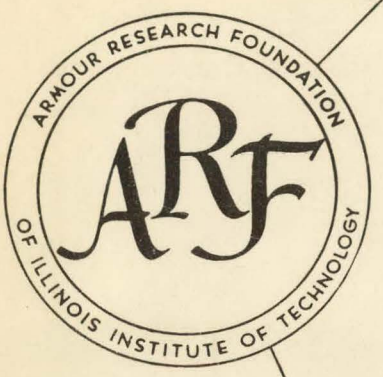
ARF 3127-6
(~~Final~~ Report)

Summary



PRELIMINARY STUDIES OF SCAVENGING SYSTEMS RELATED TO RADIOACTIVE FALLOUT

J. Rosinski and J. Stockham

Atomic Energy Commission
Washington, D. C.

Contract No. AT(11-1)-626

DISCLAIMER

Armour Research Foundation
of
Illinois Institute of Technology
Technology Center
Chicago 16, Illinois

ARF Project C 127
Contract No. AT(11-1)-626

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J. Rosinski and J. Stockham

for

Atomic Energy Commission
Division of Biology and Medicine
Washington 25, D. C.

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April 30, 1959

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FOREWORD

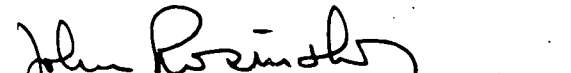
Under Contract No. AT(11-1)-626, ARF Project C 127, Armour Research Foundation conducted a program to determine (1) the importance of Brownian motion, water vapor diffusion, and sticking probability on the scavenging of submicron particles by liquid droplets and (2) the relationship between the size of particles and radioactivity in dry fallout. This program was sponsored by the Atomic Energy Commission and covered the period from April 1, 1958, to March 31, 1959.

Personnel who contributed to the work were M. A. Fisher, R. Snow, S. Radner, G. Langer, and C. Nagamoto.

Data are recorded in ARF Logbooks C 7980, C 8277, C 8626, and C 8793.

Respectfully submitted,

ARMOUR RESEARCH FOUNDATION
of Illinois Institute of Technology



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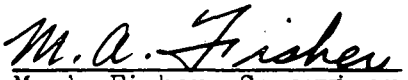

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Fine Particles Research

JR/JS/djb

PRELIMINARY STUDIES OF SCAVENGING SYSTEMS RELATED TO RADIOACTIVE FALLOUT

ABSTRACT

This program involved two related phases. In Phase I, preliminary studies were made of the relationship between the size of particles suspended in the lower atmosphere and the amount and nature of radionuclides they contain. Emphasis was placed on the distribution of strontium-90. From a limited number of analyses, it was found that strontium-90 is associated primarily with particles below 0.1 micron in diameter.

In Phase II, preliminary studies were made of scavenging of particles by liquid water droplets. This phase included studies of sticking probability and the effects of Brownian motion and water vapor diffusion. It was found that electrostatic effects are of primary importance for 1.9-micron (mean volume diameter) particles. Brownian motion and water vapor diffusion did not contribute to the scavenging. These results are based on known and new equations derived for various scavenging conditions.

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PRELIMINARY STUDIES OF SCAVENGING SYSTEMS RELATED TO RADIOACTIVE FALLOUT

I. INTRODUCTION

This final report on ARF Project C 127 covers the period from April 1, 1958, to March 31, 1959. The project consisted of two phases:

I. Preliminary experiments to relate the sizes of particles in air to specific radioisotopes

II. Preliminary laboratory studies of scavenging of particles by liquid drops, including studies of sticking probability and effects of Brownian motion and water vapor diffusion.

Phase I is primarily concerned with the relationship between particle size and radioactivity in dry fallout. Total radioactivity and the contribution of strontium-90 to the total are desired for preliminary studies. Later work will include other radioisotopes. It is well established that wet fallout is several orders of magnitude more radioactive than dry fallout, but the role and the quantity of radionuclides in dry fallout need further study.

Particle size separations were made with Andersen samplers and a cyclone separator. Analyses were performed by the New York Operations Office of the Atomic Energy Commission.

Phase II is concerned with the mechanism by which particles are captured by liquid droplets which transport them to earth. This mechanism is of interest from the standpoints of health and of atom bomb monitoring. There appear to be at least four ways by which liquid droplets can capture particles. These are:

1. Impaction - As a droplet falls through the atmosphere, suspended particles in positions coinciding with the streamlines of the droplet are collected by impaction. This mechanism is predominant for micron-size particles.

2. Brownian Motion - The random motion of air molecules drives particles to a droplet surface. This mechanism is predominant for submicron particles.

3. Electrostatic Attraction - A charge on a droplet produces an electrostatic field, which attracts particles to its surface and holds them long enough to produce strong adhesion. It is possible that a charge on the surface of a droplet also reduces the vapor pressure and thus promotes condensation.

4. Vapor Pressure Gradient - The vapor pressure gradient surrounding a condensing water droplet forces particles to the surface of the droplet. The particle size range for this mechanism has not been established.

II. THEORY OF SCAVENGING MECHANISMS

A. Impaction

Ranz and Wong[1] formulated dimensionless parameters which characterize the forces which cause impaction of a moving particle from an aerosol stream onto a collecting surface. These parameters are defined as follows:

$$\text{Inertia} = \psi = C \rho_p V_0 d_p^2 / 18 \mu d_c , \quad (1)$$

$$\text{Interception} = R = d_p / d_c , \quad (2)$$

$$\text{Settling} = G = (g_L d_c / V_0^2) \psi , \quad (3)$$

where

C is an empirical dimensionless correction factor for resistance of a gas to movement of small particles, and equals approximately 1 when $0.1 < 2f/d_p < 134$

ρ_p is the particle density, g/cm^3

V_0 is the velocity of the aerosol stream, cm/sec

d_p is the particle diameter, cm

μ is the gas viscosity, poise

d_c is the diameter of the collector, cm

g_L is the absolute value of gravity, cm/sec^2

f is the mean free path of a gas molecule, cm.

ψ is the ratio of the force necessary to stop a particle initially traveling at velocity V_0 in distance $d_c/2$, to the fluid resistance at a relative particle velocity of V_0. R is the ratio of the diameter of the particle to the diameter of the collector. G is the ratio of the force of gravity to the fluid resistance at a relative particle velocity of V_0. It is

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[1] Ranz, W. E. and Wong, J. B. Ind. Eng. Chem. 6, 1371, 1952.

also the ratio of the free settling velocity of the particle to the stream velocity.

The efficiency, e, of the collector is defined as the ratio of the cross sectional area of the unobstructed aerosol stream from which all particles are collected to the projected area of the collector normal to the direction of flow. The efficiency of interception, e_{int}, is given by the equation:

$$e_{int} = (1 + R)^2 - 1/(1 + R) . \qquad (4)$$

The efficiency of impaction caused by settling, e_s, is the number of particles per unit volume of gas times the terminal velocity of the particle times the cross sectional area of the collector projected in a vertical direction, divided by the number of particles per unit volume of gas times the gas velocity times the cross sectional area of the collector projected in the direction of flow.

Langmuir and Blodgett[2] reported that the lowest value of ψ at which collection occurs is 1/24 (0.0417). Ranz and Wong[1] showed graphically the relationship between the square root of the inertia parameter and collection efficiency. The efficiency of the above parameters is negligible for any mechanism when the parameter characterizing that mechanism is less than 10^{-2} and is unity when the parameter is unity.

B. Brownian Motion

Whytlaw-Gray and Patterson,[3] following the method of Smoluchowski,[4] gave the following equation for coagulation of aerosol particles by Brownian motion:

$$-\frac{dn}{dt} = s \pi (D_p + D_c)(r_p + r_c) n_p n_c , \qquad (5)$$

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[2]Langmuir, I. and Blodgett, K. B. General Electric Research Lab., Schenectady, N. Y., Rept. RL-225, 1944-45.

[3]Whytlaw-Gray, R. and Patterson, H. Smoke, 57, 1932. Edw. Arnold and Co., London.

[4]Smoluchowski, M. V. Z. physik. Chem. 92, 129, 1916-18.

where

t is time

s is a factor accounting for the sphere of influence of particles and collectors and equals 2 when particles brought into contact by Brownian motion coagulate

D_p and D_c are diffusion coefficients of the particle and the collector

r_p and r_c are the radii of the particle and the collector

n_p and n_c are the concentrations of the particle and the collector.

The sum of the diffusion coefficients is given by the expression:

$$D_p + D_c = \frac{R\,T}{6\,\pi\,\eta\,N}\left[\frac{1 + A\,(f/r_p)}{r_p} + \frac{1 + A\,(f/r_c)}{r_c}\right], \qquad (6)$$

where

R is the gas constant, 8.3×10^7 ergs/°C/mole

T is absolute temperature, 293°C

η is the viscosity of the air, 1.82×10^4 poises

N is Avogadro's number, 6.1×10^{23}

A is a constant equal to 0.9

f is the mean free path of air molecules, about 10^{-5} cm.

Greenfield[5] analyzed the scavenging problem in the light of the equations for impaction and for coagulation by Brownian motion. He concluded that impaction between raindrops and particles does not effect efficient removal of submicron particles, but that the process of coagulation, which takes place before rain, accounts for the removal. His work indicates that particles larger than 10 microns are nearly completely removed by impaction and particles smaller than 0.01 micron are nearly completely removed by coagulation due to Brownian motion. Particles between 0.01 and 10 microns

[5]Greenfield, S. M. Rand Corp., USAEC Contract No. AT(11-1)-3135, P-883-AEC, 1956.

are scavenged less completely by these mechanisms, and particles between 0.1 and 1 micron are poorly scavenged.

Zebel[6] presented the following equation for the diffusion of an aerosol to a single spherical collector:

$$-\frac{dn}{dt} = 4\pi (D_p + D_c)(r_p + r_c) n_p , \qquad (7)$$

where the terms are defined as in Eq. 5. He obtained a quantity, ω_0, proportional to the diffusion coefficient, and a coagulation function, K, and substituted them into Eq. 7 to obtain:

$$-\frac{dn}{dt} = \frac{\omega_0}{4} K_{(r_p,r_c)} n_p \qquad (8)$$

and

$$\omega_0 = \frac{8kT}{3\eta} = 3.6 \times 10^{-8} \text{ cm}^3/\text{min for air of } 20^\circ\text{C and 1 atm}, \qquad (9)$$

where

k is Boltzmann's constant

T is absolute temperature.

Also,

$$K_{(r_p,r_c)} = \left[\frac{1}{r_p}\left(1 + \frac{1}{r_p}(0.1 + 0.0333^{-10.83r_p})\right) + \frac{1}{r_c}\left(1 + \frac{1}{r_c}(0.1 + 0.0333^{-10.83r_c})\right)\right](r_p + r_c). \qquad (10)$$

By use of Eq. 8 the number of collisions due to Brownian motion between small aerosol particles and a single large collector can be calculated.

C. Electrostatic Attraction

Kraemer and Johnstone[7] presented curves which related collection parameters to collection efficiency, e, of aerosol particles in electrostatic

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[6] Zebel, G. Kolloid-Z. 156, (2), 102, 1958.

[7] Kraemer, H. F. and Johnstone, H. F. Ind. Eng. Chem. 47, 2426, 1955.

fields. The differential equations describing the forces contained the collection parameters K_I, K_E, K_S, K_G, and K_M as constants. K_I is the force caused by an electrical charge on the collecting sphere and by the resulting image in the aerosol particle. K_E is the coulombic attraction between a charged collector and a charged particle. The remaining parameters, K_S, K_G, and K_M, are zero except when the aerosols are charged. K_S is the repulsion of the surrounding aerosol particle on the particle being deposited. K_G, which arises only when the collector is maintained at constant voltage, is the attraction between a charged aerosol particle and the collector, which has a charge induced by the surrounding aerosol particles. K_M is the attraction between the aerosol particle and its own image in the collector.

D. Vapor Pressure Gradient

Facy[8] observed the movement of aerosol particles in a gradient of vapor pressure. He found that a water droplet captures particles by this mechanism during the condensation phase and is surrounded by a "dust-free" space during the evaporation phase. His equations for the motion of particles in a vapor pressure gradient are similar to those developed for thermal gradients. Facy found two categories for the velocity of particles in a vapor pressure gradient: one for particles small in comparison to the mean free path and the other for particles large in comparison to the mean free path. In these two categories particle velocity is independent of particle size.

The radius of the dust-free space is given by the expression:

$$S = K \frac{(\delta_R - \delta_{r_0}) R D}{(R - r_0) n}, \qquad (11)$$

[8]Facy, L. Geofisica Pura E Applicata 40, 217, 1958.

where

S is the radius of the dust-free space from the center of the droplet

K is a constant

δ_R is the vapor density at infinity

δ_{r_0} is the vapor density at the droplet surface

R is the radius of the test chamber

r_0 is the droplet radius

D is the coefficient of molecular diffusion for vapor

n is the concentration of aerosol.

For a condensing droplet, Facy obtained the following expression for force, F, which must be added to the molecular kinetic forces:

$$F = \frac{kT}{n} \frac{dn}{dx} , \qquad (12)$$

where

k is Boltzmann's constant

T is absolute temperature

dn/dx is the gradient of aerosol concentration at distance x from the center of the droplet.

In the case of a particle larger than the mean free path (10^{-5} cm), the force due to vapor pressure is:

$$F = K' \, 2 \, \pi \, r \, D \frac{dp}{dx} , \qquad (13)$$

where

K' is a constant

r is the radius of the particle

dp/dx is the vapor pressure gradient.

The velocity, V, of a particle subjected to the above force is given by the expression:

$$V = \frac{K' 2\pi r D}{6\pi \eta r} \frac{dp}{dx} = K' D \frac{dp}{dx} . \qquad (14)$$

Equation 14 shows that the velocity is independent of particles size -- a phenomenon which Facy noted.

Facy observed a velocity of 3×10^{-3} cm/sec for 2-micron particles subjected to a partial pressure gradient of 10 mm Hg/cm at normal atmospheric pressure and corresponding temperatures. He found that fall velocity for 2-micron particles with a density of about 1.2 is of the order of 30×10^{-3} cm/sec. According to Facy, the velocities of fall and motion due to vapor pressure gradient are about the same order of magnitude for particles of 0.2-0.6 micron. Because the vapor pressure gradient is extremely steep in the vicinity of the droplet, Facy observed a velocity of 10^{-2} cm/sec for large particles and a velocity larger than the velocity of fall for 1-micron-diameter particles.

III. MATHEMATICAL MODEL FOR SCAVENGING BY VAPOR DIFFUSION

An attempt was made to obtain and solve an equation which predicts the conditions under which bulk vapor motion is important in scavenging. Equation 15 was derived from the kinetic theory of gases to express the number of particles dragged toward a condensing droplet by the motion of condensing water vapor. It is similar to the equation derived by Facy[8] from an analogy of Einstein's theory of repulsion by heat radiation. Facy's equation is for the steady state and is not applicable to the collection of dust particles by water vapor condensing on droplets because of different boundary conditions.

$$N = N_w/N_g \, \bar{c} \, C = 4 \, D_w \, C \frac{\partial C_w}{\partial x} , \tag{15}$$

where

N is the net number of dust particles which move past a unit cross sectional area per unit of time

N_w is the net number of water molecules which diffuse in the same manner

N_g is the number of collisions of all gas molecules with the same unit area in unit time, as given by the kinetic gas theory

$\bar{c}$ is the average kinetic gas theory velocity of dust particles

C is the concentration of dust particles

D_w is the diffusion coefficient for water vapor in air

C_w is the concentration of water vapor in air

x is distance in the direction of motion.

Equation 15 was added to the usual diffusion equation to give:

$$\frac{\partial C}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(D \, r^2 \frac{\partial C}{\partial r} \right) + \frac{1}{r^2} \frac{\partial}{\partial r} \left(D_w \, r^2 \, C \frac{\partial C_w}{\partial r} \right) , \tag{16}$$

where

r is the radial distance from the center of the water droplet to the dust particle, cm

t is time, sec.

The first term in Eq. 16 represents the change in concentration of dust particles in time due to Brownian motion, and the second term the change due to diffusing water molecules.

A simultaneous equation holds for water vapor only:

$$\frac{\partial C_w}{\partial t} = D_w \frac{1}{r^2} \frac{1}{\partial r} \left(r^2 \frac{\partial C_w}{\partial r} \right) . \tag{17}$$

By substituting the known solution for Eq. 17 into Eq. 16 and neglecting several terms that are unimportant after 0.001 sec, the differential Eq. 16 for the concentration distribution of dust becomes:

$$\frac{\partial C}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(D\, r^2 \frac{\partial C}{\partial r} \right) + \frac{A}{r^2} \frac{\partial C}{\partial r} , \tag{18}$$

where, as illustrated in Fig. 1,

$C = C_{(r,t)}$, dust concentration at any position r and time t, particles, cm^3

$A = 4\, D_w\, R\, (C_w^{\circ} - C_w^{*})$

D_w is the diffusion coefficient for water vapor in air, approximately 0.256 cm^2/sec

R is the radius of the droplet, cm

C_w° is the initial mole fraction of water vapor in the surrounding air

C_w^{*} is the mole fraction of water vapor in equilibrium at the droplet surface

D is the diffustion coefficient for dust particles in air due to Brownian motion, approximately 5.9×10^{-5} cm^2/sec.

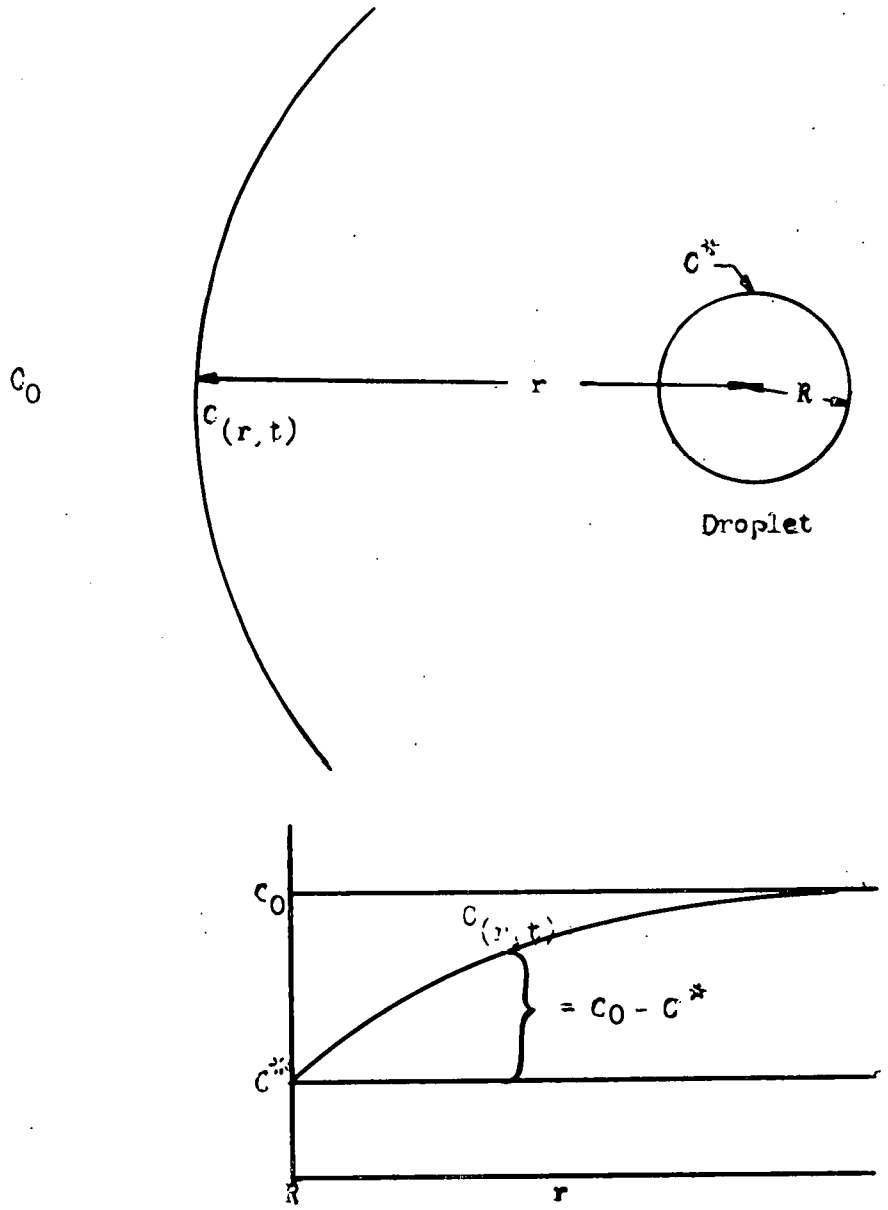

Fig. 1. GRAPHICAL REPRESENTATION OF EQ. 18

Application of the method of separation of variables to Eq. 18 yields a solution which is an exponential function of t multiplied by a function of r alone. By the method of series, the function of r is:

$$\frac{C_{(r,t)} - C^*}{C_0 - C^*} = M \sum_i e^{-\lambda_i t} \left\{ r^3 - \frac{A}{D} r^4 + \left[-\frac{D}{\lambda_i} + \left(\frac{A}{D}\right)^2 r^5 \right] + \frac{A}{D} \left[2 \frac{D}{\lambda_i} - \left(\frac{A}{D}\right)^2 \right] r^6 + \left[\left(\frac{D}{\lambda_i}\right)^2 - 3 \frac{D}{i} \left(\frac{A}{D}\right)^4 r^7 \right] \ldots \right\} \quad (19)$$

where M and λ are constants.

A representative value of A/D is 170 cm. The λ_i values have to be determined from the boundary conditions:

$$\frac{C_{(R,t)}}{C_0} = 0 . \quad (20)$$

The constant M has to be evaluated from the initial conditions:

$$\frac{C_{(r,0)}}{C_0} = 1 , \quad (21)$$

where t equals zero. Unfortunately, the above series converges slowly and many terms must be evaluated to obtain numerical results.

The following substitution was tried in an attempt to obtain a solution:

$$C = \frac{1}{x} \frac{D}{A} Z T , \quad (22)$$

where

T is the exponential function of t alone

Z is a function of r alone, expressed in terms of x

x is a new, independent variable replacing r.

The differential equation for Z becomes:

$$\frac{D^2 Z}{dx^2} + \frac{1}{x^2}\frac{dZ}{dx} + \left(b - \frac{1}{x^3}\right) Z = 0 , \qquad (23)$$

where $b = \frac{\lambda}{D}\left(\frac{A}{D}\right)^2$.

This equation was simplified into a standard form in order to find its solution in a textbook. Since a solution was not found, the equation should be solved by using a computer.

The first problem to be considered is one in which a small droplet containing an ice crystal is introduced into a still aerosol. The aerosol concentration should be small enough so that agglomeration is relatively slow. For a given droplet size, a given water vapor pressure gradient between the droplet and the surrounding air, and a given aerosol diffusivity, the equation can be solved to give the flux of dust particles at the droplet surface.

$$J = 4 \pi D R^2 \frac{dC}{dr}\bigg|_{r = R.} \qquad (24)$$

As Smoluchowski[4] explained, Eq. 24 is valid as long as other condensing nuclei on the walls of the container are far enough away not to affect the dust concentration near the droplet. In general, the flux, J, varies with time. When integrated over the length of an experiment, it gives the total number of dust particles which strike at the droplet due to Brownian motion and to condensation of water. A similar relation holds for the case in which water is evaporated.

IV. EQUIPMENT AND PROCEDURES

A. Andersen Samplers for Air Sampling

An Andersen[9] sampler (Andersen Samplers and Consulting Service, Provo, Utah) was used to separate airborne particles into the six size fractions shown in Table 1. The sampler consists of six stages, each with 400 holes. As shown in Fig. 2, 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 an inertia effect and retained on Petri dishes. The bottom of the dishes is 6.5 mm below each stage. Corn syrup (Corn Products Refining Company, Summit, Illinois) was placed in each dish to prevent bounceoff and reentrainment. This material is viscous (1132 C.S.U.), tacky, and water soluble, and thus suitable for this purpose. The syrup required only occasional replacement. Material passing the sampler was collected on 47-mm millipore filters (Millipore Filter Corporation, Watertown, Massachusetts). The millipore filters were replaced every few days to maintain a flow close to 1 cfm.

Table 1

SIZE CLASSIFICATION OF AIRBORNE PARTICLES BY THE ANDERSEN SAMPLER

Stage	Particle Size Retained, microns
1	>7
2	5.5-10
3	3.5-5.5
4	2-3.5
5	1-2
6	<1

[9] Andersen, A. J. Bact. 76, 471, 1958.

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The sampler operates at a flow rate of 1 cfm. This low flow rate is a disadvantage because 100,000 ft^3 of air must be sampled to obtain reliable radioactivity data.

Two Andersen samplers were placed 33 in. above the roof of the 3-story ARF Industrial Chemistry Building. A 48-$in.^2$ plywood sheet placed 10-1/2 in. above the top of the sampler shielded them from direct hits by sporadic raindrops. Both samplers were operated with a single vacuum pump.

Because it was desired to determine the radioactivity associated with dry fallout, a lithium chloride switch was fabricated to shut off the pump during rain. Initially, the switch consisted of two strands of 32-gage platinum wire wound alternately about a 1 x 2 x 1/8-in. ceramic core. The wires were separated by a 1-mm space which was filled with lithium chloride. When wet, the current conducted by the chloride shorted the circuit and shut off the pump. Heat from the current flow dried the chloride and restarted the pump. Although the response time was rapid, the switch required considerable maintenance and occasional replacement.

A more rugged switch was made from 2 carbon discs, 2-1/4 in. in diameter, separated by a 1/32-in. Teflon O gasket. The space between the discs was filled with lithium chloride. Holes drilled through the top disc admitted rain to shut off the samplers. Response time of the discs was slower than that of the wires but was adequate, and maintenance was nil. A sketch of the circuit is shown in Fig. 3. A timer was used to determine actual sampling periods.

B. Cyclone Separator for Air Sampling

To reduce the time necessary to sample 100,000 ft^3 of air, a cyclone separator was designed to operate at 10 cfm. It was glassblown. For an inlet velocity of 50 ft/sec, the inlet diameter was sized at 20 mm. Other dimensions

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are shown in Fig. 4; these are based on the size of the inlet and the recommendations of Perry.[10] The relation between airflow and pressure drop through the cyclone separator is shown in Fig. 5.

By using the Rosin, Rammler, and Intelmann equation[10] with values of 2 for the particle density and 5 for the number of turns, the minimum particle retained completely by the separator was calculated to be 2.6 microns. This value was checked with the apparatus shown in Fig. 6. Room air was drawn through the separator at the rate of 10.3 cfm, and the number of particles in various size fractions entering and leaving the separator was determined with the ARF particle counter.[11] The counter utilizes light scattering to obtain the distribution of particle sizes in an aerosol. It was equipped with solenoid valves to permit alternate sampling of the inlet and exit streams at 30-sec intervals.

The test results, in Table 2, show that all particles above 4 microns were retained by the cyclone separator 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 9-in.-diameter millipore filters. The filter holder was attached to the separator and the assembled apparatus as shown in Fig. 7. The pump is a Staplex high volume, model TFIA sampler (Staplex Corporation, Brooklyn, New York). The apparatus was placed on the roof near the Andersen samplers and connected to the timer and the lithium chloride cell.

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[10] Perry, J. Chemical Engineers' Handbook, p. 1024, 1950. McGraw-Hill Book Co., New York.

[11] Katz, S. et al. Soap and Chem. Specialties, Sept. 1956.

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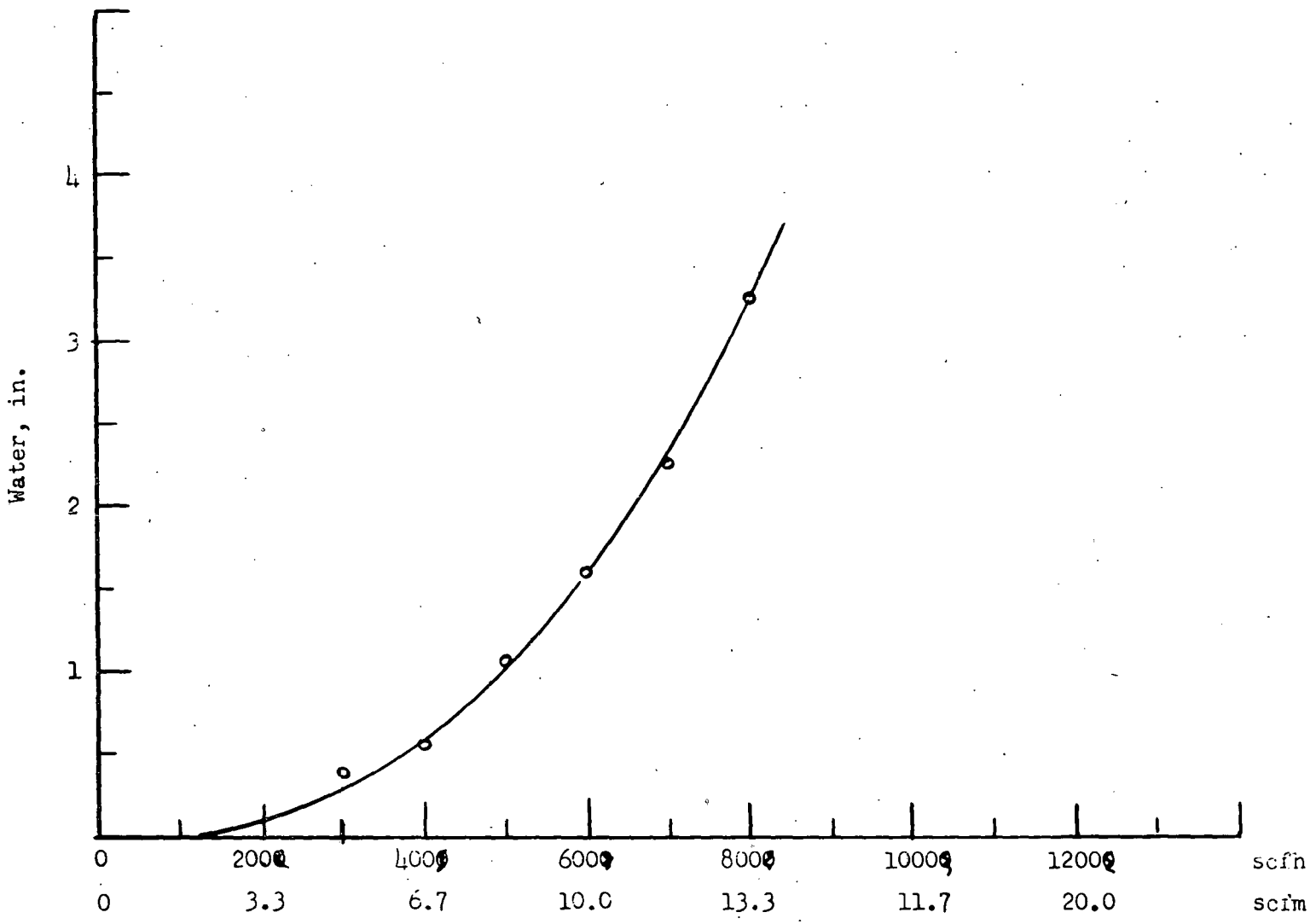

Fig. 5. PRESSURE DROP ACROSS THE CYCLONE SEPARATOR VERSUS FLOW RATE

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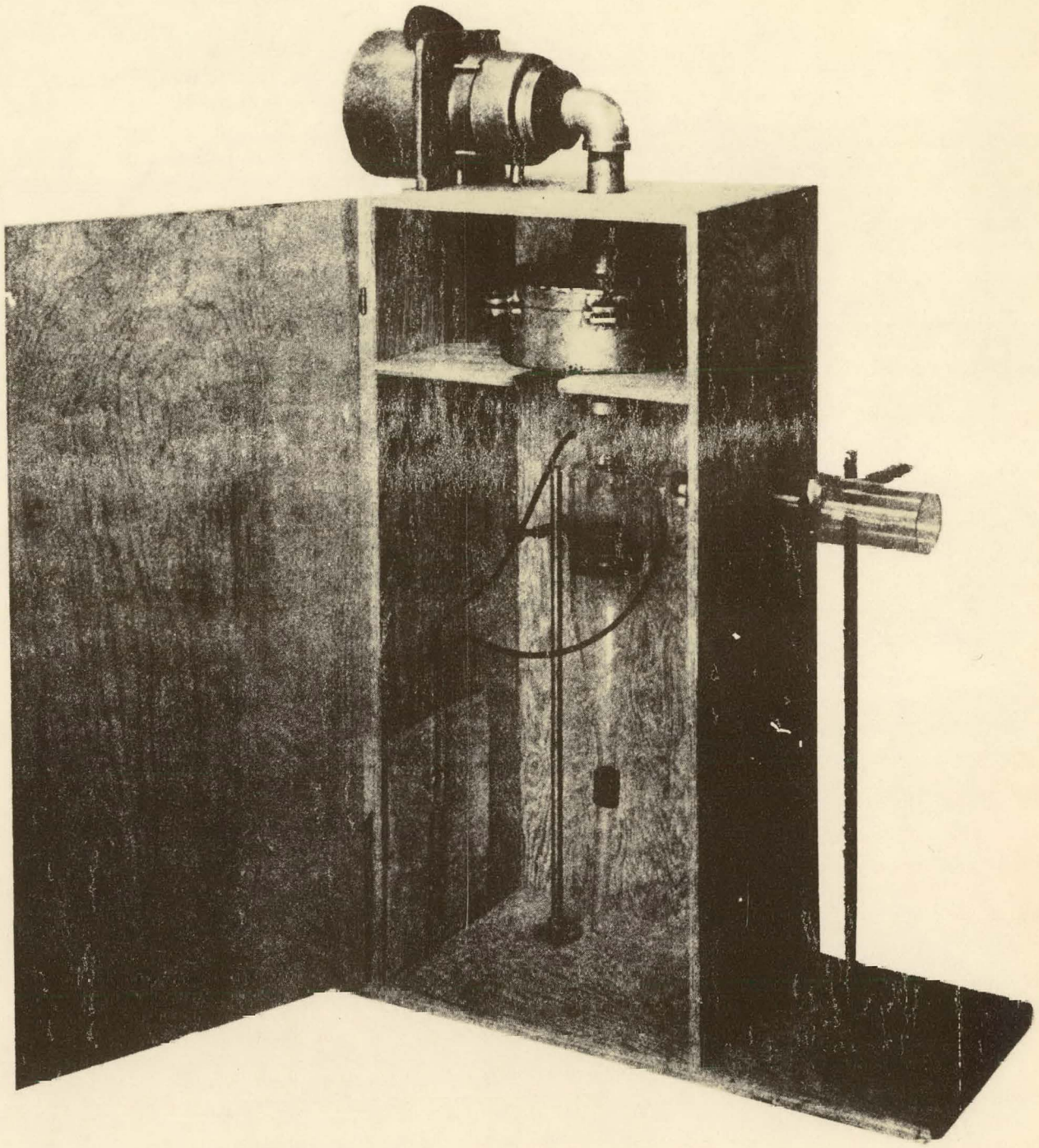

Fig. 7. CYCLONE SEPARATOR ASSEMBLY FOR CLASSIFYING AIRBORNE PARTICLES

Table 2

PARTICLE RETENTION EFFICIENCY OF THE CYCLONE SEPARATOR

	Particle Diameter, microns								
	1-1.4	1.4-2	2-2.8	2.8-4	4-5.6	5.6-8	8-16	16-32	32-64
Number of particles during 30-sec sampling period in inlet stream[a]	319	104	37	14	8	5	0	0	0
	283	95	18	7	4	7	0	0	0
	201	68	23	12	7	3	0	0	0
	184	68	24	12	6	2	0	0	0
	264	108	33	12	8	13	0	0	0
	264	72	15	8	8	6	0	0	0
	224	71	27	9	5	9	0	0	0
	1739	586	177	74	46	45	0	0	0
Number of particles during 30-sec sampling period in exit stream[a]	362	90	11	0	0	0	0	0	0
	300	92	14	1	0	0	0	0	0
	219	72	16	1	0	0	0	0	0
	202	57	9	0	0	0	0	0	0
	184	45	7	3	0	0	0	0	0
	163	54	9	0	0	0	0	0	0
	158	57	6	3	0	0	0	0	0
	1588	467	72	8	0	0	0	0	0
Retention, %	9	20	59	92	100	100	-	-	-

[a]The two airstreams were sampled alternately.

C. Equipment and Procedures for Scavenging

The equipment used to study particle scavenging by vapor pressure gradient is shown in Fig. 8. Aerosols were generated from dry powders placed in a test tube. The particles were suspended by directing a jet of air at 5-psig pressure into the powder through a 0.3-mm opening. The suspended particles were diluted with an additional quantity of air and delivered to the cyclone mixer. There the aerosol was mixed with humid air when condensing conditions were required. The humid air was obtained by passing air through an impinger containing water. The impinger was placed in a temperature-controlled water bath. By adjusting the temperature, a wide range of humidity conditions was possible.

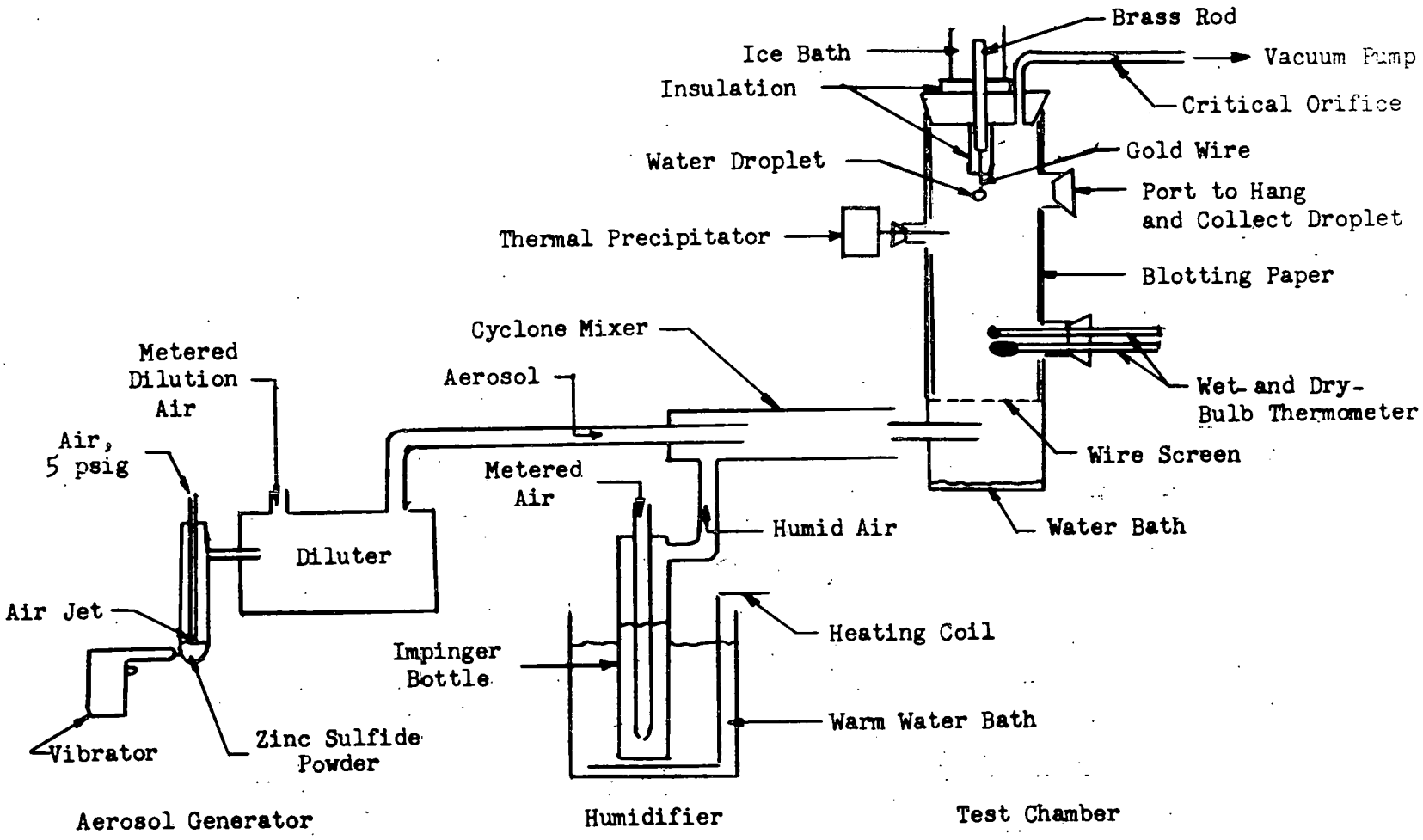

Fig. 8. EQUIPMENT USED TO STUDY SCAVENGING BY VAPOR PRESSURE GRADIENT

A portion of the aerosol was drawn upward through the test chamber by a vacuum pump. The test chamber was a glass tube 100 mm in diameter and 17 in. high. Flow was controlled by a critical orifice. A wire screen placed just about the point where the aerosol entered the chamber provided sufficient resistance to the flow so that the aerosol was uniformly distributed. Side ports were provided for sampling the aerosol and measuring the humidity with wet- and dry-bulb thermometers. Aerosol samples were taken with a thermal precipitator. The thermometers used were unmatched, and therefore the humidity readings were only approximate. For tests with condensing droplets, the wall of the chamber was covered with wet blotting paper.

The water droplets were formed by means of a 1-cc syringe and a 27-gage stainless steel hyperdermic needle. Once formed, the droplet was hung from a gold wire 1/50 in. in diameter. The gold wire was, in turn, attached to a 1/4-in. brass rod which went through a rubber stopper to the outside of the chamber. For condensation tests, the exterior portion of the rod was cooled by immersion in an ice bath. The droplet was cooled by conduction along the rod and the wire. The portion of the rod within the chamber and all but 1/3 in. of the wire were covered with cork to prevent thermal currents and condensation on the rod.

The change in droplet size was followed by means of a Leitz binocular, long focal length, low power microscope fitted with a Filar 12.5x eyepiece micrometer.

After the test, the drop was collected on a black millipore filter and the water removed by applying light suction. The deposit on the filter was examined under a microscope at 160x magnification with an ultraviolet light source. In general, all the particles collected by the droplet were counted.

Each particle in an aggregate was counted as an individual. This was permissible because the thermal precipitator samples showed the aerosol was comprised of discrete particles. Aggregates were obtained only with the evaporating and steady-state tests. Representive fields of the thermal precipitator deposits were counted to obtain a value for the aerosol concentration. The reverse sides of cover slips used in the thermal precipitator were painted black to aid in counting the samples.

For the tests on aerosols generated from dry powder, zinc sulfide powder NJZ-2210 was used. The particle size distribution of the specific lot, as determined by Rosinski (dilution method),[12] is shown in Table 3. The particle size distribution of the generated aerosol, as determined by the ARF particle counter, is also shown.

Table 3

PARTICLE SIZE DISTRIBUTION OF ZINC SULFIDE POWDER NJZ-2210

Dilution Method		Particle Counter	
Particle Diameter, microns	Distribution, %	Particle Diameter, microns	Distribution, %
<1	28.0	1-1.4	24.3
1-2	40.0	1.4-2	31.8
2-3	18.0	2-2.8	28.1
3-4	7.5	2.8-4	11.3
4-5	3.5	4-5.6	3.6
5-6	1.4	5.6-8	0.9
6-7	0.7		
>7	0.9		

[12] Rosinski, J. Anal. Chem. 28, 486, 1956.

In addition to aerosols from dry powders, aerosols of gold and polystyrene latex were generated from water suspensions by the Lauterbach generator.[13] The polystyrene was furnished through the courtesy of the Dow Chemical Company.

[13] Lauterbach, K. et al. Arch. Ind. Hyg. 9, 69, 1954.

V. RESULTS AND DISCUSSION

A. Air Sampling

Data obtained on the radioactivity of dry atmospheric particles are presented in Table 4 and 5. Table 4 shows the total beta activity and the portion due to strontium-90 obtained with the Andersen samplers from July 15 to October 20, 1958. Table 5 gives the same data with the cyclone separator for four sampling periods during November 24 to December 29, 1958. Precipitation during the sampling periods is shown in Figure 9. Sampling was suspended during precipitation. Results are not yet available on the particles separated by the Andersen samplers from November 18, 1958, to February 13, 1959, and by the cyclone separator from January 22 to January 30, 1959.

Table 4

RADIOACTIVITY ASSOCIATED WITH DRY ATMOSPHERIC PARTICLES CLASSIFIED BY THE ANDERSEN SAMPLERS FROM JULY 15 TO OCTOBER 20, 1958

Sampler No.	Particle Size Range	Radioactivity, $\mu\mu c/100{,}000\ ft^3$ of air	
		Total Beta	Strontium-90
1	5.5+ microns	628	2.38
	1-5.5 microns	2475	34.9
	<1 micron	120	0.87
	Millipore filter	2584	71.0
		5807	109.15
2	5.5+ microns	180	4.35
	1-5.5 microns	3306	46.8
	<1 micron	622	36.0
	Millipore filter	2908	85.5
		7016	172.65
1 and 2	Average	6412	140.9

Total beta activity for 16 cg of corn syrup is 7.7 dpm.

Table 5

RADIOACTIVITY ASSOCIATED WITH DRY ATMOSPHERIC PARTICLES CLASSIFIED BY THE CYCLONE SEPARATOR FROM NOVEMBER 24 TO DECEMBER 29, 1958

Date	Particle Size Range	Radioactivity, $\mu\mu c/100{,}000\ ft^3$ of air	
		Total Beta	Strontium-90
11/24-12/3/58	Retained by separator	8,205	32.6
	Passed by separator	18,746	128.0
		26,951	160.6
12/3-12/12/58	Retained by separator	6,731	28.2
	Passed by separator	14,128	80.4
		20,859	108.6
12/15-12/22/58	Retained by separator	6,740	18.9
	Passed by separator	19,687	119.0
		26,427	137.9
12/22-12/29/58	Retained by separator	7,217	35.3
	Passed by separator	33,304	161.0
		40,521	196.3
11/24-12/29/58	Average:		
	Retained by separator	7,223	28.8
	Passed by separator	21,466	122.1
		28,690	150.9

Results obtained by the two Andersen samplers were expected to be similar because their inlets were located about 4 in. apart. With the exception of the strontium-90 activity in the less-than-1-micron fraction, comparable results were obtained. From July 15 to October 20, 1958, the total beta activity was 6412 $\mu\mu c/100{,}000\ ft^3$ of air. The strontium-90 activity was 140.9 $\mu\mu c/100{,}000\ ft^3$, or 2.2% of the total activity. The activity is associated with two particle size fractions: (1) particles 1-5.5 microns in diameter contributed 45% of the total beta activity and 29% of strontium-90 activity and (2) the minute particles which passed through the samplers and were collected on the millipore filter contributed 43% of the total beta activity and 56% of strontium-90 activity. Therefore, according to these data, submicron particles contain a greater portion of strontium-90 activity.

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The choice of design characteristics for the cyclone separator proved unfortunate in that the particle size effectively retained by the separator was within the range of 1-5.5 microns. The average of the four samples analyzed by the cyclone separator was 28,690 $\mu\mu$c/100,000 ft^3 for total beta activity and 150.9 $\mu\mu$c/100,000 ft^3 for strontium-90 activity. The portion of the total activity retained by the separator was 25%, and the portion of strontium-90 retained was 19%. The filter collected 75% of the total activity and 81% of the strontium-90 activity. Again, the largest portion of the strontium-90 activity was associated with the smaller particles. The strontium-90 activity for the samples obtained by the cyclone separator system and the Andersen sampler system were comparable, but the total beta activity obtained with the cyclone separator was about 4.5 times that obtained with the Andersen samplers. Whether this variation was due to the different sampling periods or differences in the mode of collection cannot be determined from the limited number of tests.

The qualitative relationship between radioactivity and particle size, as indicated by the air sampling results, is depicted graphically in Fig. 10 and 11. It might be suggested at present that the radioactivity associated with particles below 0.03 micron is due to primary radioactive particles or small agglomerates thereof. These particles are transported to earth by airmass movements. The radioactivity associated with the 1- to 5-micron particles may represent particles scavenged by liquid water droplets or ice crystals in the free atmosphere and agglomerated after evaporation of the droplets, or may be associated with aggregates formed by coagulation of particles originating from the earth (air pollution). From the limited number of samples, it is difficult to draw any conclusions. The former mechanism of agglomeration seems more probable, however.

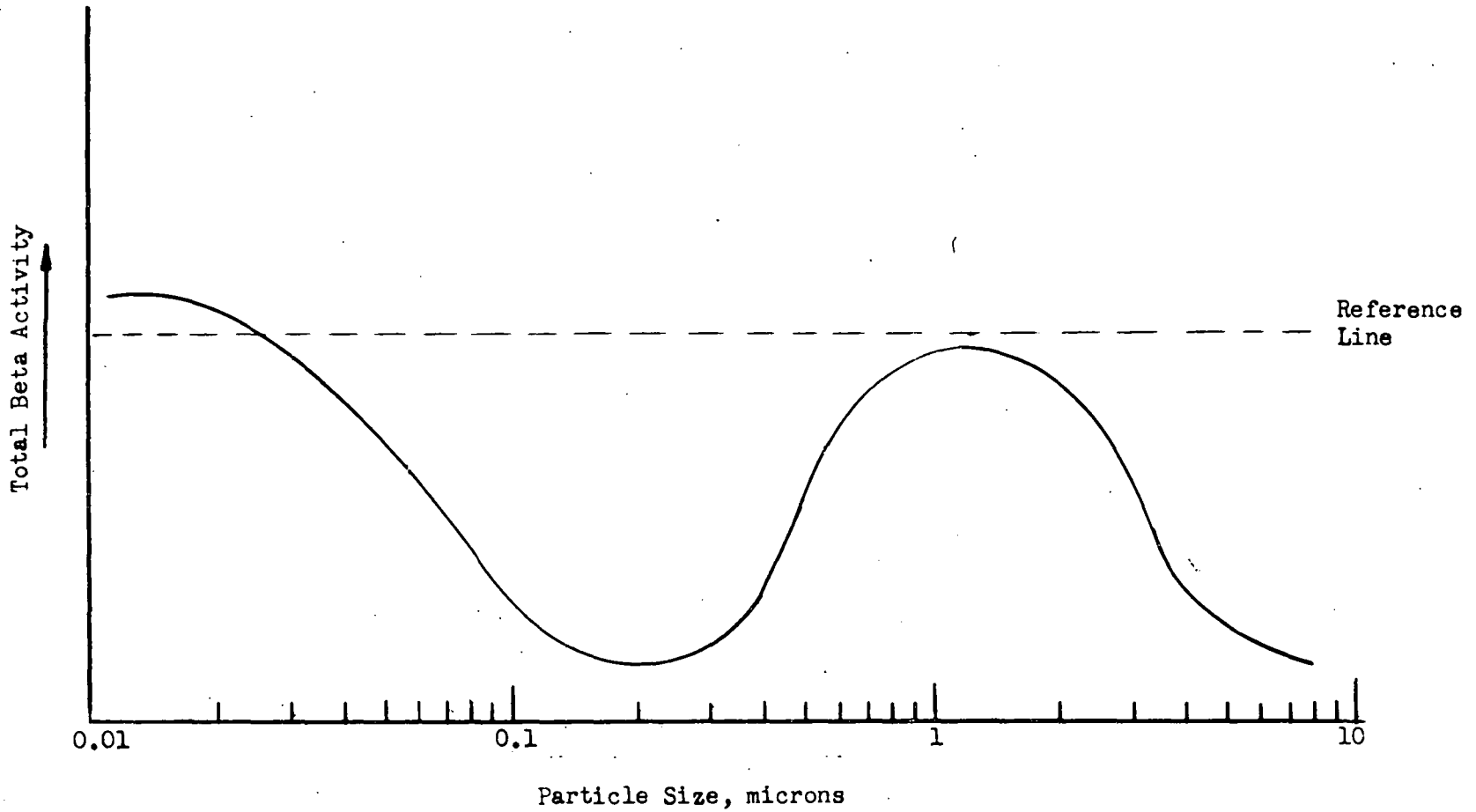

Fig. 10. RELATION BETWEEN PARTICLE SIZE AND TOTAL BETA ACTIVITY DURING DRY FALLOUT

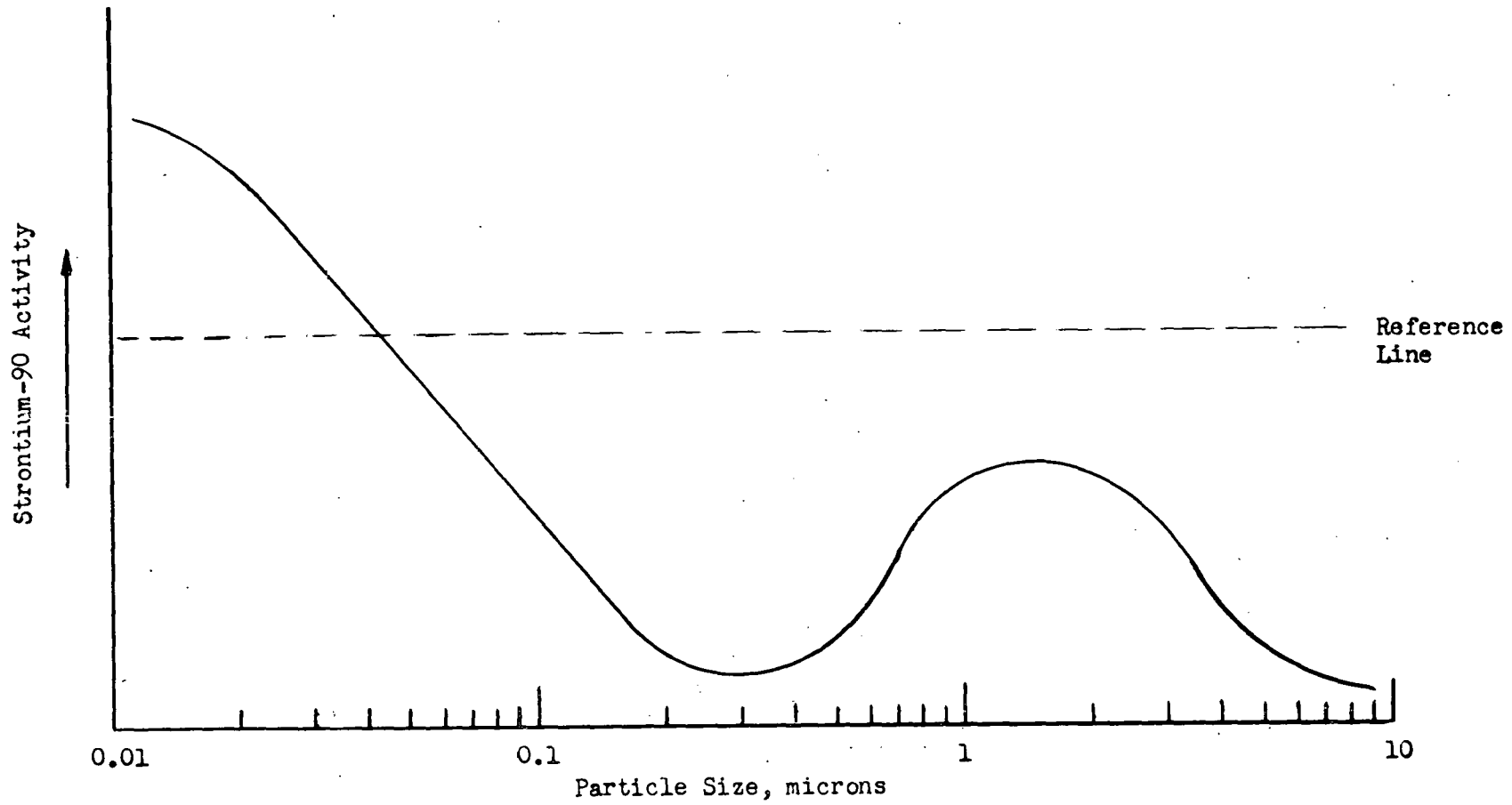

Fig. 11. RELATION BETWEEN PARTICLE SIZE AND STRONTIUM-90 ACTIVITY DURING DRY FALLOUT

Bradshaw[14] reported on the gross beta activity of atmospheric particles at Cincinnati, Ohio. His data were obtained during the winter and spring of 1955 with an Aerotec No. 2 cyclone and a glass fiber filter. The characteristics of the Aerotec cyclone are similar to those of the cyclone separator used in this study. He reported that the maximum, minimum, and average of daily samples were 30, 0.35, and 5 $\mu\mu c/m^3$. This average is equal to 14,100 $\mu\mu c/100{,}000\ ft^3$. Bradshaw's cyclone retained 34% of the average total activity. His data were obtained after nuclear detonations in Nevada. Although the article did not state so, it is assumed that sampling took place during precipitation.

B. Scavenging Mechanism

Results on the study of scavenging with a fluorescent zinc sulfide aerosol are shown in Table 6. Duplicate tests were made with evaporating, steady-state, and condensing water droplets. The value for the efficiency, e, of capture calculated from the following equation is given in Table 6.

$$e = \frac{N}{V\,C\,t}\,\frac{S_T}{S_D}\,, \qquad (25)$$

where

N is the number of particles captured

V is the airflow through the chamber, cm^3/min

C is the particle concentration, $particles/cm^3$

t is time, min

S_D and S_T are the cross sectional areas of the droplet and the test chamber, respectively.

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[14] Bradshaw, R. Public Health Rept. 73, 431, 1958.

Thus the capture efficiency is the ratio of the number of particles which are captured to the number of particles which pass through the cross sectional area of the droplet projected normal to the direction of flow during the test period.

Table 6

SCAVENGING OF ZINC SULFIDE PARTICLES BY WATER DROPLETS

	Condition of Water Droplet					
	Evaporating		Steady-State		Condensing	
	Test 1	Test 2	Test 3	Test 4	Test 5	Test 6
Aerosol flow rate, cm^3/min	550	550	550	550	550	550
Aerosol velocity, cm/min	7.6	7.6	7.6	7.6	7.6	7.6
Relative humidity, %	50-70	50-70	80-90	80-90	80-90	80-90
Particle concentration, particles/cm^3	714	1072	1380	3138	3189	3621
Test time, min	25	20	23	20	13	14
Particles captured, number	1836	2530	2404	1588	10641	5095
Expected collisions by Brownian motion	0.014	0.014	0.025	0.04	0.027	0.032
Efficiency, %	39.9	63.2	34.8	13.7	94.5	39.2

The growth histories of the droplets during the test period are recorded in Fig. 12, 13, and 14.

The true efficiency of capture of aerosol particles by a collector can be determined only by evaluating all the forces which affect the movement of the particle. While a general diffusion equation is not available, it is possible to evaluate the individual forces by means of the parameters discussed in Sections II and III. Although the forces of the various mechanisms are additive, the resulting individual efficiencies are not directly additive. Each mechanism contributes to the total efficiency, and no combination of

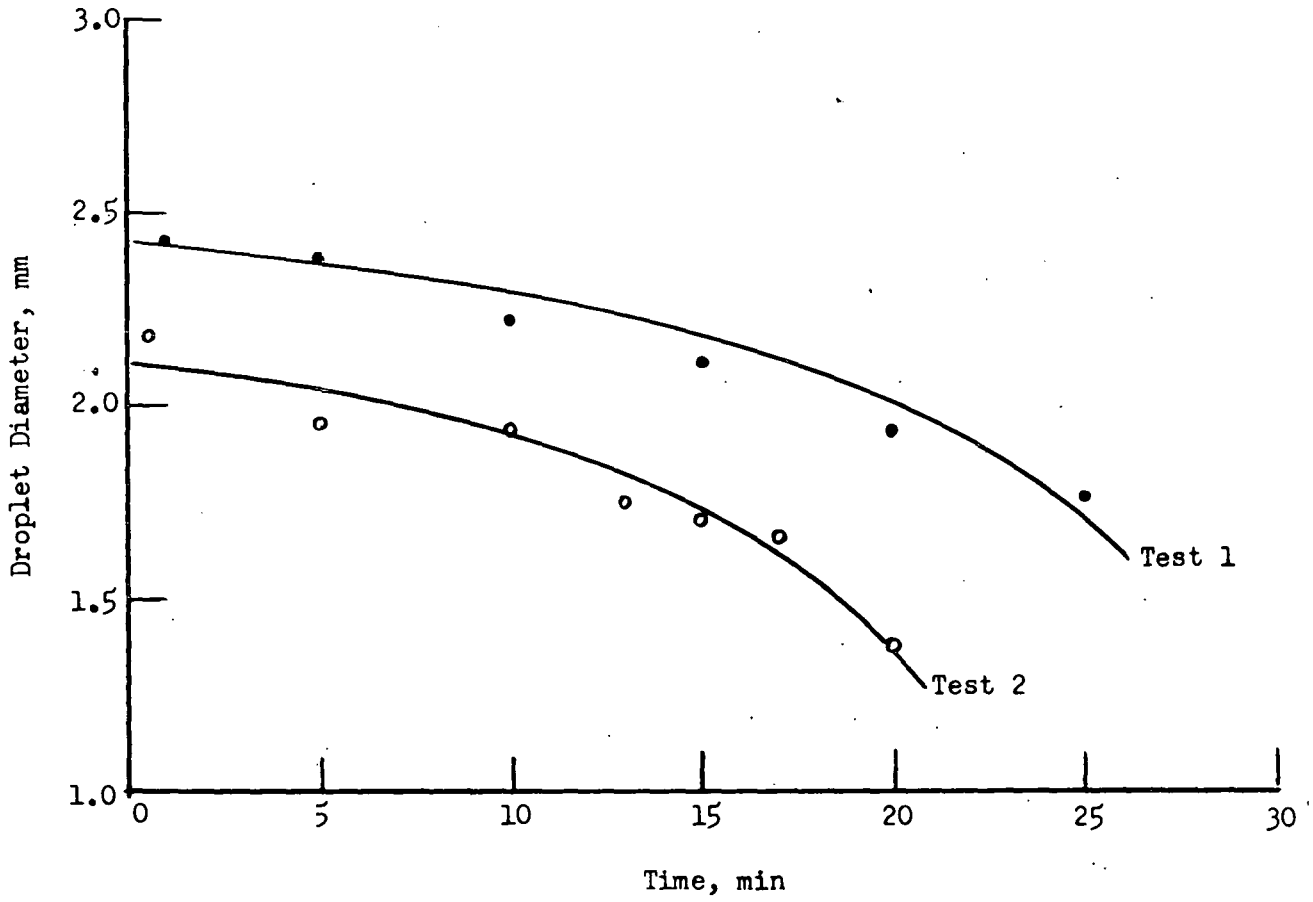

Fig. 12. CHANGE IN DROPLET SIZE DURING EVAPORATION TESTS

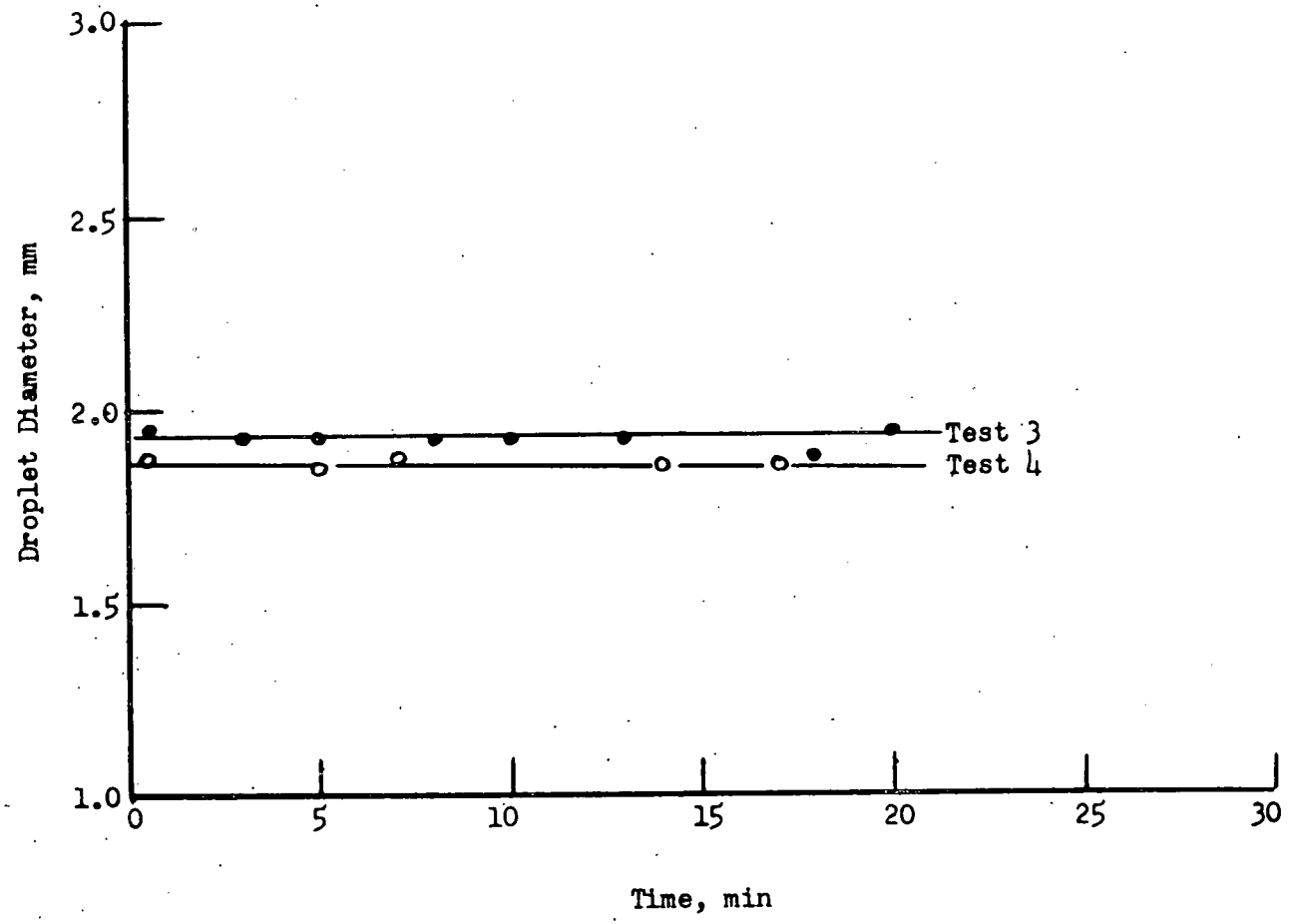

Fig. 13. CHANGE IN DROPLET SIZE DURING STEADY-STATE TESTS

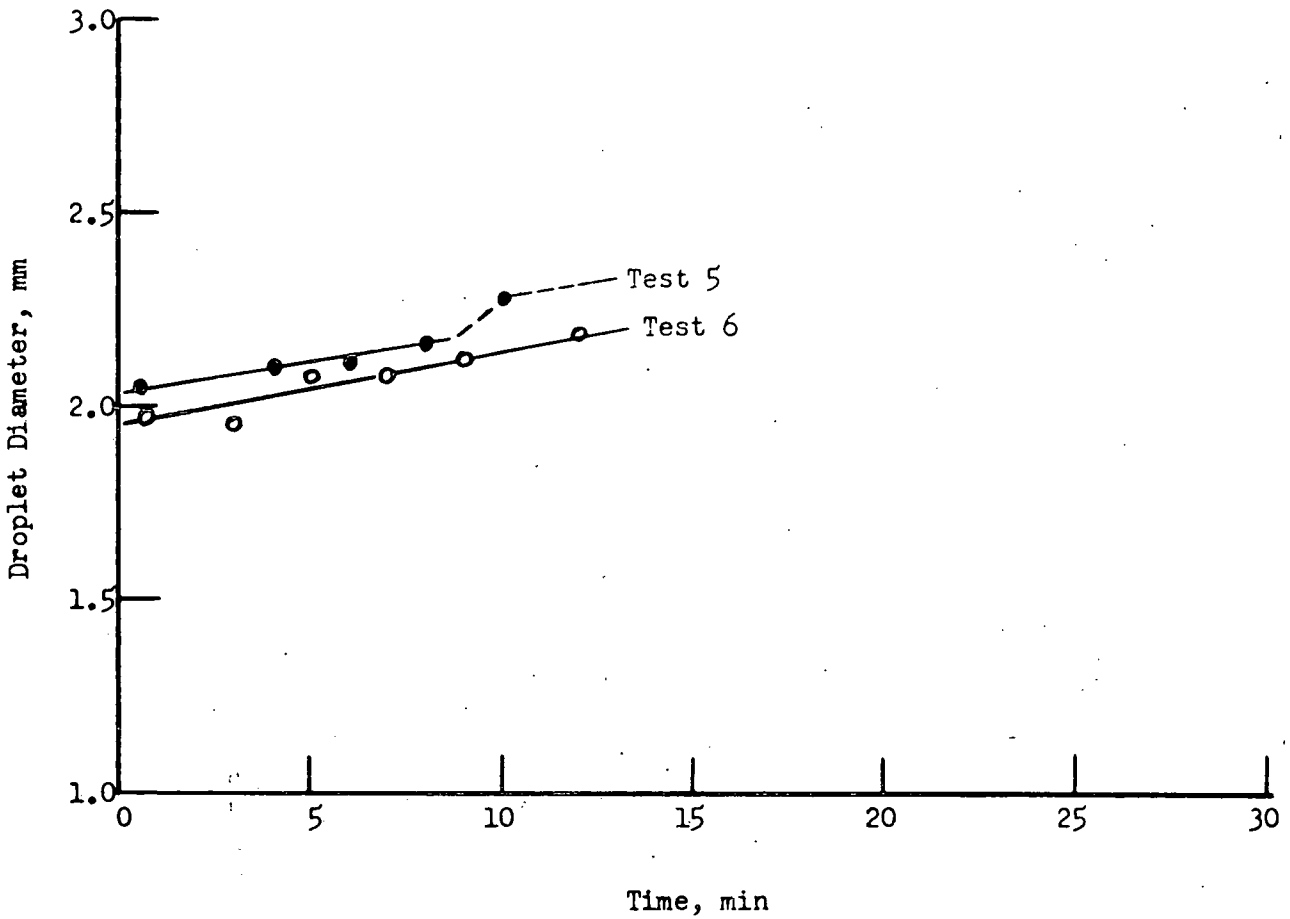

Fig. 14. CHANGE IN DROPLET SIZE DURING CONDENSATION TESTS

favorable mechanisms causes an efficiency lower than that of any one of the favorable mechanisms. Thus the efficiency, e, can be defined as follows:

$$e = \frac{B}{N_{int} + N_{in} + N_s + N_B + N_e + N_{VP} + N_0}, \qquad (26)$$

where

N is the number of particles captured

N_{int} is the number of particles captured by interception

N_{in} is the number of particles captured by inertia

N_s is the number of particles captured by settling

N_B is the number of particles captured by Brownian motion

N_e is the number of particles captured by electrostatic attraction

N_{VP} is the number of particles captured by vapor pressure gradient

N_0 is the number of particles captured by other means.

A particle size of 1.9 microns (the mean volume diameter of aerosol particles to which the droplet was exposed) and a droplet diameter of 0.2 cm were used for calculations. The interception parameter in Eq. 2 is 9.5×10^{-4}, and so the efficiency of collection by interception is 0.0027. The inertia parameter in Eq. 1 equals 2.7×10^{-5}. Comparison of this value with the value 4×10^{-2}, which is necessary for impaction by inertia to become effective, indicated that no particles were collected by this mechanism.

A value of 1.2×10^{-2} for the settling parameter in Eq. 3 indicates that settling is important in particle capture. The efficiency of settling, e_s, obtained from the ratio of the terminal settling velocity of a 1.9-micron particle to the air velocity in the test chamber, is 0.36.

Equation 8 was used to determine the expected collision between aerosol particles and water droplets due to Brownian motion. According to

the equation, less than 1 collision occurs at the aerosol concentrations used. Thus, Brownian motion can be eliminated from consideration.

From Eq. 15, the number of collisions due to diffusion of water vapor was found to be about 1.25×10^{-13} collisions per particle per droplet per sec. Thus, no particles are captured by water vapor diffusion.

Capture due to electrostatic effects were estimated from the parameters given by Kraemer and Johnstone.[7] By means of an electrometer, the mean net charge on the aerosol particles was found to be 2.7×10^{-17} coulombs/particle. Parameters K_E and K_I are not involved here because the droplet was not charged. K_S and K_M are negligible. The value for K_G proves to be the predominant mechanism of capture in these experiments.

Table 7 shows data on the number of particles which theoretically will collide with a droplet by the various mechanisms. Data for each of the six tests are reported.

Table 7

MAXIMUM NUMBER OF PARTICLES WHICH CAN BE CAPTURED BY VARIOUS MECHANISMS

	Condition of Water Droplet					
	Evaporating		Steady-State		Condensing	
Capture Mechanism	Test 1	Test 2	Test 3	Test 4	Test 5	Test 6
Interception	13	11	19	35	30	35
Inertia	0	0	0	0	0	0
Settling	1,680	1,460	2,470	4,590	4,040	5,100
Brownian motion	0	0	0	0	0	0
Vapor pressure gradient	0	0	0	0	0	0
Electrostatic attraction	2,800	3,660	7,700	32,000	28,600	35,400
Total possible	4,493	5,131	10,189	36,625	32,670	40,535
Captured, number	1,836	2,530	2,404	1,588	10,641	5,095
Captured, %	41	49	24	4.3	32	13

Not all the particles hitting the droplet should be expected to be captured. Some portion of the particles will collide with sufficient velocity to rebound. Because the particles are hydrophobic, they will remain on the surface of the droplet, where they can be knocked off by collisions of other particles. Both these factors, rebound and hydrophobicity, tend to reduce capture efficiency in highly charged, concentrated aerosols.

During investigation of the charge on the aerosol, it was noted that the dry aerosol had a net deficiency of 168 electrons/particle. Introduction of humid air (tests 3, 4, 5, and 6) gave the aerosol an opposite charge of about equal magnitude, which was probably due to production of charged water droplets by the humidifier. The effect of the change on capture is difficult to ascertain at this stage of study.

In addition to the fluorescent zinc sulfide aerosols, polystyrene latex particles produced by the Dow Chemical Company were tested. The latex particles are available in several sizes from 0.1 to 1 micron. The production process used gives good uniformity within each size range.

Determination of the number of latex particles captured is extremely difficult. Present analytical instrumentation has a detection range of 10^{-9} g of material. A 0.2-micron latex particle weighs 3×10^{-15} g. Thus, 10^{6} particles must be captured for detection. Zebel's[6] equations predict about 1000 collisions/hr. Optical counting methods were used, but it was difficult to distinguish between the latex particles, extraneous matter, and slide imperfections. Also, it was difficult to obtain a uniform deposit of the particles on a slide.

Gold sols with particles in the angstrom range were also studied. The sols were prepared free of solutes, with the exception of minute quantities

of stabilizer. Upon atomization, however, large globules of particles and stabilizer were obtained instead of individual particles. It was therefore not possible to count the number of particles captured. Passing the aerosol over a drying agent and then subjecting it to temperatures of 425°C for several seconds appears to eliminate the stabilizer from the particles.

VI. FUTURE WORK

The present contract has been renewed for an additional year under the title "Some Studies Related to Radioactive Fallout." The scope and objectives are extensions of those stated earlier in this report.

Air sampling during dry periods will continue with both the Andersen samplers and the cyclone separator. In view of the results obtained, the stages of the Andersen sampler will be analyzed separately and the new cyclone separator will be designed to obtain a more desirable point of separation.

Laboratory studies on collection mechanisms will be continued. Means of neutralizing the charge on the aerosol and eliminating random air currents will be studied. Arrangements are being made with the Dow Chemical Company for the production of 0.1- and 1-micron polystyrene latex particles from carbon-14. Gold sols with particle sizes of 0.04 micron will also be tested. After collection, the gold can be neutron irradiated and the quantity determined with suitable radiation measuring devices. Tritium can be used to evaluate the change in droplet mass during the tests for sizes which cannot be determined accurately with optical systems.