JUN 20 1960

MASTER

ARF

ARF 3127-12
(Summary Report)

AEC Research and Development Report

ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY

PRELIMINARY STUDIES OF SCAVENGING SYSTEMS
RELATED TO RADIOACTIVE FALLOUT

J. Rosinski and J. Stockham

United States Atomic Energy Commission
Washington, D. C.

DO NOT
PHOTOSTAT

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

ARMOUR RESEARCH FOUNDATION
of
Illinois Institute of Technology
Chicago 16, Illinois

ARF Project C 127

PRELIMINARY STUDIES OF SCAVENGING SYSTEMS
RELATED TO RADIOACTIVE FALLOUT

Report No. ARF 3127-12
(Summary Report)

J. Rosinski and J. Stockham

for

United States Atomic Energy Commission
Washington, D. C.

Copy No. 7

April 29, 1960

ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY

980 001

DISTRIBUTION LIST

Copies of this report are being distributed as follows:

<u>Copy No.</u>	<u>Recipient</u>
1 - 6	Mr. J. Z. Holland, Chief Fallout Studies Branch Division of Biology and Medicine U. S. Atomic Energy Commission Washington 25, D. C.
7 - 11	Chicago Operations Office P. O. Box 59 Lemont, Illinois Attn: Dr. Steven V. White, Director of Research Contracts Division
12	Technical Information Service Extension U. S. Atomic Energy Commission Oak Ridge, Tennessee
13	Alexander, Dr. Lyle T. Soil Conservation Service U. S. Department of Agriculture Beltsville, Maryland
14	Barrett, Dr. Earl W. Department of Meteorology The University of Chicago Chicago 37, Illinois
15	Bolin, Dr. Bert Institute of Meteorology University of Stockholm Lindhagensagatan 124 Stockholm, Sweden
16	Broecker, Dr. Wallace S. Lamont Geological Observatory Columbia University Palisades, New York
17	Cowan, Mr. Fred Los Alamos Scientific Laboratory P. O. Box 1663 Los Alamos, New Mexico
18	DiGiovanni, Mr. H. J. Del Electronics Corporation 521 Homestead Avenue Mt. Vernon, New York
19	Dingle, Dr. Nelson A. Department of Physics University of Michigan Ann Arbor, Michigan

**DO NOT
PHOTOSTAT**

Copy No.	Recipient
20	Dunham, Dr. Charles L. Director, Division of Biology and Medicine U. S. Atomic Energy Commission Washington 25, D. C.
21	Edvarson, Dr. K. Research Institute of National Defense 2nd Division Stockholm 80, Sweden
22	Feldman, Dr. Milton H. Defense Atomic Support Agency Department of Defense Washington 25, D. C.
23	Friedlander, Mr. S. K. Department of Chemical Engineering The Johns Hopkins University Baltimore, Maryland
24	Goeckermann, Dr. Robert University of California Radiation Laboratory Livermore, California
25	Hagemann, Dr. F. T. Argonne National Laboratory P. O. Box 299 Lemont, Illinois
26	Harley, Dr. J. H. Health and Safety Laboratory New York Operations Office U. S. Atomic Energy Commission 70 Columbus Avenue New York 23, New York
27	Harris, Mr. William Health and Safety Laboratory New York Operations Office U. S. Atomic Energy Commission 70 Columbus Avenue New York 23, New York
28	Hartgering, Lt. Col. James B. Office of Chief R & D Department of the Army Room 3D 442 - The Pentagon Washington 25, D. C.
29	Healy, Mr. Jack Hanford Atomic Products Operation Richland, Washington
30	Howell, Dr. Wallace Mount Washington Observatory Gorham, New Hampshire

DO NOT
PHOTOSTAT

Copy No.	Recipient
31	Jones, Mr. Sam P. General Mills, Incorporated 2003 East Hennepin Avenue Minneapolis 13, Minnesota
32	Kellogg, Dr. W. W. The Rand Corporation 1700 Main Street Santa Monica, California
33	Kruger, Dr. Paul Nuclear Science and Engineering Corporation P. O. Box 10901 Pittsburgh 36, Pennsylvania
34	Kuroda, Dr. Paul K. University of Arkansas Fayetteville, Arkansas
35	Langham, Dr. Wright Los Alamos Scientific Laboratory P. O. Box 1663 Los Alamos, New Mexico
36	Larson, Mr. Kermit USAEC Project, UCLA P. O. Box 4164 West Los Angeles, California
37	Leventhal, Mr. Leon Manager, Technical Services Department Tracerlab, Incorporated 2030 Wright Avenue Richmond 3, California
38	Lockhart, Dr. Luther B., Jr. Chemistry Division Naval Research Laboratory Washington 25, D. C.
39	Lough, Dr. S. Allan New York Operations Office U. S. Atomic Energy Commission 376 Hudson Street New York 14, New York
40	Machta, Dr. Lester U. S. Weather Bureau 24th and M, N. W. Washington 25, D. C.
41	Marley, Dr. W. G. Atomic Energy Research Establishment Harwell, Didcot, Berks, England

DO NOT
PHOTOSTAT

Copy No.	Recipient
42	Martell, Dr. Edward A. Air Force Cambridge Research Center Bedford, Massachusetts
43	Moses, Mr. Harry Argonne National Laboratory P. O. Box 299 Lemont, Illinois
44	Northrup, Mr. Doyle L. Technical Director, Headquarters, USAF AFTAC Washington 25, D. C.
45	Phillips, Dr. Charles, Chief Research and Development Division Ft. Detrick, Frederick, Maryland
46	Rafter, Dr. T. A. Department of Scientific and Industrial Research Wellington, New Zealand
47	Reed, Mr. Jack Sandia Corporation Albuquerque, New Mexico
48	Silverman, Dr. L. Harvard University, School of Public Health 55 Shattuck Street Boston 15, Massachusetts
49	Starr, Professor Victor Department of Meteorology Massachusetts Institute of Technology Boston 15, Massachusetts
50	Stern, Mr. Sidney C. 10 Debra Lane Saxonville Post Office Framingham, Massachusetts
51	Suess, Dr. Hans A. Scripps Institution of Oceanography La Jolla, California
52	Turkevich, Dr. John Department of Chemistry Princeton University Princeton, New Jersey
53	Volchok, Dr. Herbert Isotopes, Incorporated 123 Woodland Avenue Westwood, New Jersey

Copy No.

- 54 Weber, Dr. Francis
 U. S. Public Health Service
 Division of Radiological Health
 Washington 25, D. C.
- 55 Whitby, Dr. Kenneth T.
 University of Minnesota
 Minneapolis, Minnesota
- 56 Armour Research Foundation
 Division C Files
- 57 Armour Research Foundation
 Editors, G. S. Gordon, L. C. Kinney,
 F. Schossberger, Main Files
- 58 Armour Research Foundation
 K. W. Miller, Report Library
- 59 - 62 Armour Research Foundation
 J. Rosinski

DO NOT
PHOTOSTAT

FOREWORD

This is the summary report on ARF Project C 127, entitled "Preliminary Studies Related to Radioactive Fallout". The research program was sponsored by the Fallout Studies Branch, Division of Biology and Medicine of the Atomic Energy Commission, under Contract No. AT (11-1)-626. Mr. J. Z. Holland, Chief of the Fallout Studies Branch, and Mr. George Anton were the project officers. The project leader was J. Rosinski. J. Stockham, S. Bernsen, M. Michalchik, G. Langer, C. Nagamoto and M. A. Fisher contributed to the work. This report covers the period from April 1, 1959, to April 29, 1960. All data are recorded in ARF Logbooks C 7980, C 8277, C 8626, C 8793, C 9032, C 9230, C 9560, C 9712, and C 9831.

Approved by:

M. A. Fisher, Supervisor

JR:JS:shy
4/25/60

Respectfully submitted,

Armour Research Foundation
of Illinois Institute of Technology

J. Rosinski,
Senior Engineer

J. Stockham
Associate Chemist

PRELIMINARY STUDIES RELATED TO RADIOACTIVE FALLOUT

ABSTRACT

This program consisted of two related phases. In Phase I, an attempt was made to find the relationship between the amount and nature of radioactivity, particle size distribution and weight of particulate matter present in the lower troposphere. Emphasis was placed on the distribution of strontium-90 and total beta activity. From a limited number of analyses it was found that strontium-90 and total beta activity is associated primarily with particles below approximately 0.1 micron diameter.

Phase II consisted of experimental studies on scavenging of solid particulate matter by water droplets. It was found that water vapor gradient around a condensing droplet promotes scavenging of particles of 1.3 microns and 0.3 micron diameter. The effect of water vapor gradient around an evaporating droplet is still not too well defined.

TABLE OF CONTENTS

	<u>Page</u> viii
ABSTRACT	
I. INTRODUCTION	1
II. PHASE I - AIR SAMPLING	3
A. Andersen Samplers	3
B. Small Cyclone Separator	7
C. Roughing Cyclone Separator	11
D. Results	14
III. PHASE II - SCAVENGING STUDIES	24
A. Test Chamber	24
B. Aerosol Generators	26
C. Results with Zinc Sulfide Aerosols	27
D. Results with Titanium Dioxide Aerosols	36
IV. MISCELLANEOUS EXPERIMENTS	37
A. Atomization of Aerosols	37
B. Activation Analysis	41
C. Calibration of Tri-Carb	43
D. Radioautographs of Snow Crystals	44
V. FUTURE RESEARCH	46
APPENDIX A	47

LIST OF TABLES

1	Classification of Airborne Particles by the Andersen Sampler	3
2	Collection Efficiency of the Small Cyclone Separator	11
3	Collection Efficiency of the Roughing Cyclone	14
4	Radioactivity Associated with Dry Atmospheric Particles Classified by the Andersen Samplers	15
5	Radioactivity Associated with Dry Atmospheric Particles Classified by the Cyclone Separators	23
6	Capture of Zinc Sulfide Particles by Water Droplets	31
7	Capture of Titanium Dioxide Particles by Water Droplets	39
8	Particle Size Distribution Obtained by Atomization of Latex and Gelatin Suspension	42
9	Calibration of Tri-Carb with Tritiated Water	44
A1	New York Operations Office Radioanalyses on Classified Dust	48
A2	New York Operations Office Radioanalyses on Classified Dust	50

ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY

TABLE OF CONTENTS (contd)

LIST OF FIGURES

	<u>Page</u>
1 Details of the Andersen Sampler	4
2 Diagram of the Circuit Used for Sampling Airborne Particles During Dry Periods	6
3 Design of the Small Cyclone Separator	8
4 Relation Between Pressure Drop and Airflow in the Small Cyclone Separator	9
5 Apparatus for Determining Efficiency of the Cyclone Separator	10
6 Design of the Roughing Cyclone Separator	12
7 Relation between Pressure Drop and Airflow in the Roughing Cyclone Separator	13
8 Total Beta Activity of Particulate Matter Classified by the Andersen Samplers and the Cyclone Separator	18
9 Strontium-90 Activity of Particulate Matter Classified by the Andersen Samplers and the Cyclone Separators	19
10 Daily Precipitation at Chicago Midway Airport	20
10A Daily Precipitation at Chicago Midway Airport	21
10B Daily Precipitation at Chicago Midway Airport	22
11 Test Chamber for Scavenging Studies	25
12 Particle Size Distribution of Zinc Sulfide Aerosol Particles and of Aerosol Particles Captured by Water Droplets	29
13 Photomicrographs of Zinc Sulfide Aerosol Particles and of Aerosol Particles Captured by Water Droplets	30
14 Effect of Vapor Gradient on Capture of Zinc Sulfide Particles	35
15 Particle Size Distribution of Titanium Dioxide Aerosol Particles	38
16 Effect of Vapor Gradient on Capture of Titanium Dioxide Particles	40
17 Calibration of Tri-Carb with Tritiated Water	45

PRELIMINARY STUDIES RELATED TO RADIOACTIVE FALLOUT

I. INTRODUCTION

In Phase I of the research program, airborne particles were collected in order to determine the relationship between particle size and radioactivity in dry fallout. This phase of the program is part of the Atomic Energy Commission's routine studies to evaluate the hazards associated with the use of atomic energy. Extensive studies by various investigators have shown that strontium-90 is present primarily in wet fallout and that its contribution to the total activity of dry fallout is minor. Dry fallout is several orders of magnitude less radioactive than wet fallout, but the role and quantity of specific radionuclides in dry fallout needs further clarification.

The airborne particles were classified by size by Armour Research Foundation (ARF), and the various size fractions were analyzed for radioactivity by the New York Operations Office of the Atomic Energy Commission (AEC). Total beta activity and strontium-90 activity were determined. Because strontium-90 is a major health hazard, determination of its radioactivity is essential. Also, inasmuch as decay rates differ, a ratio of strontium-90 to total beta activity provides useful information.

Phase II of the program was a laboratory investigation of the mechanisms by which particles of micron and submicron size are scavenged by a water droplet. Present knowledge indicates that there are at least five mechanisms by which aerosol particles can be captured by a liquid droplet. These are impaction, electrostatic attraction, Brownian motion, thermal gradient, and movement of water vapor in a vapor pressure gradient.

Capture by impaction results from the inertia of particles being carried across the streamlines which are established by movement of air around a

collector, i. e., the droplet. With sufficient inertia the particles strike the droplet and are captured. In addition to inertia, relatively large particles are captured by interception; the surface of a particle need only touch the surface of the collector while the particle is following the streamlines around the collector. Impaction by settling is frequent with very large particles. Capture by impaction is generally limited to particles larger than 1 micron in diameter.

Capture by electrostatic attraction can be a potent mechanism of scavenging. A charge on either or both an aerosol particle and the droplet can drastically alter collection efficiency. Promotion of coagulation of strongly charged aerosols having opposite charges was investigated by a number of scientists and found to be proportional to the product of the charges on the particle, while inhibition of coagulation of strongly charged particles was found to be an exponential function.

Capture by Brownian motion is effective for minute particles when the relative velocity between particle and collector is low. Brownian motion is due to bombardment of aerosol particles by air molecules.

Thermal gradients of the order of $750^{\circ}\text{C}/\text{cm}$ are required for effective particle deposition, but these gradients are not generally found in nature. This mechanism probably does not play a role in the capture of radioactive particles by a water droplet, but in combination with other mechanisms it might be of importance during the formation of liquid droplets.

A vapor pressure gradient surrounding a condensing droplet provides a force field which can carry particles to the surface of a droplet. Likewise, the vapor diffusing from an evaporating droplet can hinder or prevent particle capture. For example, coagulation of hygroscopic aerosols is promoted be-

cause of the reduced pressure near each particle, and the presence of a gradient in the concentration of water vapor causes the water molecules to curve toward the droplet. This creates a flux which is capable of carrying submicron particulate matter to the collector.

II. PHASE I - AIR SAMPLING

A. Andersen Samplers

Andersen¹ samplers were used to classify airborne particles into the six size fractions shown in Table 1.

Table 1

CLASSIFICATION OF AIRBORNE PARTICLES BY THE ANDERSEN SAMPLER

<u>Stage</u>	<u>Particle Size Retained, microns</u>
1	7.0 and larger
2	5.5-10.0
3	3.5-5.5
4	2.0-3.5
5	1.0-2.0
6	< 1.0

The sampler consists of six stages, each with 400 holes. As shown in Fig. 1, these holes become progressively smaller with each stage. Thus, the velocity of the air increases as it passes through the sampler, and the particles are separated by inertia and retained on Petri dishes placed 6.5 mm below each stage. Corn syrup² was placed in each dish to a depth of a few mm to minimize bounceoff and reentrainment. This material is viscous (1132 csu), tacky, and water soluble, and thus suitable for this purpose. The

¹ Andersen, A. J. Bact. 76, 471, 1958.

² Courtesy of Corn Products Refining Co., Summit, Ill.

Schematic diagram of a six-stage Andersen sampler

Fig. 1. DETAILS OF THE ANDERSEN SAMPLER

syrup required only occasional replacement. The material collected on each stage was washed into plastic bottles with hot 5% hydrochloric acid. The acid suspensions were then sent to the New York Operations Office for analysis.

Material passing through the sampler was collected on millipore filters.³ These filters were replaced every few days to maintain a flow close to 1 cfm through the samplers. The operating flow rate of 1 cfm requires long periods of sampling because 100,000 ft³ of air must be processed to obtain reliable data. From January 1960 the millipore filters were weighed so that the weight of collected material would be known.

Two Andersen samplers were placed on a stand 33 in. above the roof of the 3-story Industrial Chemistry Building of ARF. A 4 x 4-ft plywood sheet was placed 10-1/2 in. above the top of the samplers to shield them from rain. Both samplers were operated with a single vacuum pump.

To determine the radioactivity associated with dry fallout only, a lithium chloride switch was fabricated to shut down the pump during periods of precipitation. The switch was made from two carbon discs, 2-1/2 in. in diameter, separated by a 1/32-in. Teflon O gasket. A maximum number of 1/4-in. holes were drilled through the top disc, and the space between the discs was filled with lithium chloride. Electrical connections were made to each disc; the circuitry is shown in Fig. 2. When wet by snow or rain, the current conducted by the salt shorted the circuit and shut off the pump. Heat from the current flow dried the chloride and restarted the pump. A half-hour time lag was allowed for fragments of raindrops to be removed from the atmosphere by settling and evaporation. A timer was used to determine actual sampling time.

³ Millipore Filter Corp., Watertown, Mass.

Fig. 2 DIAGRAM OF THE CIRCUIT USED FOR SAMPLING AIRBORNE PARTICLES DURING DRY PERIODS

B. Small Cyclone Separator

To reduce the time necessary to sample $100,000 \text{ ft}^3$ of air, a cyclone separator was designed to operate at 10 cfm. It was glass blown. For an inlet velocity of 50 fps, the inlet diameter was sized at 20 mm. Other dimensions are shown in Fig. 3; these are based on the recommendations of Perry.⁴ The relation between airflow and pressure drop in the cyclone is shown in Fig. 4.

The particle collection efficiency of the small cyclone was checked with the apparatus in Fig. 5 and the ARF single particle counter.⁵ The counter utilizes light scattering to detect and size particles in an aerosol. Room air was drawn through the cyclone at the rate of 10.3 cfm, and the number of particles in various size fractions entering and leaving the cyclone were examined with the particle counter. The air handling system of the counter was equipped with solenoid valves to permit alternate sampling of the inlet and exit streams at 30-sec intervals. The test results, given in Table 2, show that all particles above 4 microns in diameter were retained completely by the cyclone and particles below 1.5 microns almost completely passed through it.

To capture the particles leaving the separator, a filter holder was fabricated to hold a 9-in. diameter millipore filter. A Staplex high-volume sampler⁶ was used to draw 10 cfm of air through the cyclone and the filter.

The cyclone was connected to the lithium chloride switch so that it would be shut down during periods of precipitation. In addition, a Tork pro-

⁴Perry, J. Chemical Engineers Handbook, p. 1024, 1950. McGraw-Hill Book Co., Inc., N. Y.

⁵Katz, S. et al. Soap and Chem. Specialties, Sept. 1956.

⁶Staplex Corp., Brooklyn, N. Y. Model TFIA.

Fig. 3. DESIGN OF THE SMALL CYCLONE SEPARATOR

Fig. 4. RELATION BETWEEN PRESSURE DROP AND AIRFLOW
IN THE SMALL CYCLONE SEPARATOR

Fig. 5. APPARATUS FOR DETERMINING EFFICIENCY OF THE CYCLONE SEPARATOR

grammer was placed in the circuit so that the cyclone sampled only half the time. This was necessary because the Staplex pump is not designed for continuous duty under the conditions of its use.

Table 2
COLLECTION EFFICIENCY
OF THE SMALL CYCLONE SEPARATOR

Particle Diameter, microns	Number of Particles		% of Particles	
	Inlet Air	Exit Air	Retained	Passed
1.0-1.4	1739	1588	9	91
1.4-2.0	586	467	20	80
2.0-2.8	177	72	59	41
2.8-4.0	74	8	92	8
4.0-5.6	46	0	100	0
5.6-8.0	45	0	100	0

C. Roughing Cyclone Separator

To obtain another size fraction with the cyclone sampling system, a roughing cyclone separator was placed ahead of the small cyclone separator. This cyclone removed the larger particles from the airstream. Its construction is shown in Fig. 6; it also was glassblown. Collection efficiency was determined with the same apparatus used for the small cyclone. The results with room air are shown in Table 3. With Arizona road dust, particles larger than 16 microns were completely retained by the cyclone. The relation between airflow and pressure drop in the roughing cyclone is shown in Fig. 7.

The roughing cyclone was placed in operation on July 2, 1959, and thereafter the amounts of solid particulate matter collected in each cyclone and on the filter were determined. The solids were washed from the cyclones with ethyl alcohol into separate tared dishes. The dishes were then

Fig. 6. DESIGN OF THE ROUGHING CYCLONE SEPARATOR

Fig. 7. RELATION BETWEEN PRESSURE DROP AND AIRFLOW
IN THE ROUGHING CYCLONE SEPARATOR

reweighed after drying at 103°C. The dried material was washed into plastic bottles with 5% hydrochloric acid and shipped to the New York Operations Office for analysis. The filters were weighed before and after each run to determine the weight of collected dust.

Table 3
COLLECTION EFFICIENCY
OF THE ROUGHING CYCLONE

Particle Diameter, microns	Number of Particles		% of Particles	
	Inlet Air	Exit Air	Retained	Passed
1.0-1.4	2389	2126	11	89
1.4-2.0	1403	1084	23	77
2.0-2.8	897	461	48	52
2.8-4.0	676	251	63	37
4.0-5.6	230	61	74	26
5.6-8.0	85	10	88	12

Large millipore filters possess sufficient static electricity to give false weights on a normal beam-type analytical balance. Weighing the filters in a humid environment and in the presence of ionizing radiation permitted accurate results. Constant humidity in the environment of the balance was obtained by using a saturated solution of potassium carbonate. A 1-mc polonium source provided the ionizing radiation.

The roughing cyclone was also connected to the lithium chloride switch so that it would not operate during precipitation.

D. Results

The total beta radioactivity and the strontium-90 radioactivity associated with the dry atmospheric particles classified by the Andersen samplers are shown in Table 4. The total beta activity during the summer of 1958 was 1315 μc / 100,000 ft³ of air. The beta activity rose to about 2000 μc per

Table 4

RADIOACTIVITY ASSOCIATED WITH DRY ATMOSPHERIC PARTICLES
CLASSIFIED BY THE ANDERSEN SAMPLERS

Date	Particle Size Range	Radioactivity, $\mu\text{c}/100,000 \text{ ft}^3$ of air	
		Total Beta	Strontium-90
7/15/58-10/20/58	> 5.5 microns	83	0.7
	1.0-5.5 microns	593	8.3
	< 1.0 microns	76	3.8
	Millipore filter	563	16.0
		<u>1315</u>	<u>28.8</u>
11/18/58-2/13/59	> 7.0	75	1.1
	5.5-10.0 microns	79	4.5
	3.5-5.0 microns	243	3.1
	2.0-3.5 microns	272	3.6
	1.0-2.0 microns	601	11.0
	< 1.0 micron	210	5.7
	Millipore filter	520	10.6
		<u>1999</u>	<u>39.6</u>
2/13/59-5/26/59	> 7.0 microns	a	2.5
	5.5-10.0		1.5
	3.5-5.5 microns		1.9
	2.0-3.5 microns		13.4
	1.0-2.0 microns		15.6
	< 1.0 micron		19.5
	Millipore filter		42.3
			<u>96.7</u>
6/1/59-9/8/59	> 7.0	28	2.1
	5.5-10.0	13	1.1
	3.5-5.5	23	1.2
	2.0-3.5	40	3.3
	1.0-2.0	363	15.1
	< 1.0	261	11.9
	Millipore filter	1207	43.8
		<u>1935</u>	<u>78.5</u>
9/10/59-12/16/59	a		
1/8/59	Run not completed		

^aResults not received as yet.

100,000 ft³ during the winter of 1958-59 and remained about this level during the summer of 1959. The strontium-90 activity was 29 $\mu\mu$ c/100,000 ft³ during the summer of 1958. The activity due to this radionuclide increased through the winter of 1958-59 to a high value of 97 $\mu\mu$ c/100,000 ft³ recorded in the spring of 1959. It then dropped slightly to 78 $\mu\mu$ c/100,000 ft³ during the summer of 1959. The increased strontium-90 activity is largely due to those particles which passed the Andersen samplers and were collected on the millipore filters. These fine particles and the particles in the 1 to 2 micron range were the main contributors to both the strontium-90 and the total beta activity. The strontium-90 activity was approximately 2 to 5% of the total beta activity.

The radioactivity associated with the particles classified by the cyclone separators is shown in Table 5. The total beta activity associated with these particles collected during November and the first two-thirds of December 1958 ranged from 4300 to 5600 $\mu\mu$ c/100,000 ft³. Just before the end of 1958 the total beta activity rose sharply to about 9000 $\mu\mu$ c/100,000 ft³. At the end of January 1959 the total beta activity level was 2600 $\mu\mu$ c/100,000 ft³. The activity was at the same level in May, rose slightly to about 3000 $\mu\mu$ c per 100,000 ft³ in July, and dropped below 1000 $\mu\mu$ c/100,000 ft³ in August 1959. The strontium-90 activity followed a slightly different pattern. It rose continuously from a low value of 22 $\mu\mu$ c/100,000 ft³ in December 1958 to a high of 99 $\mu\mu$ c/100,000 ft³ in July 1959 and then dropped to 40 $\mu\mu$ c per 100,000 ft³ in August. The ratio of strontium-90 activity to total beta activity ranged from 0.4 to 4.8.

As shown with the Andersen samplers, the major share of the activity was contributed by the fine particles which were collected on the millipore filters. Results have been received on six tests in which the collected dust

ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY

was weighed. These results indicate that a gram of material collected in the small cyclone has about 10 times the total beta activity as a gram of material collected in the roughing cyclone. Further, each gram of material collected on the millipore filter is 3 to 4 times as radioactive as a gram of material in the small cyclone. Likewise, each gram of material in the small cyclone has 7 to 9 times the strontium-90 activity as a gram of material in the roughing cyclone. A gram of material from a millipore filter has between 2 to 4 times the strontium-90 activity as a gram of material in the small cyclone. The results show that all the material collected during July had 6990 $\mu\text{c/g}$ per g of total beta activity and during August 2545 $\mu\text{c/g}$. During these two months the strontium-90 activity was 229 and 114.5 c/g , respectively.

The total beta activity from the Andersen samplers and the cyclone separators are plotted against a monthly time scale in Fig. 8. A similar plot of strontium-90 activity is given in Fig. 9. The daily precipitation at Chicago Midway Airport from July 1958 to October 1959 is shown in Fig. 10. The correlation between the samplers was reasonably good except for the total beta activity during the winter of 1958. The cyclone samplers, because of their shorter sampling period, can be expected to show the effect of precipitation on the activity of dry fallout. An inverse relationship between precipitation and activity would be expected because the samplers are shut off during periods of rain when the activity is washed from the atmosphere. Thus, when the rainfall ends and the samplers restart they are sampling cleansed air. Inspection of Table 5 and Fig. 10 reveals evidence of this inverse relationship.

Fig. 8. TOTAL BETA ACTIVITY OF PARTICULATE MATTER CLASSIFIED BY THE ANDERSEN SAMPLERS AND THE CYCLONE SEPARATOR.

Fig. 9. STRONTIUM-90 ACTIVITY OF PARTICULATE MATTER CLASSIFIED BY THE ANDERSEN SAMPLERS AND THE CYCLONE SEPARATORS

Fig. 10 DAILY PRECIPITATION AT CHICAGO MIDWAY AIRPORT

Fig. 10A. DAILY PRECIPITATION AT CHICAGO MIDWAY AIRPORT

Table 5

RADIOACTIVITY ASSOCIATED WITH DRY ATMOSPHERIC PARTICLES
CLASSIFIED BY THE CYCLONE SEPARATORS

Date	Particle Size Separation Unit	Total Beta Activity		Strontium-90 Activity		
		per 10 ⁵ ft ³ c	%	per gram c	per 10 ⁵ ft ³ c	per gram c
11/24/58-12/3/58	Small Cyclone	1730	31.1		6.7	20.4
	Millipore filter	3840	68.9		26.2	79.6
		5570	100.0		32.9	100.0
12/3/58-12/12/58	Small cyclone	1380	32.2		5.8	26.0
	Millipore filter	2900	67.8		16.5	74.0
		4280	100.0		22.3	100.0
12/15/58-12/22/58	Small cyclone	1385	25.5		3.9	13.8
	Millipore filter	4050	74.5		24.4	86.2
		5435	100.0		28.3	100.0
12/22/58-12/29/58	Small cyclone	1483	16.5		7.3	18.1
	Millipore filter	7490	83.5		33.0	81.9
		8973	100.0		40.3	100.0
1/22/59-1/30/59	Small cyclone	635	24.7		10.3	21.0
	Millipore filter	1940	75.3		38.7	29.0
		2575	100.0		49.0	100.0
5/11/59-6/22/59	Small cyclone	313	12.6		6.9	8.6
	Millipore filter	2174	87.4		73.1	91.4
		2487	100.0		80.0	100.0
7/2/59-7/17/59	Roughing cyclone	52.7	1.7	360	2.1	14.2
	Small cyclone	321.0	10.6	3,990	7.6	94.0
	Millipore filter	2665.0	87.7	12,900	89.5	432.0
		3038.7	100.0	6,990	99.2	229.0
7/28/59-8/17/59	Roughing cyclone	11.3	1.2	112	1.1	10.4
	Small cyclone	72.7	8.2	1,231	7.6	94.0
	Millipore filter	806.0	90.6	4,239	33.6	177.0
		890.0	100.0	2,545	40.1	114.5
8/20/59-9/8/59	Roughing cyclone	9.7	4.7	58.4	0.14	0.80
	Small cyclone	16.6	8.0	221.0	0.00	0.0
	Millipore filter	181.5	87.3	1,120	8.47	52.2
		207.8	100.0	510	8.61	21.1
9/11/59-9/30/59	Roughing cyclone	7.8	3.2	60.8	0.82	6.4
	Small cyclone	23.5	9.5	320.0	0.37	5.1
	Millipore filter	216.0	87.3	325.0	10.65	16.1
		247.3	100.0	285.0	11.84	13.7
10/7/59-10/20/59	Roughing cyclone	10.0	5.7	56.5	0.71	4.04
	Small cyclone	45.7	25.9	582	1.53	17.3
	Millipore filter	121.2	68.4	826	a	a
		176.9	100.0	432	-	0.0
10/27/59-11/16/59	Roughing cyclone	9.5	5.4	59.7	0.0	24.2
	Small cyclone	34.1	19.3	426	1.95	24.2
	Millipore filter	134	75.3	658	5.06	24.9
		177.6	100.0	399	7.01	15.8
11/20/59-12/16/59	a					
1/8/59-2/1/60	a					

^aResults with roughing cyclone, small cyclone, and millipore filter not received as yet.

III. PHASE II - SCAVENGING STUDIES

A. Test Chamber

The apparatus used to study scavenging of aerosol particles by evaporating and condensing water droplets is shown in Fig. 11. The tests were conducted in a water-jacketed glass chamber 58 mm in diameter and 17 in. in height. The aerosol entered the bottom of the chamber at a rate of 740 cc/min. and yielded a velocity of 29 cm/min. This velocity matches the terminal fall velocity of a 10-micron particle with a density of 4. A fine screen across the entrance of the chamber eliminated air turbulence and helped distribute the aerosol uniformly in the test chamber. Two ports, each 38 mm in diameter, were located on opposite sides of the chamber about 4 in. above the entrance. One port was slightly higher than the other. A gold wire used to support the water droplet was placed in the higher port, while the lower port facilitated placement of the droplet on the tip of the gold wire and its removal after exposure to the aerosol. About 3 in. above these ports was another port for sampling the aerosol so that its concentration and particle size distribution can be ascertained. The aerosol exited at the top of the chamber, passed through a millipore filter, and was expelled through a vacuum pump. A restrictive orifice placed between the filter and the pump controlled the flow in the test chamber.

The millipore filter and its housing were electrically connected to a Keithley electrometer, model 610. The charge on the particles was discharged through the electrometer and recorded on the meter scale. A grounded metal case surrounding the filter housing shielded it from stray currents. Polystyrene tubing isolated the filter from ground and served as the connecting link between the housing and the grounded metal tubing lead-

Fig. 11. TEST CHAMBER FOR SCAVENGING STUDIES

ing from the test chamber. An oscillating thermal precipitator collected a representative sample of the aerosol. This instrument operated at a flow rate of 20 cc/min; the oscillation period was about 3 min. Under these conditions the aerosol could be sampled during the entire test. The particles were collected on a 3 x 1-in. glass slide. The deposit was about 15 x 55 mm. From this deposit the aerosol concentration and the particle size were determined.

A water droplet was hung in the center of the test chamber from a gold wire 1/50 in. in diameter. The wire was attached to a 1/4 -in. brass rod which went out the test chamber through a rubber stopper. Outside the chamber the rod went through the center of a metal basin. Ice in this basin cooled the droplet and promoted condensation. The metal rod was electrically grounded. The droplet was formed and placed on the tip of the wire from a syringe needle. The change in droplet size was followed with a Leitz binocular low-power microscope with a long focal length. One eyepiece of the microscope was fitted with a Filar 12-1/2 x eyepiece micrometer. The eyepiece was calibrated by placing a millimeter scale in the field of view.

For condensation tests the test chamber was lined with wet blotting paper. The droplet was cooled and the aerosol was humidified by passing it through humid air in a vessel containing warm water. Water at room temperature was circulated through the jacket of the test chamber. The rate of change in the droplet size is a function of droplet temperature and the temperature and the humidity of the aerosol stream.

B. Aerosol Generators

The Wright dust-feed mechanism⁷ was used to generate aerosols from dry powders. In this generator dry powder is rammed into a dust con-

⁷ Adams, Ltd. Miniver Rd., Chase Estate, London, N. W. 10, England.

tainer. The container is rotated downward against a scraper blade by means of a gear assembly. The dust scraped loose is carried by pressurized air through an 0.52-in. diameter jet and impacted against an impaction blade which breaks-up agglomerates. Several gear ratios are available to enable a wide range of aerosol concentrations.

The generator designed by Lauterbach⁸ was used to make aerosols from polystyrene latex and salts of dysprosium. Here, a jet of air is passed across or near the surface of a liquid to atomize fine liquid droplets. When the liquid evaporates, small particles of material are left suspended in the airstream.

C. Results with Zinc Sulfide Aerosols

An aerosol of fluorescent zinc sulfide dust⁹ was used to test the vapor gradient mechanism as a means of capturing particles 1 micron and larger by water droplets. The aerosol was generated with the Wright dust-feed apparatus by using an 8:1 gear ratio and an air pressure of 8 psig.

The number of captured particles was obtained by collecting the droplet after exposure to the aerosol on a black millipore filter and removing the water immediately by applying light suction. Usually this provided a fairly uniform deposit of fluorescent particles on a surface of the filter paper. The particles were placed under a light microscope and counted at 160x magnification; reflected ultraviolet light in the 4000-Å^o region was used to illuminate the particle. In most tests all the particles captured were counted. Where concentrations were high, particles along several diameters were counted and averaged. This count was then multiplied by the ratio of the total area

⁸ Lauterbach, K. A. M. A. Arch. Ind. Health 13, 156, 1956.

⁹ Rosinski, J. et al. Anal. Chem. 28, 486, 1956. Dust #2210.

of the deposit to the counted area to obtain the total number of particles captured.

The size distribution of the aerosol, as shown by the thermal precipitator deposit, is given in Fig. 12. The skewness in the distribution of the larger particles can be attributed to the velocity in the test chamber, which was just sufficient to match the terminal fall velocity of a 10-micron particle. The mean particle diameter was 1.14 microns, and the mean volume diameter was 1.9 microns. The size distribution of the captured particles is also shown in Fig. 12. Skewness is lacking in this distribution because of particle aggregation on the surface of the droplet. Photomicrographs from which these distributions were obtained are shown in Fig. 13.

The results of the zinc sulfide tests are shown in Table 6. The slope refers to the rate of change of the droplet diameter during the test. A negative slope indicates an evaporating droplet and a positive slope a condensing or growing droplet. The slopes were calculated by the method of least squares to obtain the straight-line equation of best fit. The fit of the data to the straight-line equation is shown by the correlation coefficient, r , which is equal to

$$r = \frac{n \sum xy - \sum x \sum y}{\sqrt{[n \sum x^2 - (\sum x)^2] [n \sum y^2 - (\sum y)^2]}}$$

where

n is the number of measurements

x is time variable

y is droplet diameter at time x .

The correlation coefficient can vary between 0 and ± 1 . Perfect correlation

Aerosol Particles

Particles Captured

Fig. 13. PHOTOMICROGRAPHS OF ZINC SULFIDE AEROSOL PARTICLES
AND OF AEROSOL PARTICLES CAPTURED BY WATER DROPLETS

ARMOUR RESEARCH FOUNDATION OF ILLINOIS INSTITUTE OF TECHNOLOGY

Table 6
CAPTURE OF ZINC SULFIDE PARTICLES BY WATER DROPLETS

Growth Curve			Av. Diam. mm	Aerosol Conc. particles per cc	Particles Captured		Theoretical Number of Particles Captured			
Slope, mm/min	Inter- cept	r			Number	%	Inter- ception	Inertia	Brownian Motion	Electro- static
-0.0575	2.083	0.99	1.709	1,300	101	0.90	27	0	0	5
-0.0464	2.199	0.99	1.851	64,000	4801	0.65	1624	0	15	-
-0.0445	2.269	0.99	1.935	193	60	2.4*	5	0	0	0
-0.0393	2.143	0.99	1.907	4,260	351	0.83	111	0	0	53
-0.0380	2.328	0.98	2.106	2,515	500	1.6	59	0	0	24
-0.0353	2.332	0.94	2.120	3,250	124	0.29	82	0	0	43
-0.0305	2.192	0.97	1.863	89	32	3.0*	2	0	0	0
-0.0050	1.814	0.70	1.776	5,740	1103	1.7	156	0	0	103
-0.0013	1.820	0.29	1.811	2,238	455	1.9	58	0	0	21
+0.0017	2.090	0.70	2.100	752	306	3.1	18	0	0	7
+0.0059	1.848	0.56	1.877	3,350	1110	4.3	56	0	0	26
+0.0120	1.620	0.88	1.695	3,550	891	3.2	67	0	0	28
+0.0133	1.948	0.96	2.015	2,715	1751	6.9	48	0	0	20
+0.0153	2.124	0.93	2.201	699	495	6.3	9	0	0	25
+0.0186	1.962	0.98	2.055	3,720	2095	6.1	68	0	0	48

* Droplet deposit ill defined.

is ± 1 ; 0 is no correlation. In general, the fit of the data to the straight line is excellent. The low values of r were obtained when there was little change in droplet size. Here, subtraction of two values which differ by a few thousandths magnifies any errors in calculation. The percent capture is based on the number of particles collected divided by the number of particles which passed beneath the cross-sectional area of the droplet projected in a direction of the aerosol flow during the test period. Based on theoretical considerations, the number of particles captured by four mechanisms is given.

The estimated particle capture by the impaction mechanisms of interception and inertia is based on the equation developed by Ranz and Wong.¹⁰ Impaction arises primarily from inertia which carries a particle across streamlines so that it hits and in many cases sticks to the obstruction, i.e., the water droplet, which has disturbed the streamlines. Interception is a common mechanism for relatively large particles, since it is necessary only for the surface of a particle to come into contact with the collector. A particle touches the collector whenever its center approaches within a distance equal to the radius of the collector. The efficiency of interception for a spherical collector is:

$$n_0 = (1 + R)^2 - 1 / (1 + R) \quad (1)$$

where R is the ratio of the particle's diameter to the collector's diameter. From this equation an interception efficiency of about 0.3% was calculated.

Impaction by inertia was determined from the formula:

$$\eta = \frac{C_p \rho_p V D_p^2}{18 \mu D_c} \quad (2)$$

where

¹⁰

Ranz and Wong. Ind. Eng. Chem. 44, 1371, 1952.

C is an empirical correction factor for resistance of a gas to movement of small particles

ρ_p is the particle density

V is the velocity of the aerosol stream

D_p is the particle diameter

μ is the viscosity of the gas stream

D_c is the collector diameter.

A critical value for Ψ below which impaction by inertia does not occur was found to be 0.042. All values for Ψ in these experiments were on the order of 10^{-4} , well below the critical value.

Zebell¹¹ gives formulae for diffusion by Brownian motion of aerosol particles with radii of r to a larger particle with radius R

$$\frac{dn}{dt} = \frac{W_0}{4} k(R, r) n(r, t) , \quad (3)$$

where

$W_0 = 3.6 \times 10^{-8}$ cm/min for air at 20°C and 1 atm.

$k(R, r)$ is the coagulation function

$n(r, t)$ is the number of r particle/cc at time t.

With the exception of one test when the aerosol concentration was high, no particles were captured by Brownian motion.

Kraemer and Johnstone¹² present data which relate collection parameters to collection efficiency of aerosol particles in electrostatic fields. The collection parameter of interest in these experiments is K_g , which arises

¹¹ Zebell, G. Koll. Zeit. 156. No. 2, 1958.

¹² Kraemer, H. and Johnstone, H. Ind. Eng. Chem. 47, No. 12, 1955.

only when the collector is maintained at constant voltage. It represents the attraction between a charged aerosol particle and a collector which has a charge induced by the surrounding aerosol particles.

$$K_g = \frac{C Q^2 N R^2}{3 \pi \mu D_p V \epsilon D_c} \quad (4)$$

where

C is Cunningham's factor

Q is the charge on the aerosol particle in coulombs

N is the concentration of aerosol particles

R is the radius of the test chamber

μ is the viscosity of air

D_p is the particle diameter

V is the velocity of the aerosol

ϵ is the permittivity of free space, 8.85×10^{-21} coulombs/dyne-cm²

D_c is the collector diameter.

In Fig. 14 the percent of particles captured is plotted against the slope of the growth curve. The plot therefore shows the effect of the vapor gradient of the particle captured. Evaporating droplets gave particle capture below 2%. As the rate of evaporation increased, particle capture decreased slightly. For condensing droplets a slight increase in growth rate produced a large increase in the percent captured. The situation seems to be analogous to coagulation of electrostatically charged aerosols, where promotion of particle capture by condensation increases exponentially but retardation of capture during evaporation, if it does occur, seems to be influenced to a much lesser degree by the evaporation rate.

Fig. 14. EFFECT OF VAPOR GRADIENT
ON CAPTURE OF ZINC SULFIDE PARTICLES

D. Results with Titanium Dioxide Aerosols

An aerosol of titanium dioxide particles¹³ was used to study the capture mechanism of submicron particles. The titanium dioxide used has an anatase crystal structure and is water dispersible. The tests were conducted in a manner similar to that of the zinc sulfide aerosol. The titanium dioxide was dispersed in the Wright dry-dust feeder. Air for the feeder was dried in an electric dryer.¹⁴ Humidity was adjusted by passing the aerosol through a flask containing warm water. The aerosol was then drawn by the vacuum pump through the test chamber, which contained the water droplets hanging from the gold wire. The aerosol concentration was sampled with the thermal precipitator and the charge measured with the electrometer. The change in the water droplet size was followed with the 12.5x Filar eyepiece attached to the telescope. After exposure to the aerosol, the droplet was collected on a white-grid millipore filter and the water removed by applying vacuum.

The number of particles captured was determined by counting the particles in representative areas of the millipore filter deposit under the microscope. A magnification of 250x was generally used. The total area of the deposit was obtained by determining the coordinants of the circumference of the deposit at intervals. This was done under the microscope with the micrometer stage. The circumference was then retraced on graph paper and the area found with a planimeter. The total number of particles captured was therefore the average number counted in a specific area multiplied by the

¹³ Titanox A-WD, Titanium Pigment Corp.

¹⁴ Pittsburgh Lector dryer Co., Pittsburgh, Pa.

ratio of the total area to the area counted. The median diameter of the particles was 0.3 micron, and the mean volume diameter was 0.37 micron. The particle size distribution of the aerosol is shown in Fig. 15. The test results are shown in Table 7. Based on theoretical considerations, impaction by interception, Brownian motion, and electrostatics all contributed to particle capture but to a minor extent. Capture by impaction due to inertia did not occur.

Figure 16, in which percent capture is plotted against the slope of the growth curve, shows a grouping of points with a particle capture of 1 to 3%. In order to define the effect of vapor pressure gradient on particle capture, the data need to be extended to higher evaporation rates and higher condensation rates. Of interest is the test that gave 6% capture with a slope of -0.0115. During the first 3 min of this test the water droplet vibrated intensely from the tip of the gold wire, and it is believed that the turbulence caused by the vibrating droplet resulted in the high particle capture.

IV. MISCELLANEOUS EXPERIMENTS

A. Atomization of Aerosols

Polystyrene latex aerosols suspensions can be made which have essentially monodispersed particles. Two latex suspensions were labeled to contain 0.138-micron spheres and 0.814-micron spheres. Electron photomicrographs of a drop of each of these suspensions showed that these particle sizes were true. An undiluted and several diluted latex suspensions were atomized in the Lauterbach generator. The aerosol was passed through a furnace into a 34-in.-diameter plastic sphere, where it was mixed and diluted with clean air. The aerosol was then sampled with the ARF particle counter. Distilled water atomized in this manner had no particles. The data for the atomization

Fig. 15. PARTICLE SIZE DISTRIBUTION OF TITANIUM DIOXIDE AEROSOL PARTICLES

Table 7

CAPTURE OF TITANIUM DIOXIDE PARTICLES
BY WATER DROPLETS

Growth Curve			Av. Droplet Diam. mm	Aerosol Conc. particles per cc	Particles Captured		Theoretical Number of Particles Captured			
Slope, mm/min	Inter- cept	r			Number	%	Inter- ception	Inertia	Brownian Motion	Electro- static
-0.0387	2.404	0.99	2.212	39,800	35,100	8.0*	222	0	34	44
-0.0333	2.437	0.84	2.270	59,100	18,800	2.8	335	0	50	98
-0.0308	2.273	0.99	2.135	280,000	34,200	1.4	1308	0	216	1764
-0.0284	2.347	0.98	2.178	52,800	6,965	1.2	291	0	45	74
-0.0118	2.127	0.88	2.069	10,964	2,887	2.6	60	0	9	3
-0.0115	2.358	0.92	2.289	10,500	9,052	6.0**	72	0	11	36
-0.0018	2.564	0.46	2.555	151,400	75,700	3.4	970	0	127	892
-0.0005	2.471	0.08	2.468	50,700	16,600	2.1	349	0	47	101
+0.0007	2.254	0.31	2.557	1,145	178	1.4	6	0	1	0
+0.0054	2.541	0.85	2.571	870	858	6.0	6	0	0	0
+0.0074	2.522	0.76	2.562	42,700	15,000	2.3	285	0	36	66
+0.0085	2.506	0.83	2.553	42,800	7,550	1.1	299	0	40	69
+0.0085	2.104	0.86	2.155	18,631	18,976	8.2	120	0	19	14
+0.0237	1.993	0.97	2.123	22,200	4,050	1.6 ⁺	133	0	21	16

* Thermal precipitator not operating properly.

** Droplet vibrated during experiment.

⁺ Droplet deposit ill-defined.

Fig. 16. EFFECT OF VAPOR GRADIENT ON CAPTURE OF TITANIUM DIOXIDE PARTICLES

experiments are given in Table 8. Atomization of the undiluted latex suspension did not produce polystyrene particles of the size indicated by the manufacturer.¹⁵ The counter does not count particles below 0.5 micron. Therefore, no counts with the 0.138-micron polystyrene spheres were expected. However, particles up to 2.8 microns in diameter were found. Heat treatment reduced the number and the size of the aerosol particles, but particles 1 micron in diameter were still present. Results were somewhat improved with the 0.814-micron spheres, although 30 to 50% of the particles were oversized when heat treated. After heat treatment the particles can grow in a humid atmosphere. Heating to 360°F apparently decomposed the particles.

Dr. Van der Hoff of the Dow Chemical Company stated that the latex suspensions contain 7% polystyrene and 2% stabilizer of an undisclosed nature. The stabilizer is present in a quantity sufficient to place a monomolecular layer on 0.3-micron particles. The results in Table 8, however, indicate that the stabilizer is present throughout the liquid.

A 0.1% gelatin suspension was atomized in the Lauterbach generator to show the particle size distribution of the type of material often used to stabilize colloids. It is believed that the oversized particles are due to stabilizer particles, which hold water tenaciously.

B. Activation Analysis

Because of the difficulties associated with microscopic counts, determination of particles captured by neutron irradiation techniques was considered. Dysprosium has a high thermal neutron cross-section approximately 2700 barns and can be detected in amounts as small as 10^{-11} g. The

¹⁵ Dow Chemical Co., Midland, Mich.

Table 8

PARTICLE SIZE DISTRIBUTION OBTAINED BY ATOMIZATION OF LATEX AND GELATIN SUSPENSION

Suspension	Diluting Air, cfm	R.h. of Aerosol, %	Air Temp. after Heater, °F	% of Airborne Particles after Atomization						Number of Airborne Particles/cc
				.5-.7 μ	.7-1 μ	1-1.4 μ	1.4-2 μ	2-2.8 μ	2.8-4 μ	
0.138-% Latex diluted 1:500	0.5	10	75	45	27	17	9	2	0	90
	3.0	4	75	72	19	5	3	1	0	15
	5.5	4	75	90	8	1	1	0	0	4.2
	3.0	4	330	75	25	0	0	0	0	0.2
0.814-% Latex diluted 1:100	3.0	4	75	12	24	52	12	0	0	109
		4	330	58	42	0	0	0	0	0.2
0.814-% Latex diluted 1:10	3.0	4	75	3	13	36	33	13	2	500
		4	250	14	31	36	16	1	0	147
		4	330	57	30	12	1	0	0	3.2
		4	340	55	34	11	0	0	0	2.4
		4	360	0	0	0	0	0	0	0
0.814 - Latex diluted 1:10	3.0	19	330	17	26	42	14	1	0	137
		19	350	33	34	29	4	0	0	52
		100	340	36	37	22	5	0	0	100
0.1% Gelatin	3.0	4	360	(a)	92	8	0	0	0	180
		4	395	(a)	95	5	0	0	0	53
		4	425	0	0	0	0	0	0	0

^aChannel out of order.

gamma decay of dysprosium can be followed on a 236 channel analyzer, and the beta decay of dysprosium chloride (Dy-165 , $\text{Cl}_3\text{-38}$) can be followed with a Tri-Carb liquid scintillation counter. To test the suitability of activation analysis, dysprosium nitrate was purchased and standard water solutions were prepared.

However, irradiation at Argonne National Laboratory was not possible because of refueling of their reactor. Therefore facilities for neutron activation analysis were installed at the ARF reactor and will be used after they have been thoroughly tested and calibrated.

C. Calibration of Tri-Carb

The Packard Tri-Carb liquid scintillation counter was calibrated for counting tritiated water. The tritium can be counted simultaneously with captured radioactive aerosol particles.

One gram of water with one curie of activity was purchased¹⁶ for the preparation of standard samples. This gram of water was diluted with distilled water to give a solution with an activity of $1 \mu\text{c/ml}$; 0.1 ml of this solution was further diluted to 100 ml to yield an activity of $1000 \mu\text{c/ml}$. The scintillation solution contained 4 g/liter of 25 diphenyl oxazole (PPO), 1 g/liter of 1,4-bis-2-(5-phenyloxazolyl)-benzene (POPOP), 100 ml of absolute ethanol, and 900 ml of toluene. Six standards were then prepared containing 20, 40, 80, 120, 160, and 200 $\mu\text{c/ml}$. Each standard contained 0.005 ml of water in a total of 25 ml of scintillation solution. These standards were placed in 22-ml vials and counted at -10°C in the Tri-Carb counter.

¹⁶ New England Nuclear Corp., (Boston, Mass.)

The counting time was 30 min with the discriminator level at 10, 50, and 100 volts. Results are shown in Table 9 and Fig. 17.

Table 9

CALIBRATION OF TRI-CARB WITH TRITIATED WATER

<u>Activity,</u> <u>$\mu\mu$ c/ml</u>	<u>Total Activity</u> <u>in Vial, $\mu\mu$ c</u>	<u>DPM</u>	<u>Tri-Carb,</u> <u>CPM*</u>	<u>Counting</u> <u>Efficiency, %</u>
20	440	968	92	9.5
40	880	1936	155	8.0
80	1760	3872	330	8.5
120	2640	5808	518	8.9
160	3520	6644	686	10.3
200	4400	9680	828	8.5

* Above blank.

D. Radioautographs of Snow Crystals

An attempt was made to obtain a radioautograph of fallout from radioactive debris in snow crystals. The purpose was to determine whether specific radioactivity sites occur and, if they do, whether they are associated with crystal growth sites.

Snow flakes were collected on x-ray films and placed in a freezer for a period of time to allow for exposure from any radioactivity that might be present. Regular dental x-ray films were used. They were taped to a brass plate with an aluminum foil cover. The plates were kept in a freezer until collection. Snow covering about half of the film was collected. A second plate with film was placed over the film on which the snow was collected. The two plates were quickly taped together and placed back in the freezer. Snow already on the ground and falling snow were collected.

The first film was developed 23 days after collection with a GE Super-mix X-ray Developer. Development time was 3 min; fresh, clean solutions

Fig. 17. CALIBRATION OF TRI-CARB WITH TRITIATED WATER

were used. No definite exposure patterns could be observed. Under the microscope there were too many dust particles on the film itself to single out any small exposed areas. Therefore a technique of keeping the films dust free must be devised. Films developed after 30 days of exposure also gave no trace of radioactive exposure. The exposure time should probably be increased.

V. FUTURE RESEARCH

Air sampling during rainless periods was discontinued with both the Andersen samplers and the cyclone separators. In view of the results obtained, separate analysis of the data will be made to better explain and understand the relationship between total beta radioactivity, strontium-90, and particle size distribution and weight associated with particulate matter present in the lower part of the troposphere. The problem of sampling and size separation will be analyzed and recommendations will be made for future experimental work in this area of research.

Laboratory studies on collection mechanisms of aerosol particles by water droplets will be continued. The emphasis will be placed on the investigation of the role of water vapor flux present around a condensing or evaporating water droplet. Scavenging of $150\text{-}\text{\AA}$ diameter gold particles by an evaporating water droplet will be investigated using the falling drop technique which is being developed. Scavenging by a condensing droplet will be studied for $150\text{ }\text{\AA}$, 0.3 and 1.3 micron diameter particles. The number of particles collected will be determined by the most suitable analytical technique. Neutron activation analysis and radioactive tracers will be used where applicable.

APPENDIX A

Analyses of Classified Dust

Table A1

NEW YORK OPERATIONS OFFICE RADIOANALYSES
ON CLASSIFIED DUST
(Andersen Samplers)

Date	Air Vol. Sampled cu ft	Sample Location*	Particle Size Range	HASL No.	Radioactivity, dpm	
					Total Beta	Strontium-90
7/15/58-10/20/58	102,480	E	> 5.5 μ	8,867	290 $\pm$ 116 ⁺	1.1 $\pm$ 6.6
				8,869		
			1.0-5.5 μ	8,871	1143 $\pm$ 124	16.1 $\pm$ 2.2
				8,873		
				8,875		
	102,279	W	< 1.0 μ	8,877	55 $\pm$ 141	0.4 $\pm$ 1.3
			Millipore filter	8,879	1193 $\pm$ 150	32.8 $\pm$ 1.8
			> 5.5 μ	8,868	83 $\pm$ 121	2.0 $\pm$ 1.4
				8,870		
			1.0-5.5 μ	8,872	1523 $\pm$ 157	21.6 $\pm$ 2.2
11/18/58-2/13/59	96,756	E		8,874		
				8,876		
			< 1.0 μ	8,878	286 $\pm$ 133	16.6 $\pm$ 2.7
			Millipore filter	8,880	1340 $\pm$ 162	39.4 $\pm$ 1.9
			> 7.0 μ	9,535	137 $\pm$ 12	1.7 $\pm$ 0.8
			5.5-10.0 μ	9,537	195 $\pm$ 16	< 0.7
			3.5-5.0 μ	9,539	138 $\pm$ 9	3.8 $\pm$ 0.9
			2.0-3.5 μ	9,541	545 $\pm$ 25	8.7 $\pm$ 0.7
			1.0-2.0 μ	9,543	1880 $\pm$ 60	24.2 $\pm$ 2.9
			< 1.0 μ	9,545	490 $\pm$ 22	13.7 $\pm$ 1.6
	104,025	W	Millipore filter	9,548	1080 $\pm$ 90	21.7 $\pm$ 1.3
			> 7.0 μ	9,536	194 $\pm$ 16	3.2 $\pm$ 0.6
			5.5-10.0 μ	9,538	148 $\pm$ 12	2.0 $\pm$ 1.1
			3.5-5.0 μ	9,540	965 $\pm$ 35	10.0 $\pm$ 1.2
			2.0-3.5 μ	9,542	430 $\pm$ 19	7.2 $\pm$ 1.1
			1.0-2.0 μ	9,544	728 $\pm$ 25	24.4 $\pm$ 2.5

Table A1 (contd)

Date	Air Vol. Sampled cu ft	Sample * Location	Particle Size Range	HASL No.	Radioactivity, dpm	
					Total Beta	Strontium-90
2/13/59-5/26/59	102,524	E	< 1.0 μ	9,546	438 $\pm$ 18	11.4 $\pm$ 1.1
			Millipore filter	9,549	1215 $\pm$ 90	25.2 $\pm$ 1.7
			> 7.0 μ	10,605		7.4 $\pm$ 0.24
			5.5-10.0 μ	10,606		17.83 $\pm$ 0.17
			3.5-5.5 μ	10,607		2.99 $\pm$ 0.26
			2.0-3.5 μ	10,608		24.4 $\pm$ 1.0
			1.0-2.0 μ	10,609		25.1 $\pm$ 1.2
	103,059	W	< 1.0 μ	10,610		44.6 $\pm$ 0.5
			Millipore filter	10,604		96.8 $\pm$ 1.5
			> 7.0 μ	10,612		3.84 $\pm$ 0.24
			5.5-10.0 μ	10,613		5.09 $\pm$ 0.25
			3.5-5.5 μ	10,614		5.44 $\pm$ 0.27
			2.0-3.5 μ	10,615		13.5 $\pm$ 0.4
			1.0-2.0 μ	10,616		45.8 $\pm$ 0.5
6/1/59-9/8/59	103,988	E	< 1.0 μ	10,617		43.6 $\pm$ 0.5
			Millipore filter	10,611		99.5 $\pm$ 4.5
			> 7.0 μ	11,244	76.2 $\pm$ 65	4.59 $\pm$ 0.60
			5.5-10.0 μ	11,245	40.0 $\pm$ 5.7	3.54 $\pm$ 0.70
			3.5-5.5 μ	11,246	51.2 $\pm$ 5.9	3.19 $\pm$ 1.72
			2.0-3.5 μ	11,247	120 $\pm$ 82	37.86 $\pm$ 2.44
			1.0-2.0 μ	11,248	972 $\pm$ 20	37.86 $\pm$ 2.44
	102,088	W	< 1.0 μ	11,249	546 $\pm$ 11	27.13 $\pm$ 1.86
			Millipore filter	11,243	3105 $\pm$ 49	102.81 $\pm$ 2.43
			> 7.0 μ	11,251	50.6 $\pm$ 5.8	4.77 $\pm$ 0.67
			5.5-10.0 μ	11,252	18.1 $\pm$ 6.7	1.42 $\pm$ 0.52
			3.5-5.5 μ	11,253	52.2 $\pm$ 7.5	2.35 $\pm$ 1.67
			2.0-3.5 μ	11,254	60.8 $\pm$ 8.0	4.41 $\pm$ 1.24
			1.0-2.0 μ	11,255	668 $\pm$ 12	30.72 $\pm$ 2.49
			< 1.0 μ	11,256	638 $\pm$ 11	26.82 $\pm$ 1.78
			Millipore filter	11,250	2598 $\pm$ 42	98.24 $\pm$ 2.49

* East (E) sampler, 6 in. to the east of other sampler.

Table A2

NEW YORK OPERATIONS OFFICE RADIOANALYSES
ON CLASSIFIED DUST
(Cyclone Samplers)

Date	Air Vol. Sampled cu ft	Particle Size Separation Unit	HASL No.	Radioactivity, dpm	
				Total Beta	Strontium - 90
11/24/58-12/3/58	97,111	Small cyclone	9,061	3,588 + 189	14.3 + 1.8
		Millipore filter	9,047	8,198 + 241	55.8 + 2.7
12/3/58-12/12/58	114,584	Small cyclone	9,082	3,471 + 184	14.6 + 1.7
		Millipore filter	9,080	7,290 + 245	41.5 + 2.0
12/15/58-12/22/58	108,969	Small cyclone	9,108	3,307 + 262	9.3 + 2.8
		Millipore filter	9,106	9,660 + 368	58.2 + 2.6
12/22/58-12/29/58	91,310	Small cyclone	9,109	2,969 + 238	14.5 + 1.7
		Millipore filter	9,163	15,002 + 1010	66.1 + 2.8
1/22/59-1/30/59	98,619	Small cyclone	9,547		20.6 + 1.2
		Millipore filter	9,515		78 + 5
5/11/59-6/22/59	95,000	Small cyclone	10,855	730 + 170 580 + 150	17.0 + 2.3 12.1 + 1.8
		Millipore filter	10,797	4,544 + 36	152.89 + 3.43
7/2/59-7/17/59	103,581	Roughing cyclone	11,057	100 + 60 140 + 70	5.4 + 1.8 4.1 + 2.1
		Small cyclone	11,058	700 + 130 760 + 140	14.2 + 2.1 20.2 + 3.8
		Millipore filter	11,037	6,071 + 73	204 + 3.80

Table A2 (contd)

Date	Air Vol. Sampled cu. ft	Particle Size Separation Unit	HASL No.	Radioactivity, dpm	
				Total Beta	
7/28/58-8/17/59	98,176	Roughing cyclone	11,258	24.5 + 60	2.27 + 0.71
		Small cyclone	11,257	157 + 79	11.72 + 1.16
		Millipore filter	11,291	1,740 + 37	72.55 + 2.74
8/20/59-9/8/59	100,590	Roughing cyclone	11,648	19 + 5	0.6 + 0.9
				24 + 6	0 + 0.9
		Small cyclone	11,649	38 + 6	0 + 0.8
				36 + 7	0 + 1.4
		Millipore filter	11,560	374 + 15	20.7 + 3.0
				431 + 17	16.8 + 2.4
9/11/59-9/30/59	109,503	Roughing cyclone	11,650	23 + 6	3.9 + 2.7
				14 + 5	0.2 + 0.8
		Small cyclone	11,651	61 + 6	0.9 + 1.8
				53 + 6	0.8 + 1.6
		Millipore filter	11,561	507 + 20	26.4 + 4.1
				542 + 21	25.7 + 4.2
10/7/59-10/20/59	127,590	Roughing cyclone	11,905	27 + 7	0.3 + 2.1
				28 + 7	0 + 1.0
		Small cyclone	11,097	117 + 9	6.2 + 1.5
				142 + 11	2.4 + 2.6
		Millipore filter	11,909	369 + 15	
				317 + 13	
10/27/59-11/16/59	104,262	Roughing cyclone	11,906	26 + 7	0 + 0.6
				17 + 6	0 + 1.5
		Small cyclone	11,908	79 + 8	3.6 + 2.1
				78 + 8	5.4 + 2.9
		Millipore filter	11,910	319 + 13	11.2 + 1.8
				297 + 14	12.1 + 2.1