a complication in the nondimensionalization which arises because there are two characteristic lengths in the y -direction: the thickness of the velocity boundary layer, $\delta \simeq L/Re^{1/2}$, and the thickness

of the "diffusion layer", δ_* , say. These two lengths are approximately equivalent for the special case when the diffusion of vorticity (diffusivity ν) proceeds at approximately the same rate as the diffusion of the particles (diffusivity $\simeq D$). For the case of scavenging of sub-micron particles the ratio of these two diffusivities, the Schmidt number, $Sc = \nu/D$, is generally large compared to unity. Thus δ_* is smaller than δ .

If the density of particles is N_0 at distances far from the plate, and if the plate is a perfect absorber of particles so that $N = 0$ at $y = 0$, then the order of magnitude of the term $\partial N / \partial y$, in Equation 5, is N_0 / δ_* . In addition, since $u = 0$ at $y = 0$ and $u \simeq U_0$ at the outer edge of the velocity boundary layer, then at $y = \delta$, $u \sim (\delta_*/\delta) U_0$. Consequently the following nondimensionalized (primed) variables suggest themselves:

$$\begin{aligned} x &= Lx'; & y &= \delta_* y'; \\ u &= (\delta_*/\delta) U_0 u' \end{aligned} \quad (6)$$

To obtain an appropriate nondimensionalization for the y-component of the velocity, the variables defined by Equation 6 are substituted into the continuity equation for the gas,

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \quad (7)$$

and yield

$$v = (\delta_*^2 / \delta L) U_0 v' \quad (8)$$

Substituting these nondimensional quantities in Equation 5 and dropping the primes leads to

$$\begin{aligned} u \frac{\partial N}{\partial x} + v \frac{\partial N}{\partial y} &= \frac{\delta L^2}{\delta_*^3} \frac{1}{Pe} \left[\frac{\partial^2 N}{\partial y'^2} + \frac{\delta_*^2}{L^2} \frac{\partial^2 N}{\partial x'^2} \right. \\ &\quad - \frac{\rho \nu U_0 \delta_*}{5p L \delta} \left\{ \frac{\partial}{\partial x'} \left(\frac{\partial u}{\partial y'} \frac{\partial N}{\partial y'} \right) \right. \\ &\quad \left. \left. + \frac{\partial}{\partial y'} \left(\frac{\partial u}{\partial y'} \frac{\partial N}{\partial x'} \right) \right\} \right], \end{aligned} \quad (9)$$

where $Pe = Re Sc$ is the Péclet number.

The left-hand side of Equation 9 describes the convection of the Brownian particles by the fluid. At the outer edge of the "diffusion layer" the dominant term on the right-hand side of Equation 9, which is evidently the first term, becomes of order unity. Thus an estimate for the thickness of the "diffusion layer" can be found from

$$\frac{\delta L^2}{\delta_*^3 Pe} \simeq 1 \quad (10)$$

Since $\delta \simeq L/Re^{1/2}$, then Equation 10 predicts that

$$\frac{\delta_*}{L} \simeq \frac{1}{Re^{1/2} Sc^{1/3}} \quad (11)$$

This result is given in Landau and Lifshitz, Reference 7. Returning to Equation 9, it is seen that the second term on the right-hand side is of order $(Re Sc^{2/3})^{-1}$ and therefore is ignorable since both the Reynolds and

Schmidt numbers are assumed to be large compared to unity.

Now, an estimate for the new terms in Equation 9 can be obtained. The coefficient is

$$\frac{\rho v U_o \delta_*}{5p L \delta} \simeq \frac{\gamma}{5} \left(\frac{U_o}{\gamma RT} \right)^2 \frac{v}{U_o L} \frac{Re^{1/2}}{Re^{1/2} Sc^{1/3}}$$

$$\simeq \frac{\gamma}{5} \frac{M_o^2}{Re Sc^{1/3}}, \quad (12)$$

where γ is the usual ratio of specific heats for the gas and M_o is the Mach number of the main-stream. Since for a falling raindrop $M_o \ll 1$, the influence of anisotropic Brownian diffusion in below-cloud scavenging is insignificant.

However, the effect can be significant in other circumstances, for example in supersonic and hypersonic boundary layers. Other applications can be seen by recasting Equation 12 into the form

$$\frac{\rho v U_o \delta_*}{5p L \delta} \simeq \frac{\bar{c} \gamma U_o}{\bar{c}^2 L Sc^{1/3}} \simeq \frac{Kn M_o}{Sc^{1/3}}, \quad (13)$$

where $\bar{c}$ is the mean thermal speed of the molecules and where Kn is the Knudsen number of the flow field (which is to be distinguished from the Knudsen number of the Brownian particle). For fixed M_o and Sc , it is seen that if Kn is increased (to, for example, free molecule flow for which $Kn > 1$) then the effect can become important. Of course for free molecule flow, the Chapman-Enskog distribution function, which

was used to obtain Equation 1, is no longer appropriate. However, a detailed analysis will soon be reported* which shows that the phenomenon of anisotropic Brownian diffusion occurs also in free molecule flow.

In conclusion, the asymptotic form for the nondimensionalized mass transfer, the Sherwood number, can be estimated. In the case of perfect retention efficiency, the flux of submicron particles to a raindrop's surface is of order $D N_o / \delta_*$. Using Equation 11 and nondimensionalizing the flux with $D N_o / R$, where R is radius of the raindrop, then for large Reynolds and Schmidt numbers, the Sherwood number is

$$Sh \simeq Re^{1/2} Sc^{1/3} \quad (14)$$

Perhaps it is useful to mention that Equation 14 should be asymptotically valid for arbitrarily large Reynolds and Schmidt numbers.

Based on a heat transfer analogy to the usual convective diffusion, the empirical expression

$$Sh = 1 + 0.39 Re^{1/2} Sc^{1/3} \quad (15)$$

was used in our previous report.⁽¹⁾ Now, with some confidence, we can state that it is acceptable to use Einstein's diffusion coefficient in this equation.

* See the contribution in this volume entitled: "Anisotropic Brownian Diffusion in Free Molecule Couette Flow."

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OLYMPIC PENINSULA RAIN: CHARACTERIZATION OF SIZE AND ELECTRIC CHARGE

M. T. Dana

A number of raindrop charge and size distributions, computed from data collected at a research site on the Olympic Peninsula, have been grouped, and their median values characterized, in terms of rainfall intensity. The median diameter increases at approximately the one-sixth power of rainfall intensity J_0 , while the median charges increase at powers of J_0 from one-half to one. Comparisons of gross size and charge spectra indicate a dependence of the charge upon raindrop area, in agreement with the theoretical proposals of Gunn and Sartor. The rainfall intensity dependency apparently is complicated by a number of meteorological variables.

Three seasons of data on characteristics of precipitation have now been collected at our field research site, Quillayute Air Base, Olympic

Peninsula. Data that have been obtained include rainfall intensity, raindrop electric charge and raindrop size spectra. To date, the

analysis of these data allows us to make gross characterizations of the electric charge and raindrop size parameters for the storm-frontal activity rain which predominates at the site.

It should be emphasized that these characterizations may hold only for a limited geographic region. As such, the orographic influences of the coastline and mountain slopes probably cause a rapid change in rainfall characteristics with distance inland. For example, it is known that the annual amounts of rainfall are significantly greater, closer to the crest of the Olympic Mountains. The existing research site is located

approximately 5 miles inland at an elevation of 195 ft.

Thirty-seven raindrop size spectra were collected as spot images on water sensitive paper. These spectra were grouped according to the rainfall intensity which was computed from the individual spectra. In Table 1 are listed the rainfall intensity intervals, the number of raindrops which were sized and the number of individual spectra included. Also presented are the geometric number-median diameters of the lognormal distributions.

In Figure 1 are plotted these geometric number-median diameters for the lognormal distributions as a func-

TABLE 1. Raindrop Size Spectra

Spectrum	Rainfall Intensity ^(a) Interval, mm hr ⁻¹	Raindrops Sized	Individual Spectra	Number Median Diameter, mm
B	0.2 - 0.4	1510	7	0.41
C	0.4 - 0.6	427	2	0.43
D	0.6 - 0.8	907	4	0.84 ^(b)
E	0.8 - 1.0	818	4	0.46
F	1.0 - 1.2	429	2	0.90 ^(b)
G	1.2 - 1.4	193	1	0.68
H	1.4 - 1.6	627	3	0.54
I	1.6 - 2.0	638	3	0.40
J	2.0 - 3.0	1520	6	0.62
K	3.0 - 5.0	525	2	0.57
L	5.0 - 7.0	522	2	0.80
M	7.0 - 9.0	281	1	0.52
TOTAL		8397	37	0.58

a. Rainfall intensity computed from individual spectra

b. Normal distribution

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FIGURE 1. Relationship of Median Raindrop Diameter to Rainfall Intensity

tion of rainfall intensity. The least-squares fit to the data is given as the solid curve

$$D = 0.49 J_0^{0.15} \quad (1)$$

where D is the median diameter in mm and J_0 is the rainfall intensity in mm hr⁻¹. The rather special case normal distributions have been omitted.

The raindrop charge data were collected with the spectrometer developed at Battelle-Northwest. The principle and functions of this spectrometer are described elsewhere.^(1,2) The

records which were analyzed were obtained from different days and separate storms. It was found that the rainfall intensity dependence of the charge varies significantly with these different occasions and therefore it is apparent that, until detailed knowledge of the mechanisms which are responsible for the charging is determined, characterization must be made on the basis of individual storms.

Sufficient data were collected during 2 days in December, 1968, and these are grouped according to rainfall intensity in Table 2. The

TABLE 2. Raindrop Charge Spectra

Spectrum	Rainfall Intensity ^(a) Interval, mm hr ⁻¹	Number of Individual Samples		Number of Raindrop Charges			
				12/7/68		12/13/68	
		12/7/68	12/13/68	+	-	+	-
A	0.3 - 0.5	2	5	96	24	224	137
B	0.5 - 1.0	5	11	131	229	741	361
C	1.0 - 1.5	2	5	177	33	264	116
D	1.5 - 2.0	13	11	1146	442	906	357
E	2.0 - 2.5	2	4	140	47	290	136
F	2.5 - 3.0	3	6	96	58	385	280
G	3.0 - 4.0	8	8	585	186	326	188
H	4.0 - 5.0	6	4	576	144	298	140
I	5.0 - 7.0	2	4	175	65	296	136
J	7.0 - 9.0	3	-	164	72	---	---
TOTAL		46	58	3286	1300	3740	1851

a. Rainfall intensities from weighing rain gage

rainfall intensities were those measured by a weighing rain gage which was developed in the laboratory.

The geometric number-median magnitudes of the charges are plotted in Figures 2 and 3. It was found that each polarity of the charge samples had close to lognormal distributions. Some of the samples which were listed in Table 2 were combined to obtain the plots shown in the figures. Previously, mean charges were computed.^(1,3,4) However, this method of reduction of the data is now considered to be too sensitive to the charges of large magnitude, since the distributions are so broad. That is, a wide range of magnitudes is included in each sample.

The data for December 13 fit fairly closely to a power-law rela-

tion between the median charge and the rainfall intensity. However, those of December 7 are quite flat at low rainfall intensity. The least-squares fit to the curves in Figure 3 are approximately

$$q_+ = 63 J_0^{1/2}, \quad (2)$$

$$q_- = -20 J_0, \quad (3)$$

where median charge q is given in units of 10^{-15} coulomb and rainfall intensity J_0 is measured in mm hr^{-1} .

An analysis, similar to those which led to Equations 2 and 3, was made on the total bipolar charge distributions. Since these distributions fit neither the normal nor the lognormal regimes, the actual, median charge value from the data

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FIGURE 2. Median Charge-Rainfall Rate Relationship: Data of December 7, 1968

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FIGURE 3. Median Charge-Rainfall Rate Relationship: Data of December 13, 1968

was taken. The resulting median net charge is given by

$$q_n \approx +15 J_0^{0.75}. \quad (4)$$

When displayed as a linear-probability plot, the shape of the bipolar distributions is approximately normal but with a deficiency of magnitude near zero. To some extent, this deficiency is expected because of the limited sensitivity of the instrument.⁽³⁾ However, an examination of the sensitivity characteristics of the instrument and the conditions under which the measurements were made leads to the conclusion that there must be a natural deficiency of near-neutral raindrops.

Simultaneous measurements of size and charge of individual raindrops were not made in this study. However, a guarded comparison of the raindrop size and electric charges can be made by comparing gross spectra. Examination of the corresponding spectra for each rainfall intensity group has indicated that the geometric standard deviation of the unipolar charge distribution agrees closely with the geometric standard deviation of the distribution of the square of the size.

This result gives some insight into possible charging mechanisms. Several theoretical developments have indicated a dependence of the electric charge on the raindrop area.^(5,6,7) If we accept that the charge is proportional to the raindrop area, then the rainfall intensity dependence of the charge can

be determined. Thus, from Equations 1 through 4, we obtain:

$$q_+/D^2 = 266 J_0^{0.2}, \quad (5)$$

$$q_-/D^2 = -83 J_0^{0.7}, \quad (6)$$

and

$$q_n/D^2 = +63 J_0^{0.45} \quad (7)$$

There is no obvious reason why the negative charge per unit area should differ from the positive. However, it may arise from different sizes of the negatively charged rain drops. To investigate this hypothesis, studies are in progress to determine the charge distributions which are recorded when the transducer of the raindrop charge spectrometer is oriented in various directions relative to the wind. The wind has, of course, a greater influence on smaller raindrops. Consequently, we should be able to obtain preliminary results about the magnitude and polarity of the charge as a function of raindrop size.

Further work is in progress in an attempt to test the theories of the charging raindrops which have been proposed by Gunn⁽⁵⁾ and Sartor.⁽⁶⁾ These theories indicate that the value of the charge-to-area ratio should vary with the thickness of cloud layer, liquid water content of the cloud, cloud-droplet size distribution and other meteorological parameters which are expected to vary as does the rainfall intensity. In this regard, it is noted that the

charge per unit area for the December 13 data is an increasing function of rainfall intensity.

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PHORETIC AND BROWNIAN DIFFUSION PROCESSES IN THE
IN-CLOUD SCAVENGING OF AEROSOL PARTICLES*

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

An analysis is made of the relative importance of thermophoresis, Brownian diffusion and diffusiophoresis, for the in-cloud scavenging of aerosol particles, accompanying the condensation and Bergeron-Findeisen process. Under the assumption that all of the latent heat associated with vaporization (condensation) is conducted to (from) the precipitation embryo, it is shown that thermophoresis is of dominant importance for particles of radii between 0.01 to 1.0 μ . The steady-state solution to the appropriate convective-diffusion equation is presented. It is suggested that thermophoresis could be a significant mechanism for the in-cloud contact ice nucleation of water droplets in a subfreezing cloud.

Although it is known that precipitation scavenging accounts for more than 80% of the long-term removal of radioactive material from the atmosphere,⁽¹⁾ neither experiments nor theories have yet established whether below- or in-cloud scavenging processes are of dominant importance. For a review of some of the contradictory statements in the literature about this point, see Reference 2. However, it is probably not too controversial to state that much more is known about below-cloud than in-cloud scavenging.

Although there are many physical processes involved in the below-cloud scavenging of pollutants from the

atmosphere, the theory of the rain-scavenging of aerosol particles has been fairly well developed by Langmuir,⁽³⁾ Greenfield,⁽⁴⁾ Ziman,⁽⁵⁾ and others. However, there remains a major uncertainty about the role played by electrical effects. In another contribution to this report,^t hereafter referred to as I, we have demonstrated that thermophoretic effects can be significant if the relative humidity of the atmosphere is less than 100%. Future analyses of the below-cloud scavenging problem should concentrate on the rain-scavenging of gases as well as the removal of all pollutants by other types of precipitation.

* This report is based on a portion of an article by the authors which has been submitted for publication in the Proceedings of the 1970 Precipitation Scavenging Meeting.

† See the report by the authors entitled "Phoretic and Brownian Processes in the Below-Cloud Scavenging of Aerosol Particles by Rain." p. 70.

The in-cloud scavenging theory is even more difficult and less developed. A major complication is that the dynamics of the clouds must be considered. Further, there are problems involved in understanding the interaction of the pollutant with "microphysical" processes such as condensation and ice nucleation. Recently, Hicks⁽⁶⁾ investigated the removal of aerosol particles by the direct condensation processes. It is expected that this effect would be most significant for particles of radii larger than about 10 μ , since it would require approximately an hour and a half for a particle of a few tenths of a micron to grow by condensation to a size of about 20 μ , at which size coalescence is significant.⁽⁷⁾ Little is known about the interaction of aerosol particles with the primary stages of ice nucleation.⁽⁶⁾ The objectives of this paper are to study the scavenging of aerosol particles by other processes which occur during the formation of precipitation.

In I, the phenomena of thermo- and diffusiophoresis were described. In this report, an investigation will be made of the influences of Brownian diffusion and the phoreses effects on the motion of aerosol particles which are in the neighborhood of precipitation embryo.

The analysis is appropriate for two different mechanisms of precipitation growth. For one, the Bergeron-Findeisen process,⁽⁷⁾ we consider an ice crystal which is growing by

condensation at the expense of the evaporation of cloud droplets in its neighborhood. For the other process, which is applicable to nonfreezing clouds, we consider the growth of individual droplets by condensation. In both these cases, the aerosol particles are influenced by the heat and water-vapor fluxes, to and from (as appropriate) the precipitation elements.

We make the following assumption which is fundamental to our analysis. It is assumed that all the precipitation elements within the cloud are at, approximately, the temperature of the environment. That is, it is assumed that the latent heats which are associated with vaporization (condensation) are supplied to (removed from) the precipitation elements by conduction from the environment.

That this assumption is a reasonable one can be seen by studying the equation which describes the temperature of a "dormant" precipitation element. If no water vapor is being transferred, then the temperature, T , of the element is approximately governed by

$$m c \frac{dT}{dt} = \left[\frac{k(T - T_a)}{R} \right] 4\pi R^2 \quad (1)$$

where m , c , and R are the mass, specific heat, and radius of the element and k and T_a are the thermal conductivity and temperature of the air. The e-fold, relaxation time for the temperature of the particle is then

$$\tau = \frac{\rho \cdot c \cdot R^2}{3k} \quad (2)$$

where ρ is the density of the embryo.

For a water droplet of 1 μ radius,
 $\tau \approx 10^{-4}$ sec.

From Equations (3), (5), and (9) of I, we can obtain an estimate of the relative importance of the thermophoretic, Brownian diffusion and diffusiophoretic fluxes. The convective terms in these fluxes ($Re^{1/2}$, $Pr^{1/3}$, etc.) probably can be ignored, but whether or not they are, we obtain the same result that thermophoresis is of dominant importance for particles of radii between about 0.01 and 1 μ .

If this result is applied to the two in-cloud processes under consideration, then the following conclusions can be drawn. In the process of condensation growth of cloud droplets, in nonfreezing clouds, thermophoresis tends to drive the aerosol particles of the above size range away from the cloud droplets. That is, the heat flux from the droplet, which is liberated because of the latent heat associated with condensation, has a greater influence on the aerosol particles than the flux of water vapor to the droplet. Similarly, in the Bergeron-Findeisen process, thermophoresis tends to drive the particles away from the ice crystal and toward the evaporating droplets. Since the droplets are supercooled and since the particles impinge on the droplets, it is proposed that thermophoresis could be a

trigger for the contact ice nucleation of the droplets.

Other authors have studied the role of diffusiophoresis in the in-cloud scavenging of aerosol particles. The impetus for these works was Facy's original observations⁽⁸⁾ of a dust-free space in the neighborhood of an evaporating body and his suggestion that this process could be significant to in-cloud scavenging. Goldsmith et al.⁽⁹⁾ studied the role of diffusiophoresis in nonfreezing clouds and concluded that it was not significant. Vittori and Prodi⁽¹⁰⁾ suggested that in the Bergeron-Findeisen process, diffusiophoresis would be enhanced over the values predicted by Goldsmith et al. because of the drift of the particles away from the cloud droplets. We are grateful to Dr. Jan Rosinski⁽¹¹⁾ for sharing with us his thoughts on their suggestion.

In both of the above mentioned studies, thermophoresis was ignored. Concerning this, Goldsmith and May wrote:⁽¹²⁾ "This is justified on the ground that the temperature relaxation time of small droplets in the atmosphere is very short and also that condensation will tend, because of the liberation of latent heat, to warm the droplets and so any temperature gradient would be small." On the other hand, we propose that, exactly for the reason why the temperature relaxation is short (namely, there is significant heat flux involved) then thermophoresis can not be ignored.

There remains the necessity of explaining their experimental

observations^(8,9,10) of a dust free space in the neighborhood of an evaporating liquid. To this end, it is noted that these experiments were conducted so that energy for vaporization might have been supplied by modes other than conduction from the surrounding gas for example, by radiation from illumination sources or by transient conduction from the structure which was supporting the liquid. This would tend to lower the thermophoretic deposition during these experiments. It is suggested, therefore, that although these experiments demonstrated a valid phenomenon, they may not be representative of in-cloud behavior.

A more rigorous comparison of the relative importance of thermophoretic, Brownian diffusion, and diffusiophoretic processes can be obtained by solving the appropriate convective-diffusion equation.^(13,14,15) In contrast to the below-cloud problem, for the in-cloud problem it is reasonable to neglect the velocity field of the air. Thereby, the equation regains its spherical symmetry and more progress toward a solution can be expected.

The time-dependent convective diffusion equation is

$$\frac{\partial N}{\partial t} = \nabla \cdot \left[D_p \nabla N - \vec{v}_d N \right] \quad (3)$$

where N is the number density of aerosol particles and $\vec{v}_d$ is their drift velocity resulting from thermo- and diffusiophoresis. Using the results in I, the drift velocity is seen to be

$$\begin{aligned} \vec{v}_d &= \frac{f}{p} (1 - \kappa) \frac{k(T_e - T_a)}{R} \frac{R^2}{r^2} \hat{e}_r \\ &\equiv \sigma \frac{D_p}{R} \frac{R^2}{r^2} \hat{e}_r \end{aligned} \quad (4)$$

where $\hat{e}_r$ is a unit vector in the r direction, and R is the radius and T_e is the temperature of the precipitation element. The symbols f and κ are defined in Equations I-(5) and I-(14), respectively. The dependence of $\vec{v}_d$ on r results from obtaining the heat flux from the solution to Laplace's equation for the temperature. The parameter σ is essentially the Péclet number of the particle, based on the drift velocity for $r = R$.

The solution to the steady-state version of Equation (4), subject to the perfect retention boundary condition, $N(R) = 0$, is

$$N = N_0 \left[\frac{e^{-\frac{\sigma}{R} r}}{1 - e^{-\sigma}} \right] \quad (5)$$

It is interesting that this is exactly the same form as the solution to the convective-diffusion equation which describes Brownian diffusion and the electrical interaction between a particle and a droplet.⁽¹³⁾

From Equation (5), the number flux of particles to the precipitation element is found to be

$$\vec{F} = \frac{D_p N_0}{R} \left[\frac{\sigma}{e^{\sigma} - 1} \right] (-\hat{e}_r) \quad (6)$$

For a growing ice crystal or water droplet, with σ large and positive, it is seen that the flux of aerosol particles to the element is reduced to

essentially zero. On the other hand, for an evaporating droplet, with a large and negative, the flux of aerosol particles to the droplet is enhanced over that from Brownian diffusion by a factor approximately equal to the magnitude of a . As was mentioned earlier, we propose that this enhancement could significantly influence the in-cloud ice nucleation cloud droplets.

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A NUMERICAL MODEL TO DESCRIBE
OROGRAPHIC PRECIPITATION IN THE PACIFIC NORTHWEST

B. C. Scott

A two-dimensional numerical model is being developed to describe the meteorological events occurring as moisture laden air is orographically lifted over the coastal mountain ranges. The basic equations and limitations of the model are discussed, and some preliminary results from the first test case are presented.

Inside a cloud, moisture content, temperature, and turbulent motions are all known to be related to the volume and type of precipitation, and hence to the degree of in-cloud scavenging and subsequent deposition of pollutants upon the earth's surface. The exact relationships of these variables to the rate and efficiency of scavenging will remain poorly defined until either a large, active, precipitating cloud is physically observed and continuously monitored, or until the precipitating cloud is numerically described. Of these two possibilities it is more feasible to develop a numerical model capable of providing estimates of the meteorological parameters at the time of precipitation scavenging experiments, and thus establish a sufficient background for the description and understanding of the observed events.

To identify regions of nucleation within a cloud and to aid in determining the efficiency of in-cloud scavenging, a numerical model of moisture-laden airflow is being developed. This model will be used

in conjunction with the present sampling of naturally occurring radio-nuclides in an attempt to describe the physical processes occurring in a precipitating air-mass.

Specifically, the model is expected to provide detailed information about the orographic precipitation on the windward side of the Olympic Mountains. The basic procedures for constructing the model have been outlined and developed by Myers.(1) This steady-state, two-dimensional model is independent of mountain width and is applicable for use in a stable atmosphere. The numerical output will provide the data necessary for constructing streamlines of air parcels moving over the mountain. Vertical profiles of temperature, wind speed, and moisture content will be obtainable at several points between the foot and the crest of the mountain.

List of Symbols

P = atmospheric pressure
 P_e = effective pressure
 ρ = density of air

V = wind speed along a streamline
 V_h = horizontal component of the wind speed along a streamline
 T = temperature ($^{\circ}\text{A}$)
 t = time, temperature ($^{\circ}\text{C}$)
 T^* = virtual temperature
 g = gravitational constant
 $= 980 \text{ cm sec}^{-2}$
 z = height
 w = vertical velocity
 R = gas constant for air
 $= 2.87 \times 10^6 \text{ ergs g}^{-1} \text{ deg}^{-1}$
 C_p = specific heat at constant pressure
 $= 1.0 \times 10^7 \text{ ergs g}^{-1} \text{ deg}^{-1}$
 E_s = saturation vapor pressure
 L_w = latent heat of condensation
 W_s = saturation mixing ratio
 k = friction coefficient

The subscripts 1,2,3,4 associated with some of the variables refer to the points labeled 1,2,3,4, in Figure 1.

The compressible, laminar flow of the model can be described by four basic equations: continuity, motion, energy, and state. Several additional relationships have been used to describe latent heat release, saturation vapor pressure, and the mixing ratio of water vapor. A description of each equation and their related assumptions will be sufficient to describe the model. The development is essentially that of Myers.

The vector equation of motion can be reduced to the Bernoulli equation and the hydrostatic equation. The flow along a streamline is described by the Bernoulli equation:

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FIGURE 1. The Streamlines of Airflow over a Barrier

$$\frac{dp}{\rho} + \frac{1}{2} d(V^2) + g dz = 0 \quad (1)$$

This restricts the analyses to laminar, nonviscous flow and ignores the Coriolis Force. If the vertical accelerations are ignored, then the third component of the equation of motion

$$\frac{dw}{dt} = - \frac{1}{\rho} \frac{\partial p}{\partial z} - g \quad (2)$$

reduces to the hydrostatic equation. Thus, the inflow atmosphere is required to be conditionally stable. If orographic lifting in the Pacific Northwest is found to produce frequent instability, the vertical accelerations will have to be included. However, at present, it is believed that a large percentage of the prefrontal precipitation on the Olympic Peninsula falls from a stable atmosphere.

The equation of state is the next basic equation:

$$P = \rho RT^* \quad (3)$$

where T^* is the virtual temperature. Myers apparently neglected the virtual temperature in his development. However, its influence is important because an error of as little as one degree in the average temperature of a lifted layer can produce outflow wind speed errors on the order of 1 to 2 m/sec. Equation (3) is used to eliminate the density from Equations (1) and (2).

By integrating around the closed curve 1,2,4,3 in Figure 1, an energy-velocity relationship in the steady state flow is obtained. Equation (2) is used in the vertical integration and Equation (1), in the integration along the streamlines. The resultant energy relationship is

$$2R \oint T^* d(\ln P) = V_3^2 + V_2^2 - V_1^2 - V_4^2. \quad (4)$$

The continuity equation is used to satisfy conservation of mass:

$$\int V_h dP = \text{Constant}; \quad (5)$$

that is, the total air mass entering the flow between points 1 and 3 leaves between points 2 and 4. The expression used in the model can be obtained by approximating V_h by V ,

and assuming $\bar{V}_{13} = (V_1 + V_3)/2$ and $\bar{V}_{24} = (V_2 + V_4)/2$. This yields

$$\begin{aligned} & (V_1 + V_3) (P_1 - P_3) \\ & = (V_2 + V_4) (P_1 - P_4) \end{aligned} \quad (6)$$

If the airflow remains unsaturated, the dry adiabatic law is used to describe the temperature-pressure relationships

$$\frac{T_1^*}{T_2^*} = \left(\frac{P_1}{P_2} \right)^{R/C_p} \quad (7)$$

The energy equation for the flow of moist air has been derived by assuming that the pseudo-equivalent potential temperature, (θ_e) , is a conservative property of a saturated air parcel. By subtracting the expressions for θ_e at two different levels, an expression for the moist temperature lapse rate results:

$$\begin{aligned} T_4 = T_3 & \left[\left(\frac{P_4 - E_{s4}}{P_3 - E_{s3}} \right)^{R/C_p} \right. \\ & \left. \left(1 + \frac{L_3 W_{s3}}{C_p T_3} - \frac{L_4 W_{s4}}{C_p T_4} \right) \right]. \end{aligned} \quad (8)$$

The temperature expression in Equation (8) allows the latent heat and the saturation mixing ratios to vary as functions of pressure and temperature. At this time only the latent heat of condensation (L_w) is included in the model, although it would be relatively simple to replace L_w with the latent heat of sublimation, or with combinations of both.

The expression for L_w has been obtained from the Smithsonian Tables(2)

$$L_w = 596.73 - 0.601t \text{ cal g}^{-1} \quad (9)$$

where t is in $^{\circ}\text{C}$. The saturation vapor pressure is approximated by(3)

$$E_s = 6.11 \times 10^{at/(t+b)} \quad (10)$$

where $a = 7.5$, and $b = 237.3$. The saturation mixing ratio is defined by the standard expression

$$W_s = 0.622 \left[E_s / (P - E_s) \right] \quad (11)$$

To prevent unrealistic acceleration on streamlines near the ground, a modification of Myers' arbitrary friction approximation is being used. If the reported radiosonde winds used for input data are assumed to be frictionally reduced in the lowest layers, Myers' equation becomes

$$k \left[(V_2^2)_{\text{no Fr}} - \frac{(V_1^2)_{\text{Fr}}}{k} \right] = \left[(V_2^2)_{\text{Fr}} - (V_1^2)_{\text{Fr}} \right] \quad (12)$$

The subscript "Fr" defines the terms enclosed by brackets as being frictionally reduced, and conversely, "no Fr" identifies the bracketed terms as being unaffected by friction. The constant k is defined as a function of height above an "effective" ground surface; e.g., the top of a cold stable dome of air, or the top of a strong surface inversion.

$$k = 0.5 \left[1 + \frac{P_e - P}{0.1 P_e} \right] \text{ for } P \geq 0.9 P_e;$$

$$k = 1.0 \text{ for } P < 0.9 P_e. \quad (13)$$

Here P_e equals the pressure at the effective ground.

From Equation (12), the frictionless wind at point 1 in Figure 1 becomes

$$(V_1)_{\text{no Fr}} = \frac{(V_1)_{\text{Fr}}}{\sqrt{k}}.$$

Similarly, the frictionless wind at point 2 becomes

$$(V_2)_{\text{no Fr}} = \frac{1}{\sqrt{k}} (V_2)_{\text{Fr}}.$$

Newton's method⁽⁴⁾ has been used in conjunction with Crout's compact elimination algorithm⁽⁵⁾ to solve the above set of nonlinear and linear equations for the outflow temperature, wind, and moisture profiles. Crout's algorithm was chosen because, truncation and round-off errors can easily be determined from the step size of the final iterations. The wind speed is usually the last variable to converge and hence has the largest truncation error. Truncation and round-off errors vary from 10^{-3} cm/sec for the wind speed to 10^{-8} g/g for the saturation mixing ratios.

There has been one observed weakness with Crout's method in solving the above system of equations. Occasionally the determinant of the system of equations will go to zero and terminate the iteration process before a final solution has been

found. This occurs most frequently in the solution of equations with dry adiabatic motions. However, with the aid of a more laborious iteration scheme, it has been found that for these cases the actual solution to the equations violates the laminar assumptions of the model.

The computer programming of the model has been completed and comparisons of hand calculations with computer output indicate that the program is running correctly. A series of case studies is now being conducted in order to test the various assumptions of the model.

Rawinsonde data are available from two experimental sites on each side of the Cascade Range. The sites are about 80 km apart and are separated by mountains rising to elevations between 1200 and 2100 m. The first trial runs of the model utilize data from 0400 PST January 30, 1969. This particular set of data was selected because from the surface to great heights the horizontal airflow was nearly perpendicular to the mountain ridges.

Figure 2 shows the complete calculated airflow pattern using as input the data the rawinsonde reports from Enumclaw (ENC) and Cle Elum (CLE), Washington. As can be seen from the figure, the model has been extended to the lee side of the mountain range where verification of the total flow pattern is believed to be indicative of successful descriptions at points in between. The computed results are generally in agreement with the observed meteorological events although the wind

speeds at CLE have been overestimated. CLE was reporting overcast skies at 0400 PST and a station near the mountain crest was reporting light to moderate snow throughout the study period.

In Figure 3, the calculated temperature profile at CLE is compared with the observed temperature at 0400 and 1000 PST. The calculated temperature profile takes on the major characteristics of both observed soundings, i.e., the cooling in the lower levels and the large scale warming above 600 mb. These preliminary results are encouraging because the computed results apply to the time the slowest air parcel completes its trip from ENC to CLE, or at about 0700 PST.

Figure 4 has been included for general interest and is a plot of the computed vertical velocities. The packing of the isotacks at the mountain crest is a result of the discontinuity in the airflow streamlines above the mountain peak. The subsidence to the lee of the mountain was expected although the magnitude is not so great as anticipated. The airflow has apparently been lofted over an arctic air mass to the east of the mountain.

Many of the present assumptions will no doubt prove to be too restrictive or unrealistic for initial successes with the case studies and several modifications are anticipated. Among those considered for revision in the near future are the friction and latent heat estimates. In addition, tests are presently being conducted to determine

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FIGURE 2. Airflow Streamlines for the Test Case Study of January 30, 1969. Shaded area is the orographically formed cloud.

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FIGURE 3. Computed Versus Observed Temperature Profiles, January 30, 1969

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FIGURE 4. Computed Vertical Velocities (m/sec) for the Test Case Study of January 30, 1969

the effect of various mountain heights and various cloud liquid water contents on the outflow profiles.

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COSMOGENIC RADIONUCLIDE TRACERS OF IN-CLOUD SCAVENGING PROCESSES*

R. W. Perkins, C. W. Thomas, J. A. Young, and H. G. Rieck, Jr.**

Technology has been developed(1-3) which allows the measurement of the short-lived cosmogenic radionuclides ^{24}Na , ^{38}Cl , and ^{39}Cl in rain during consecutive sampling periods as short as 10 min. The technique involves collection of ^{24}Na on cation and ^{38}Cl and ^{39}Cl on anion resin beds, followed by direct counting on multi-dimensional gamma-ray spectrometers. Measurement precision is better than $\pm 10\%$, providing excellent resolution for studying in-cloud processes.

Observation of three rainstorms in Washington with this technique suggests the following sequence in rainwater development. Cloud droplets form on condensation nuclei which have been labeled with the cosmogenic radionuclides. During subsequent growth of the cloud droplets through coalescence and condensation, additional collection of newly formed cosmogenic radionuclides appears to be small or negligible. During their in-cloud development, the raindrops may be subjected to several cycles of partial evaporation followed by further coalescence and condensation, particularly in light rains.

The results indicate that light rains have spent a substantially longer period in their development than heavy rains. They show both higher radionuclide concentrations and higher, long- to short-lived radionuclide ratios. The time period for raindrop development ranged from approximately 30 to 130 min in the light to medium rains which were studied, based on the $^{39}\text{Cl}:$ ^{38}Cl ratio of rainwater relative to that of the atmosphere. Rainout coefficients for the storms were in the range of 10^{-3} to 10^{-2} sec^{-1} but could be some two-to three-fold lower, depending on the effective altitude of the cloud top. These rainout coefficients were determined by comparing the radionuclide deposition per unit area at ground level (decay corrected to the time of nucleation) to the radionuclide content per unit area of cloud in an air column of cloud thickness. The apparent "time since previous rainout" in the rainstorms ranged from 30 to 200 min from estimates of the $^{24}\text{Na}:$ ^{38}Cl ratio in the rainwater. These very short values would require that essentially complete rainout of the cosmogenic radionuclides occur at these time intervals. This apparent behavior is in agreement with the observed high rainout coefficients (Figure 1).

* This report also appears in
BNWL-2307 Part 2.

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FIGURE 1. Rainout Coefficients for ^{38}Cl Observed During Two Rainstorms, February 15, 1969 and April 3, 1969.

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AN INTERPRETATION OF THE IN-CLOUD SCAVENGING OF PARTICULATES

W. E. Davis

Results are presented of a comparison of computed rates of deposition, arrived at by assuming an in-cloud scavenging coefficient, with rates of deposition computed from observations of cosmogenic radionuclides in rainwater. In-cloud scavenging coefficients of 10^{-4} to 10^{-3} sec^{-1} would agree with the deposition rates calculated from observations for ^{24}Na . However, the same coefficients would not predict the rates of deposition calculated from observations of ^{38}Cl .

Understanding in-cloud scavenging is important for estimating the removal of radioactive debris as well as other pollutants from the atmosphere. In the past few years, Soviet scientists^(1,2) have evaluated in-cloud scavenging coefficients and obtained average values of 10^{-4} to $\sim 5 \times 10^{-3} \text{ sec}^{-1}$. Recently, R. W. Perkins et al.⁽³⁾ have presented unique observations of cosmogenic radionuclides in rainwater. These results are useful for interpreting in-cloud scavenging. The following presentation utilizes these data to calculate an in-cloud scavenging coefficient, assuming a continuous process for the pickup and removal of the aerosol particles which are tagged with the radio-nuclide ^{24}Na .

In this report, the transfer of aerosol particles from free air to cloud water will be referred to as in-cloud scavenging. The nuclide concentration, N_a , in an air parcel in the presence of cloud water is determined by

$$\frac{dN_a}{dt} = -\alpha N_a - \lambda N_a + B, \quad (1)$$

where α is the decay coefficient; A is the in-cloud scavenging coefficient; and B is the rate of production. Assuming α , A , and B are constants, then the solution to Equation (1) is

$$N_a = \frac{\lambda N_o}{\alpha + \lambda} e^{-(\lambda + \alpha)t} + \frac{B}{\alpha + \lambda}, \quad (2)$$

where N_o is the free-air equilibrium concentration when decay is equal to production.

A similar equation describes the cloud water concentration, N_w , of cosmogenic radionuclides. For $t < t_o$, where t_o is the in-cloud time before precipitation begins, the equation is

$$\frac{dN_w}{dt} = -\alpha N_w + \lambda N_a, \quad t < t_o. \quad (3a)$$

After precipitation begins,

$$\frac{dN_w}{dt} = -\alpha N_w + \lambda N_a - \psi N_w, \quad t > t_o, \quad (3b)$$

where Ψ is the precipitation removal coefficient. Thus $-\Psi N_w$ is the rate of removal of radionuclides in the cloud water, by raindrops leaving the cloud.

Equations (3a) and (3b) do not include production terms. The inclusion of the effects of production can be important if the radionuclides are attached directly to the water drops without using the aerosol particles as intermediaries. The results of the inclusion of the production term in Equations (3a) and (3b) as well as deleting it from Equation (1) will be discussed in a later paper.

Assuming Ψ to be constant, then the solution for N_w is

$$N_w = N_{w_0} e^{-(\Psi+\alpha)t} + \frac{\lambda^2 N_0 e^{-(\lambda+\alpha)t_0}}{(\alpha + \lambda)(\Psi - \lambda)} [e^{-(\alpha+\lambda)t} - e^{-(\Psi+\alpha)t}] + \frac{\lambda B}{(\alpha + \lambda)(\Psi + \alpha)} [1 - e^{-(\Psi+\alpha)t}],$$

$$t \geq t_0. \quad (4)$$

Here, N_{w_0} is the water concentration at time t_0 , obtained from the solution of Equation (3a).

If evaporation and below-cloud scavenging are ignored, then the rate of deposition on the ground can be determined from

$$\frac{dN_g}{dt} = \Psi N_w - \alpha N_g. \quad (5)$$

In Equation (5), the decay term, $-\alpha N_g$, may be ignored, since the data

which will be used have been corrected for this decay.

On February 15, 1969 and April 3, 1969 observations of the deposition of cosmogenic radionuclides were made at Quillayute, Washington. For these test periods, radiosonde observations were investigated for cloud height and depth. A cloud extending from 1 to 2 km was indicated by the soundings. Using this thickness, assuming a cloud water concentration⁽⁴⁾ of 0.25 g/m^3 and a mean rainfall rate of 0.15 cm/hr , a precipitation removal rate of $1.4 \times 10^{-3} \text{ sec}^{-1}$ was calculated. Further, using this cloud height and depth, then a clear air concentration of 0.2 dpm for a column 1 km by 1 m^2 was calculated for ^{24}Na from the date of February 15, and the first half of the data from April 3, 1969. This clear air concentration and an assumed t_0 of 1000 sec was then used in computing a deposition rate of ^{24}Na from an individual air parcel.

As can be seen in Figure 1, the deposition rate remains fairly constant for the first 10 min after deposition begins (t_0 to $t_0 + 10 \text{ min}$). Assuming horizontal homogeneity, the deposition rate may be calculated from Equation (5). For this example, a time since precipitation from the air parcel began was selected as 5 to 10 min.

Comparing the deposition values calculated from $N_w \Psi$ in Figure 1 with those calculated from observed rates, the following was found. The observed values appear to be in a range

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FIGURE 1. Plot of Deposition Rate, ψ_{N_w} , of ^{24}Na for Various In-Cloud Scavenging Coefficients as a Function of Time Following an Air Parcel

of in-cloud scavenging coefficient of 10^{-4} to 10^{-3} sec^{-1} . These values can be altered considerably by assum-

ing a different cloud water content or a different cloud depth. A decrease from 0.25 to 0.1 g/m^2 would

cause the values of the deposition rate to increase, which in turn would cause the values associated with the in-cloud scavenging coefficient to decrease from the above range to the range: 5×10^{-5} to $8 \times 10^{-4} \text{ sec}^{-1}$.

In-cloud scavenging coefficients* of 1×10^{-4} to 10^{-3} sec^{-1} were obtained through a comparison of calculated rates of deposition with rates of deposition calculated from ^{24}Na data taken during the storms of February 15 and April 3, 1969. It was assumed that there is a continuous process of uptake and removal of the radionuclides by the cloud water. Since these results can be dramatically altered by assuming different liquid water contents of the cloud and cloud thickness, it is recommended that cloud water content be measured as well as cloud depth.

The model described here should be considered as exploratory since

it does not yet adequately explain the scavenging of aerosol particles tagged with ^{38}Cl . Nevertheless the model has sufficient potential to warrant further investigation of its details.

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* Note that this coefficient, defined on p. 107, differs from that used by Perkins et al. (3)

ANISOTROPIC BROWNIAN DIFFUSION IN FREE-MOLECULE COUETTE FLOW*

W. G. N. Slinn**

Results are given for the theory of Brownian motion of large Knudsen number aerosol particles, hosted by linearized, free-molecule Couette flow. The tensorial diffusion coefficient displays the dependence of diffusion on the viscous stress within the gas.

Atmospheric physicists are accustomed to the concept that turbulent diffusion is anisotropic. Recently we reported that Brownian diffusion can also be anisotropic.^(1,2,3) The degree of this anisotropy depends upon the ratio of the viscous stress tensor, $\tilde{\tau}$, to the pressure, p , within the gas. Specifically, for the case of an aerosol particle with large Knudsen number, diffusing in a Chapman-Enskog host gas, the Brownian diffusion coefficient was shown to be the second order tensor.

$$\tilde{D} = D_E [\tilde{I} - \frac{\tilde{\tau}}{5p}] \quad (1)$$

where D_E is the (scalar) Einstein diffusion coefficient and $\tilde{I}$ is the identity or metric sensor.

Since the degree of anisotropy, $\tilde{\tau}/5p$, is necessarily small in order

that the Chapman-Enskog solution to the Boltzmann equation be valid, it is of interest to investigate Brownian diffusion in other, nonuniform gas flows with larger $\tilde{\tau}/p$ values and for which the velocity distribution function for the molecules is known. In this report, a review will be given of our recent studies of Brownian diffusion in free-molecule Couette flow.

Of course, there is no guarantee that $\tilde{D}$ would still depend on $\tilde{\tau}/p$, but it might be expected on the basis of Curie's theorem.⁽⁴⁾ Thus, if Brownian diffusion is dependent on nonuniformities in the gas, then the only measure of the nonuniformity which is a second or higher even-order tensor is the viscous stress. As an aside, it might be useful to remark that, by the same principle, turbulent diffusivity should depend linearly on the Reynolds stress.

To describe the velocity distribution function for the gas molecules in a free-molecule Couette flow,⁽⁵⁾ consider an inertial coordinate system (X', Y', Z') in which two infinite plates are parallel to the $X'-Y'$ plane. The

* This report is based on a portion of an article submitted for publication in The Physics of Fluids.

** A part of the financial support for this work was obtained from Battelle Memorial Institute, Physical Sciences Program.

distance, L , along the Z' axis, between the plates, is assumed to satisfy

$$a \ll L \ll \lambda \quad (2)$$

where a is the radius of the aerosol particle and λ is the mean free path for the molecules of the gas. Inequality (2) ensures that the Knudsen numbers, for both the particle and the flow field, are large compared to unity.

If the upper of the two parallel plates moves in the positive Y' direction with uniform speed U , and if the lower plate is stationary, then using the velocity $\vec{\xi}'$ of the gas molecules with respect to (X', Y', Z') , the velocity distribution function for the gas molecules is

$$f = f_b h(\xi'_z) + f_a [1 - h(\xi'_z)] \quad (3)$$

where the molecules from "below" have the Maxwellian distribution function

$$f_b = (\gamma/\pi)^{3/2} \exp[-\gamma \vec{\xi}' \cdot \vec{\xi}'] \quad (4)$$

and those from "above" have

$$f_a = (\gamma/\pi)^{3/2} \exp[-\gamma (\vec{\xi}' - u \hat{J})^2] \quad (5)$$

In these equations, $\gamma = m/2kT$, where m is the mass of a gas molecule, k is Boltzmann's constant, and T is the common temperature of the plates. The h functions are Heaviside step functions.

There are two aspects of this analysis which limit the usefulness of the results. First, what might be called a "shadow" effect has been

ignored. That is, when the particle approaches within a few radii of one of the plates, it seems that there would be fewer molecules hitting the particle on its "near-plate" side, because of the "shadow" cast by a particle. If this effect were included, the stochastic Langevin and Fokker-Plank equations⁽²⁾ would be considerably more complicated, not to mention the difficulty itself of formulating the effect.

Of more consequence, though, is that we were unable to evaluate some innocuous-looking integrals such as

$$\int_0^a \operatorname{erf}(\lambda \xi) \exp(-\xi^2) d\xi.$$

Because of this, we were forced to abandon the attempt to obtain a result which was valid for all $\tilde{v}/p$. Instead, the results are adequate only to terms linear in $\tilde{v}/p$.

To evaluate the mean and variance of the stochastic force on the particle, it is necessary in our formalism^(2,6,7) to express the velocity distribution function, Equation (3), in a coordinate system (X, Y, Z) moving with the particle. If the velocity of a molecule with respect to the particle is $\vec{\xi}$ and if the velocity of the particle with respect to (X', Y', Z') is $\vec{V}$, then

$$\vec{\xi}' = \vec{\xi} + \vec{V} \quad (6)$$

Now, the desired form for the velocity distribution function can be obtained by substituting Equation (6) into Equations (3) through (5) and by expanding for small $\vec{V} = \vec{V} \gamma^{1/2}$. The

Heaviside functions are extended according to

$$h(\zeta_Z + v_Z) = h(\zeta_Z) + v_Z \delta(\zeta_Z) + O(v^2) \quad (7)$$

where $\zeta = \xi \gamma^{1/2}$ and δ is the Dirac delta function.

The details of the calculation will not be given. The methods are similar to those used in previous reports^(2,6) and are described in the complete paper.

The results for the mean and "variance" of the momentum, $\vec{p}$, delivered during Δt to the moving particle, arising from the collisions of N gas molecules, are:

$$N\langle\vec{p}\rangle = -M\beta \Delta t \left[\vec{V} - \frac{U}{2} \hat{J} + \frac{9}{8} \frac{15}{128} \pi \vec{V} \cdot \frac{\vec{\tau}}{p} \right] \quad (8)$$

$$N\langle\vec{p}\vec{p}\rangle = M^2\beta \Delta t \frac{kT}{M} \left[\hat{I} - \frac{15}{128} \pi \frac{\vec{\tau}}{p} \right] \quad (9)$$

where M is the mass of the particle, β is Epstein's drag coefficient per unit mass and the viscous stress tensor has been identified as:⁽⁵⁾

$$\vec{\tau} = \frac{1}{2} \rho U \bar{c} (\hat{J}\hat{K} + \hat{K}\hat{J}) \quad (10)$$

where ρ is the mass density and $\bar{c} = (8kT/\pi M)^{1/2}$ is the mean thermal speed of the molecules. The last term in Equation (8) gives rise to what we have previously called "stressphoresis."

Equations (8) and (9) are used to obtain a Fokker-Planck equation for the particle. In turn, if this equation is reduced to a Smoluchowski and thence a convective-diffusion equation, then the diffusion coef-

ficient for large Knudsen number particles hosted by a free-molecule, linearized, Couette flow is found to be

$$D = D_E \left[\hat{I} - \frac{13}{4} \left(\frac{15}{128} \right) \pi \frac{\vec{\tau}}{p} \right]. \quad (11)$$

The numerical factor is approximately 1.2, as compared with the factor 0.2 for the continuum, Chapman-Enskog case.

It is reasonable to predict that anisotropy is a universal feature of Brownian diffusion in a nonequilibrium environment. The degree of anisotropy is expected to depend linearly on the viscous stress in the host medium. However to theoretically test this prediction, much work remains to be done, especially for small Knudsen number particles and for the case when the host fluid is a liquid. Toward this end, the recent analysis by Mazo⁽⁸⁾ is important for he has (independently) discovered "stressphoresis" and shown that this phenomenon is a universal property of the motion of arbitrary size particles in any nonequilibrium environment. Unfortunately, though, it is not yet feasible to obtain numerical values from his more general, time-correlation analysis.

On the other hand, the numerical values can be obtained from experiments. In this regard, the experimental observations by Elrick⁽⁹⁾ of anisotropy of the Brownian diffusion of small Knudsen number particles is most interesting. It is hoped that the above result, Equation (11), would be tested experimentally. For this, it might be

useful to point out that the diffusion coefficient is independent of a Galilean coordinate transformation and therefore Equation (11) is valid for, and it may be more convenient to perform, the experiment in which the plates of the Couette flow move in opposite directions with equal speeds.

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A KINETIC THEORY OF DIFFUSELY REFLECTING BROWNIAN PARTICLES*

W. G. N. SLINN**

An analysis is made of the motion of a spherical Brownian particle whose surface can diffusely reflect the molecules of an equilibrium host gas. The analysis is based on Newton's second law and a limiting form of Markoff's method. It is shown, both for specular and diffuse reflections that equipartition of energy is a consequence of the dynamics and randomness of the motion. In addition, it is demonstrated that the diffusion coefficient can depend on the temperature of the particle. The entire analysis is restricted to the case for which the Knudsen number of the particle is large compared to unity.

After the report of our recent study⁽¹⁾ of the precipitation scavenging of submicron particles, Professor S. F. Shen, Cornell University suggested⁽²⁾ that an attempt be made to evaluate the influence on Brownian diffusion, of nonuniformities in the host gas. To this end, we developed a kinetic theory of Brownian motion⁽³⁾ for particles whose Knudsen number (the ratio of the mean free path of the gas molecules to the radius of the particle) is large compared to unity. Subsequently, the effect on Brownian motion of nonuniformities in the host gas⁽⁴⁾ and the influence on the precipitation scavenging of

submicron particles, of the resultant anisotropy of the diffusion,⁽⁵⁾ were evaluated.

As an aside to this study of the effect of nonuniformities in the gas, an attempt was made to describe Brownian diffusion of particles whose surfaces could diffusely reflect the molecules of the host gas. It appeared that in our first analysis, some phenomenon had been overlooked (see comments on page 26 of Reference 4). Recently, Professor R. M. Mazo, University of Oregon, demonstrated to us that correlations between the incident and reflected molecules had been omitted. Because of his assistance, we can now report on a complete theory of Brownian diffusion of diffusely reflecting, large Knudsen number, aerosol particles. Although the analysis can easily be extended to the case of a nonuniform gas (see comments on page 30 of Reference 4), it will be illustrated here only for the case of an equilibrium environment.

* *This report is based on a portion of an article, with the same title, submitted for publication in the Journal of Statistical Physics.*

** *A part of the financial support for this work was obtained from Battelle Memorial Institute, Physical Sciences Program.*

It is assumed that the motion of the Brownian particle (B particle) is adequately described by Newton's second law

$$\vec{F} = M \frac{d\vec{V}}{dt} \quad (1)$$

where M is the mass and $\vec{V}$ is the velocity of the B particle and $\vec{F}$ is the sum of the forces applied to the particle from collision by the gas molecules. What is sought is a statistical description of this stochastic force, $\vec{F}$.

By extending Markoff's method⁽⁶⁾ we have shown⁽⁴⁾ that it is sufficient to calculate the mean $N \langle \vec{p} \rangle$ and the "variance" $N \langle \vec{p} \vec{p} \rangle$ of the momentum, $\vec{p}$ delivered during Δt to the B particle by N gas molecules. In the complete paper, it is shown that the mean is given by

$$N \langle \vec{p} \rangle = - M \vec{V} \beta \Delta t \left[1 + f \frac{\pi}{8} \left(\frac{T_p}{T} \right)^{1/2} \right] \quad (2)$$

where β is Epstein's drag coefficient (per unit mass), T_p and T are the temperatures of the particle and gas, respectively, and f is the fraction of the total number of molecules incident on the sphere that is reflected diffusely. It is assumed that the particle is a perfect thermal conductor and that the diffusely reflected molecules are perfectly accommodated at the surface of the particle. In the complete paper, it is shown also that the "variance" is given by

$$N \langle \vec{p} \vec{p} \rangle = \beta \Delta t M k T \gamma \left[2 - f + f \frac{T_p}{T} + 2 f \frac{\pi}{8} \left(\frac{T_p}{T} \right)^{1/2} \right] \quad (3)$$

where k is Boltzmann's constant and γ is the identity or metric tensor.

Following Chandrasekhar's presentation,⁽⁶⁾ Equations (2) and (3) can be used to obtain a Fokker-Planck equation for diffusely reflecting B particles:

$$\frac{\partial W}{\partial t} + \vec{V} \cdot \nabla_R W = \beta^* \nabla_V \cdot W \vec{V} + \frac{kT}{M \tau^*} \nabla_V^2 W \quad (4)$$

$$\text{where } \beta^* = \beta \left[1 + f \frac{\pi}{8} \left(\frac{T_p}{T} \right) \right]^{1/2} \quad (5)$$

$$\text{and } (\tau^*)^{-1} = \beta \left[1 + \frac{f}{2} \left(\frac{T_p}{T} - 1 \right) + f \frac{\pi}{8} \left(\frac{T_p}{T} \right)^{1/2} \right] \quad (6)$$

In Equation (4), $W(\vec{R}, \vec{V}, t; \vec{R}_0, \vec{V}_0, t)$ $d\vec{R} d\vec{V}$ is the probability that the β particle is within the volume element $d\vec{R} d\vec{V}$ about the phase point $\vec{R}, \vec{V}$ at time t if the particle were known to be at $\vec{R}_0, \vec{V}_0$ at time t_0 . The solution in velocity space to Equation (4), subject to the initial condition

$$W(\vec{V}, t) \rightarrow \delta(\vec{V} - \vec{V}_0) \text{ as } t \rightarrow 0,$$

can be obtained simply by relabeling quantities in Chandrasekhar's solution (his Equation 239):

$$W(\vec{V}, t) = \left[\frac{2\pi kT}{M\tau^*\beta^*} (1 - e^{-2\beta^*t}) \right]^{-\frac{5}{2}} \exp \left\{ \frac{-|\vec{V} - \vec{V}_0 e^{-\beta^*t}|^2}{\frac{2kT}{M\tau^*\beta^*} (1 - e^{-2\beta^*t})} \right\} . \quad (7)$$

Equation (7) is one of the main results of this analysis. Taking into account Equations (5) and (6), it is seen that for $t \gg \beta^{-1}$, Equation (7) predicts that the velocity distribution function for the B particle becomes Maxwellian regardless of the number of molecules which is reflected diffusely, provided that the particle has come to its equilibrium temperature, $T_p = T$. This result, that at equilibrium each degree of freedom of the B particle attains its equipartition value of energy, $kT/2$, is a prediction rather than a postulate of the theory. It follows more or less directly from Newton's second law and the requirements of the central limit theorem. Thus equipartition is a consequence of the dynamics and randomness of the motion and is independent of the specific molecule reflection mechanism under consideration.

The complete phase-space solution to Equation (4) can be obtained from Chandrasekhar's analysis (his Equation 286). For $t \gg \beta^{-1}$, it becomes

$$W(\vec{R}, \vec{V}, t \gg \beta^{-1}) = \frac{W(\vec{V})}{(4\pi D^*t)^{3/2}} \exp \left\{ \frac{-|\vec{R} - \vec{R}_0|^2}{4D^*t} \right\} \quad (8)$$

In Equation (8), a diffusion coefficient has been identified as

$$D^* = \frac{kT}{M\beta^*} \frac{1}{\tau^*\beta^*} = \frac{kT}{M\beta^*} \frac{1 + f/2 (T_p/T - 1) + f \pi/8 (T_p/T)}{\left[1 + f (\pi/8) (T_p/T)^{1/2} \right]} . \quad (9)$$

Two interesting features of Equation (9) are the following. For $T_p \neq T$ and $f \neq 0$, it is seen that the diffusion coefficient depends on the temperature of the particle. This is reasonable because if, for example, the temperature of the particle is greater than the temperature of the gas, then as the diffusely reflected molecules leave the particle's surface, they "kick back" harder than do specularly reflected molecules. Thereby, the diffusion coefficient would increase.

A second prediction from Equation (9) is that for $T_p = T$, regardless of f , Einstein's relation is regained.

$$D = kT/M\beta^* . \quad (10)$$

In turn, this demonstrates that the fluctuation-dissipation theorem⁽⁷⁾ is extendable also to those circumstances for which some parameter, such as f , external to the thermodynamic bath, influences the response of the thermodynamic system to the external perturbation. Perhaps this is a rather surprising result. However, it is not just the fluctuations of the thermodynamic system which are important (for they do not

contain any information about f) but it is the fluctuations of the influence of the thermodynamic system on the external perturber. Further, though, notice that for the non-equilibrium situation, $T_p \neq T$, it would be necessary to know not only β^* to obtain D^* , but also f and T_p .

Finally, it might be of interest to add a few comments about the dependence of the entropy of the parameter f . If Equation (7) is used in Boltzmann's expression for the entropy (or H function) (8)

$$H = \int d\vec{V} W \log W, \quad (11)$$

then an explicit dependence of the entropy on f and T_p can be obtained. This is an illustration of Prigogine's discussion⁽⁹⁾ of the dependence of entropy on "internal coordinates." From an information-theory interpretation of entropy, it is reasonable that the entropy should depend on f when $T_p \neq T$. For example, if $T_p \neq 0$, then introducing a diffusely reflected B particle into a ($T = 0$) beam of molecules will randomize their motion. Thus the entropy of the gas molecules will increase as a function of how many gas molecules are reflected diffusely. A similar phenomenon, discussed by Sommerfeld⁽¹⁰⁾ occurs when a speck of soot is introduced into a chamber filled with electromagnetic radiation. The soot can transform the radiation field into black-body radiation. In analogy to the present result, it can be said that when the radiation field

is not in equilibrium, then the entropy of the system depends on the amount of soot which is present. As a further extension of these concepts, it might be appropriate to determine the dependence of the entropy on the "coordinates" of Maxwell's familiar demon. (11)

Professor R. M. Mazo, University of Oregon contributed significantly to these results by demonstrating to us a method of calculating the correlations between the incident and reflected molecules.

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RADIOISOTOPES AS PARTICLES AND VOLATILES

Radioactive particles and volatile radioactive materials released from operations involving nuclear materials constitute a potential hazard to man through ultimate deposition in lung or other body tissue. Along the route from the generating source to man, particles and vapors react and interact with liquids, solids, and gases, each of which may remove, temporarily retain, or have no effect on these particles or volatile materials. Although data are available to account for particle behavior in some circumstances and to permit prediction of the ultimate fate of a particle or vapor in these circumstances, much remains to be learned about the many variables controlling particle deposition and re-entrainment, and trace level vapor interactions with solids, gases and liquids. Improved understanding of particles and vapor interactions with surfaces will permit better estimates and interpretation of particle deposition and retention in ducts, tubes, surfaces in the open air, lung passageways, in particle samplers, and delivery lines.

An important objective of this research is to establish fundamental relationships among variables controlling deposition through theory and experiments and to develop models which correctly relate the variables. Should phenomena prove too complex to permit mathematical expressions with a theoretical basis to be derived, empirical relationships from the experimental measurements will, nevertheless, prove useful in predicting particle behavior.

WIND TUNNEL FOR PARTICLE DEPOSITION MEASUREMENTS

G. A. Sehmel and L. C. Schwendinian

The turbulent deposition of particles was measured for deposition onto 0.7 cm high plastic grass in a 2 ft × 2 ft cross section wind tunnel. Both the particle fluxes and airborne concentration profiles were measured. Deposition velocities are defined to show the mass transfer resistance for the last 1 cm above the grass surface. These dimensionless deposition velocities, K^+ , are nearly independent of experimental conditions for the present limited range of experimental conditions.

The transport of airborne particles from the bulk carrying stream across boundary layers to a solid surface is a process of considerable theoretical and practical significance. A full understanding of the parameters controlling the deposition of fine particulate material on surfaces would permit calculation of particle depletion from an atmosphere moving over terrain, would allow estimates of particle losses to the wall of ducts and sampling lines, and would clarify the significance of rebounding and re-entrainment of particles in a variety of situations.

Particle deposition has been characterized by a deposition velocity, K , defined as the number of particles deposited per unit area per second per unit average concentration in the gas over the surface. In previous atmospheric oriented research, the average concentration has been measured far from the deposition surface. Consequently, a considerable amount of information is lacking about the detailed mechanisms of particle deposition over vegetation.

The purpose of this study is to determine the deposition behavior of particles on simulated large scale surfaces in controlled laboratory experiments. The experimental relationships between deposition, particle concentration profiles in the air, and turbulence of the carrying airstream are being investigated.

The deposition experiments are being performed in a single pass wind tunnel, 2 ft × 2 ft in cross section, constructed of 1/8 in. steel. The air enters the wind tunnel through a 6 ft × 6 ft bank of high efficiency filters. Immediately downstream is a section for inserting screens and baffles for controlling air turbulence. Monodispersed uranine particles generated with a spinning disc aerosol generator are injected downstream of the screens. The tunnel is next contracted to a 2 ft × 2 ft cross section through a nozzle designed to reduce turbulence.

The test section is 30 ft long. Windows are provided (2 ft 4 in. by 1 ft 9 in.) to introduce the test

specimens with various geometries. Downstream is the blower capable of pulling air through the test section at a maximum flow rate of 31 miles/hr. The air leaving the blower is passed through high efficiency filters.

The deposition surface chosen for initial study is a 0.7 cm high plastic grass. For this surface, the friction velocity, the deposition flux, and the particle concentration profile above the surface are measured.

The friction velocities, u_* , were determined from the velocity profiles measured with a hot wire anemometer. The velocity profile is given by the expression

$$v = \frac{u_*}{k} \ln \frac{z + z_0}{z_0}, \quad (1)$$

where v is the measured velocity at a height z above the grass base, k is von Kármán's constant equal to 0.4, and z_0 is the roughness height. Equation (1) adequately represents the velocity profile up to about 12 cm above the grass. Typical values of u_* and k are shown in Table 1.

TABLE 1. Air Characteristics

Maximum Air Velocity, miles/hr	u_* , cm/sec	z_0 , cm
5	21	0.13
10	42	0.19
15	64	0.23
20	87	0.27
30	137	0.34

The roughness height increases with an increase in velocity. This means that models to describe deposition must be developed in terms of both u , and z_0 .

The deposition flux is measured 20 ft downstream of the converging inlet section of the wind tunnel. A total of 22 ft is covered with grass to ensure a fully developed boundary layer. The deposition flux is measured by removing a 7 in. wide (middle third) $\times$ 8 ft long strip of grass. This strip is cut into smaller 6 in. $\times$ 9 in. sections to determine deposition as a function of length. The grass sections are washed and the solution analyzed fluorimetrically for the total mass deposited.

The airborne particle concentration is measured as a function of height above the grass surface at a distance of 20 ft downstream from the start of the grass. The concentration is determined by impacting the particles on a series of 0.035 in. diameter wires which are placed horizontally and at right angles to the air flow. These wires are covered with a thin layer of petroleum jelly to prevent particle re-entrainment. After impaction, the wires are washed and the solutions analyzed fluorimetrically to determine the mass of uranine.

Computer techniques are used to convert the fluorimeter readings to mass concentrations. These calculations account for the variation of wire impaction efficiency as the velocity changes as indicated by Equation (1). The computer also plots the final results and compares

model predictions with the experimentally observed concentration profiles.

Particle eddy diffusion coefficients as a function of distance above the grass were calculated from the measured concentration gradient and grass deposition flux in the wind tunnel. The basic assumption for these calculations was that an infinite source of particles determined the steady state flux to the grass. The ratio of eddy diffusivity to kinematic viscosity (ϵ/ν or reciprocal Schmidt number) increases from about 30 to 600 for y^+ values from 300 to 10^4 . For distances of about 1 to 20 cm above the grass, the data show that particle eddy diffusion coefficients are less than eddy diffusion coefficients of air momentum. This is qualitatively as anticipated. However, the data suggest that the friction velocity may be influencing the particle eddy diffusion coefficient. Particle diffusion coefficients increase with increasing friction velocity. Additional experiments are needed to confirm this generalization.

The experiments were originally designed to estimate the particle diffusion coefficients in the region away from the surface and to measure the deposition velocity at the grass surface. In these experiments, particles were introduced near the tunnel centerline in order to generate measurable concentration profiles from the centerline to very near the grass surface. It became apparent that high enough concen-

trations of particles near the surface could not be maintained in this mode of particle introduction to permit the deposition velocity to be measured reliably and at the same time permit the particle diffusion coefficient to be estimated from concentration profiles. The deposition velocities have been measured in subsequent experiments under conditions yielding nearly constant particle concentrations for the first few inches above the grass. These conditions were achieved by introducing particles through a distributor located near the wind tunnel floor.

A typical concentration profile is shown in Table 2 for 12 μ diameter particles depositing from air at 15 miles/hr ($u_* = 70$ cm/sec.) The concentrations calculated from the wire collections are shown as a function of the distance above the grass. The concentration is nearly uniform with height since the particles were introduced near the bottom of the wind tunnel

TABLE 2. Particle Concentration Above Grass Surface

Height Above Surface of Grass, cm	Airborne Particle Concentration, Particle/cm ³ $\times 10^4$
0.7	4.8
2.4	5.1
3.5	4.5
4.7	5.8
5.7	5.6
7.0	6.0

A deposition velocity can be defined in terms of the ratio of the particle flux divided by the first measured concentration (0.7 cm above grass surface). For this example

$$K = \frac{2.9 \times 10^{-3} \text{ cm}^{-2} \text{ sec}^{-1}}{4.8 \times 10^{-4} \text{ cm}^{-3}} = 6.0 \text{ cm/sec.} \quad (2)$$

Experimental deposition velocities are shown in Table 3 as a function of particle size for two friction velocities. The deposition velocities at a u_* of 70 cm/sec are nearly independent of particle size. Since the deposition velocities are much greater than the terminal settling velocity, the increase in deposition velocity over the settling velocity is attributed to the effects of eddy diffusion. The last column shows that the deposition velocity increases with an increase in the friction velocity.

TABLE 3. Deposition Velocity to Artificial Grass			
Particle Diameter, μm	Terminal Velocity, cm/sec	K, cm/sec	
		$u_* = 70$	$u_* = 144$
6	0.17	5.4	9.1
12	0.66	6.7	
16	1.2	5.8	

These data for a u_* of 70 cm/sec are a factor of 5 to 8 times greater than the deposition velocities interpolated for 6 μm particles from Chamberlain's data.⁽¹⁾ Closer agreement is obtained for the larger particles.

Particle eddy diffusion coefficients were estimated for the region less than 1 cm above the grass surface. These estimates were used to predict from the measured flux the particle concentration at the first sample location. The results show that the particle eddy diffusivities must be much greater than those that can presently be predicted.⁽²⁾ These high required diffusivities may be caused by the combined effect of gravity and the filtering action of the grass stems. Both this theoretical analysis and the experimental work is continuing.

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PARTICLE DEPOSITION FROM TURBULENT AIR FLOW

G. A. Sehmel and L. C. Schwendiman

A theoretical model was developed to describe the turbulent deposition of particles through the boundary layer adjacent to a deposition surface. In this study, the particle eddy diffusivities are back-calculated from the deposition data. From these calculations it is concluded that particle turbulent diffusivities are greater than those for air momentum transfer in turbulent flow.

The transport of airborne particles from the bulk carrying stream across boundary layers to a solid surface is a process of considerable theoretical and practical significance. Aerosol particles will deposit on a surface due to the action of the turbulent air motions of the carrying stream. The rate of turbulent deposition can be significantly greater than the deposition rates due to effects of gravity, Brownian motion, electrical charge effects, diffusiophoresis, and thermophoresis. A full understanding of the parameters controlling the deposition would also clarify the significance of rebounding and re-entrainment of particles.

It is the purpose of this study to determine the quantitative relationships among the variables governing particle deposition during turbulent flow of air and to establish deposition models supported by the data. The objective of recent studies⁽¹⁾ was to develop an improved analyti-

cal deposition model which is substantiated by the deposition data for vertical tubes.

Particle deposition has been experimentally defined in terms of a deposition velocity, K . For a tube, this velocity is defined as the deposition flux of particles to the wall divided by the average concentration of particles in a circular cross sectional segment of the tube above the deposit. That is,

$$K = \frac{\text{flux}}{C_{\text{avg}}} = \frac{\text{number of particles deposited}/(\text{cm}^2\text{-sec})}{\text{average number of particles}/\text{cm}^3} \quad (1)$$

The deposition velocity has units of length per time.

In existing models the assumptions of the free-flight velocity and/or equality of particle diffusivity to air diffusivity must not be adequate since the model predictions⁽¹⁾ do not agree with the experimental measurements of deposition velocity. However,

these assumptions for free-flight velocity and equality of diffusivity no longer need to be made since data now exist to back calculate the required velocities and diffusivities from measured deposition velocities.

The back-calculated free-flight velocities are a function of the dimensionless relaxation time, τ^+ . The decrease in velocity with an increase in relaxation time can be represented by the least squares correlation,

$$(v_0)^+ = 1.49(\tau^+)^{-0.49}. \quad (2)$$

This is the relationship which should be used to predict the free-flight velocity if equality of eddy diffusivities is assumed for the non-isotropic turbulent boundary layer.

The approach used in developing an effective eddy diffusivity correlation was to assume that particle transport from y^+ of 30 to a^+ was solely by diffusion. For y^+ greater than 30, the concentration was assumed to be uniform or decreased so little that the experimental variations in deposition velocities would not warrant more refined assumptions. To develop the correlation, assumed eddy diffusivity correlations were used to predict deposition velocities. The logarithm of the deposition velocity predicted from the correlation was plotted against the logarithm of the observed deposition velocity. The variance from the 45 degree line representing perfect agreement was

determined for each correlation. The correlation which yielded the minimum variance from the 45 degree line was accepted as the best of those developed.

To develop this correlation, the one-dimensional continuity equation,

$$K^+ = - \frac{\varepsilon}{v} \frac{dC^+}{dy^+} \quad (3)$$

was used to describe particle transport adjacent to the deposition surface. The predicted deposition velocities were calculated from Equation (3). By using iterative procedures, an effective eddy diffusivity correlation was sought which was reasonably simple to use, and the complexity of which was no greater than that warranted by the scatter in existing experimental deposition velocity data.

The correlation developed to predict the effective particle eddy diffusivities is graphically represented in Figure 1 and shows that the diffusivities may be represented as a function of both the distance y^+ and particle relaxation time τ^+ . The diffusivities for y^+ greater than 20 lie along the line,

$$\frac{\varepsilon}{v} = 0.4 y^+, \quad (4)$$

or for y^+ less than 20, in the shaded region given by

$$\frac{\varepsilon}{v} = 0.011(y^+)^{1.1} (\tau^+)^{1.1} \quad (5)$$

which is bounded on top by Equation (4) and on the bottom by

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FIGURE 1. Eddy Diffusivity Distributions near a Pipe Wall

$$\frac{\varepsilon}{v} = \left(\frac{y^+}{10}\right)^3. \quad (6)$$

These particle eddy diffusivities are indeed greater than the eddy diffusivities of air which are approximated by Equation (6).

All three Equations (4,5,6) for effective eddy diffusivities may be used in Equation (3) to predict deposition velocities. This means that up to three continuity equations

must be solved with appropriate matching of the particle fluxes and integration limits.

To calculate deposition velocities, one must first determine on Figure 1 the line represented by Equation (5). This line must have an intercept with either the top or bottom boundary represented by the shaded region. Next, the continuity equation in rectangular coordinates is solved with distance limits of a^+ and the intercept. For greater y^+ distances the additionally required continuity

equations are solved in the cylindrical coordinate system with effective eddy diffusivities given by the respective boundary of the shaded area and finally by Equation (4). For the continuity equation in cylindrical coordinates, the distance integration limit is a y^+ of 30.

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Nomenclature

- a = particle radius, cm
 a^+ = $a u_* / \nu$, dimensionless
 C = average particle concentration per unit tube volume downstream in the tube or local particle concentration, particles/cm³
 C_0 = average particle concentration per unit tube volume upstream or in the tube, particles/cm³
 C^+ = C/C_0 , dimensionless local concentration
 d = particle diameter, assuming spherical shape, cm
 f = Fanning friction factor, dimensionless
 K = deposition velocity, cm/sec (number of particles depositing/(cm²sec)/average number concentration/cm³ of air)
 K^+ = K/u_* , dimensionless
 u_* = friction velocity, $V \sqrt{f/2} = \sqrt{\tau_0/\rho}$, cm/sec
 $(v_0)^+$ = v_0/u_* , dimensionless
 v_0 = particle velocity at start of free-flight distance, cm/sec
 V = average axial velocity in tube, cm/sec
 y = distance from tube wall in rectangular coordinate, cm
 y^+ = distance from tube wall, $y u_* / \nu$, dimensionless
 ϵ = eddy diffusivity, cm²/sec
 μ = gas viscosity, g/(cm sec)
 ν = kinematic viscosity, μ/ρ , cm²/sec
 ρ = gas density, g/cm³
 τ_0 = shear stress at tube wall interface, g force/cm²
 τ = $\rho d^2 V / 18 \mu$, sec⁻¹
 τ^+ = $\tau u_*^2 / \nu$
-

ECOLOGICAL MICROMETEOROLOGY AND CLIMATOLOGY

The area of concern in this program is the study of the ecological importance of energy and moisture exchange at and near soil and organism surfaces. Two parallel studies are simultaneously employed to focus these efforts on appropriate biological phenomena and climatological studies: 1) long term studies of important meteorological facts which complement biological studies of the structure of biotic communities and 2) micrometeorological studies, i.e., detailed, relatively short term studies of energy and moisture transfer in relation to the physics of fluid flow and physiology of organisms, which complement biological studies of a function within biotic communities, or individual organisms. Climatological studies are currently concentrated on determining the magnitude of climatological fluctuations, elevational and topographical alterations in micro climates, and related studies affecting plant phenology. Micrometeorological studies currently are oriented toward identification of important meteorological influences on the water balance of individual organisms and communities, and the effect of topographic or edaphic alterations on biological processes.

PRERECORDED CLIMATOLOGY OF THE HANFORD REGION

W . T. Hinds and J. M. Thorp

Evidence for climatological fluctuations before the start of Weather Bureau cooperative observations in 1923 is being gathered, using growth ring widths of big Sagebrush (Artemisia tridentata). Early indicators seem to be that a dry spell near 1900 ended sooner in the Hanford region than in the Okanogan region, 400 miles north. In the relation of ring width to climate, only 30% of the variance of ring widths is due to winter precipitation.

One of the fundamental precepts of ecological investigations is that the structure of existing communities is a function of their history. To extend our knowledge of the history of the Hanford region beyond the records that started in 1913, we took advantage of the relatively long-lived proclivities of sagebrush. Big sagebrush (Artemisia tridentata) ordinarily lives for 50 to 150 years, puts on an annual growth ring that is usually readily discernible, and even after death decomposes relatively slowly, due to the arid regions it inhabits. It is thus a prime tool for determining past climates, and the Dendrochronology Laboratory at the University of Arizona has developed the botanical knowledge to assure accuracy. Some early results of our application of this technique to the Hanford region are described here.

Our efforts were pointed to two objectives: first, a specification of the sequence of favorable and unfavorable growth years, and second, a quantitative relationship between sagebrush ring widths and standard climatic measurements.

Specimens were collected at four sites. Two sites were on south-facing talus slopes (habitats that the Arizona workers suggested as likely to yield long-lived shrubs) and two in mature stands on level terrain over rather deep soil. So far, we have located older specimens on the flat sites than on the talus slopes, contrary to the experience of the Arizona researchers.

Combining records from living and dead sagebrush, we have established a "master chronology" of growth years beginning in 1862. The chronology indicates that the Hanford region has not experienced a prolonged drought (i.e., 6 or 8 years of substantially subnormal winter precipitation). An interesting, albeit not well established, contrast

* *Ecosystems Department*

between the Arizona workers' results in the Okanogan region, 400 miles north of Hanford, and ours in the Hanford region occurred at the turn of the twentieth century. A period of relatively low growth ended about five years earlier at Hanford than in the Okanogan. This discrepancy may not be real, because our results are based on only 5 specimens, but if real, the difference is quite significant for interpretation and interpolation of tree ring data in other regions.

Quantitative relations (or correlations) between ring width and climate measurements were sought,

using the precipitation occurring during the winter months (the period when moisture accumulates faster than it evaporates) and the mean monthly maximum temperature during the season of active growth, spring. To date, only total winter precipitation, October through February, appears to be very important, accounting for 30% of the variance in ring widths. Without doubt, other important measurements will be found, even though sagebrush is not extremely sensitive to climatic fluctuations in this region, being thoroughly adapted to growing in the Hanford region.

CLIMATE AND PHENOLOGY ON THE ALE RESERVE

W. T. Hinds* and J. M. Thorp

Results of a phenological survey of more than 100 species are described, including an indication that the persistent mountain-valley inversion over the Columbia Basin may speed phenologic progression at 1000 ft compared to the valley floor (600 ft).

After setting up the network of temperature and precipitation sensors on the ALE Reserve, described in the 1968 annual report, the effect of climatic gradients on phenology was examined. In this initial survey, more than 100 species were followed

through their springtime development.

By and large, the results were as anticipated: increasing elevation induced a lag in initiation of reproductive activity, but did not greatly increase the length of time between reproductive phases.

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There was one unanticipated observation: reproductive development seemed to begin earlier at about 1000 ft elevation than at lower elevations. This behavior was observed in diverse species, such as phlox, cheatgrass, lupine, balsam root, and sunflower.

The phenological advancement of plants growing at about 1000 ft elevation is probably a temperature effect; phenology has for many years been successfully related to accumulated temperatures. Nearly every night, a temperature inversion is set up over the Hanford region and this inversion intersects Rattlesnake Mountain on the ALE Reserve at about

1000 ft elevation, causing the night temperatures there to be some 5 to 15 °F warmer than on the valley floor. The daytime temperatures at 1000 ft are, of course, a few degrees cooler than on the valley floor.

Should the warmer night temperatures actually induce more rapid phenological progress, some interesting physiological conclusions should follow. Therefore, the phenological observations will be continued, on a more selective species list, using identified individuals to assure there will be no uncertainties in the data due to missed observations or failure to follow the same individuals.

FOLIAR INTERCEPTION OF PRECIPITATION IN SAGEBRUSH COMMUNITIES

W. T. Hinds and J. M. Thorp

Preliminary results of measurements of precipitation throughfall and nutrient accumulation are presented. Data seem to indicate that potassium is readily available to throughfalling rain, and is probably not due to dissolving minerals in soil particles held on foliage.

Precipitation and its distribution in time and space is of prime importance in arid and semiarid plant communities. Clearly, then, the

effect of overstory plants on understory plants may extend beyond light interception to include precipitation interception. To examine the effect of foliar interference with moisture accumulation, sampling of rainfall under sagebrush was begun in Autumn

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1969. Ten individual (i.e., not members of clumps) mature shrubs were selected at random. Under each shrub, three rain gauges were placed at random, the positioning of the gauges to be re-randomized after each significant period of rainfall, a slight modification of a well-known sampling method.

Results to date are only preliminary, but quite suggestive. Interception is quite dependent upon position under the canopy, as expected. Close to the stem, some gauges have collected only one-tenth the amount of rainfall collected in unobstructed gauges. Farther out from the stem, or under tall shrubs, a much higher fraction falls through the canopy.

Analysis of the water collected under the shrubs compared with unobstructed rainfall is shown in Table 1, from which it is clear that significant alterations in nutrient content of the precipitation are caused by contact with the foliage.

Of special interest is the great increase in potassium in the throughfall. It is not yet entirely clear whether this increase is due to solution of potassium compounds from dust particles on the leaves, or due to removal of potassium from exudates from within the leaves. However, if a value of 2 is assumed for leaf area per unit area canopy cover, and the area of an average leaf is assumed to be about 1 cm^2 , it can be shown that, if all the potassium in the throughfall came from attached soil particles, then the soil weight per leaf is about the same as the leaf weight, a most unlikely prospect in midwinter. Hence, some if not most of the potassium in the throughfall may well result from leaching material exuded or otherwise brought to the leaf surface from within the leaf. However, a great deal remains to be done before the ecological consequences of this facet of mineral cycling becomes clear.

TABLE 1. Composition of Water Samples

	Rainwater, ppm	Foliar Throughfall, ppm
Total N	0.15	0.70
Na	0.4	0.5
P	0.025	0.20
S-SO ₄	0.9	2.4
Mg	<1	<1
Ca	1.9	4.0
K	<0.5	23.0
pH	7.0	6.35

BIOMETEOROLOGICAL STUDIES OF CONTRASTING SLOPE EXPOSURES

W. T. Hinds*

Results of studies in north- and south-facing slopes are described, including phenology (a 30-day Zag was noted in the north) and energy fixation (about 8% more energy was fixed per gram biomass on the south-facing slope than on the north, although about 40% more energy was fixed per unit area on the south slope). Average meteorological conditions over the growing seasons were surprisingly similar, but the contrasts at a given moment were notable.

One of the most important modifiers of the structure and function of plant communities is the alteration of meteorological elements induced by slopes and their exposure. In 1969, a systematic study was begun of some of the changes produced by contrasting exposures. These studies were significantly different from most ecological studies of slope environments, in that the slopes studied did not have a pronounced species differentiation on the contrasting exposure. Species composition is one of the most obvious indicators of micro-environment character. Consequently, most ecological studies of slope environment and its influence on plant communities have been taxonomic in nature. Cheatgrass (Bromus tectorum) was the dominant species on both north- and south-facing slopes of our study site, so that

the effect of environmental alteration on the single species could be examined.

The study site was a series of earth mounds left from construction activities about 15 years ago (the mounds are described in some detail in the annual report for 1968). The meteorological data taken during the spring of 1969 were insolation to the inclined surfaces, air temperature and relative humidity at 10 cm above the surface, and soil temperature at 40 cm below the surface. Biological observations, made by scientists in Terrestrial Ecology, included above-ground biomass (sampled weekly), plant density, seed germinability, and caloric content of the collected samples.

The meteorological results indicated several items of interest. Phenology was delayed by 30 days on the north exposure compared to the south exposure, a much longer delay than usually reported, probably due to the steep (36 degree) slopes we studied. Furthermore, observations

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showed that, at least in salubrious times (as was the winter of 1968-69), most of the shoot production on the north-facing slopes occurs in the spring, in contrast with the south-facing slopes, where shoot production was about equally divided between the autumn and the spring. The extension of the growing season on the north-facing slopes served to de-emphasize considerably the apparent difference in microenvironment felt by the plants. Table 1 shows the average conditions during the spring period of active growth, and the similarity is clear. Also included in the table is the condition observed on a slope during the period of growth on the opposite slope, which makes clear the contrast between the slopes at a given time. It was this contrast that induced the difference in phenologic progression observed on the two slopes.

Energy fixation by the plants growing on the contrasting slopes is shown in Table 2. Included in this table are the estimated and/or measured total short wave incident radia-

tion; the energy fixed per unit area; energy fixed per unit shoot biomass; and efficiency of energy fixation. There was a great deal of variability in the results from vegetation sampling, so close interpretation is impossible. However, there is some indication that the efficiency of energy fixation was lower on the north-facing slope than on the south-facing slope during the autumn period of growth - a very reasonable thing, considering the low solar elevation in October and November that would restrict noticeably the incident radiation on the north-facing slope. The energy fixed per gram of shoot biomass was significantly ($p = 0.05$) less on the north-facing slope than the south-facing slope, a suggestive result for speculation. Further work on biotic response to contrasting slope would be profitable indeed, but it should be carried out on specially constructed slopes so that soil properties (e.g., moisture, nutrients, root penetration) could be properly measured.

TABLE 1. Average Conditions During Spring Growth

Slope Exposure	Temperature, °F			Daytime Saturation Deficit, mb	Daily Insolation, cal cm ⁻²	Interval
	Day	Night	Soil			
North	65	47	54	17	400	North Growing Season
South	64	43	51	17	520	South Growing Season
North	59	43	48	11	270	South Growing Season
'South	69	48	61	19	520	North Growing Season

TABLE 2. Energy Fixation in Contrasting Slope Exposures

Slope Exposure	Season	Incident Radiation		Energy Fixation, per cm^2	Efficiency, %	Energy Fixation per Gram Biomass
		Measure cal cm^{-2}	Calculated, cal cm^{-2}			
North	Autumn		2400	2.7	0.11	
South			8000	21.5	0.27	
North	Spring	17000	16000	30.3	0.18	
South		12000	14000	25	0.21	
North	Total		19000	33	0.17	3700
South			21000	46.5	0.23	3940

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