Rain Scavenging of Zinc Sulfide Particles

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Abstract

Several scavenging experiments were conducted using artificial rain in the outdoors with zinc sulfide tracer particles. The measured collection efficiencies for raindrops are compared to the theoretical predictions of Langmuir, and to the measurements with freely falling drops made by Kinzer and Cobb, Walton and Woolcock, and McCully et al.

The four main characteristics of the efficiencies found are 1) higher efficiencies for drops of about 0.1 mm diameter, 2) efficiencies greater than 1.0 with all drops for particles larger than 13 microns, 3) a rapid increase in efficiency with particle size in the general region of ten microns diameter, and 4) an apparent minimum for all particle sizes with drop sizes of about 0.8 mm diameter.

The explanation offered utilizes the classical inertial impaction of particles on the leading side of the drops with additional wake effects. Particles that narrowly miss the drops are drawn into the wake, fall therein with respect to the drop, and impact on the upper side of the drop. This favors the collection of the larger particles. The vortices in the wakes of the larger drops are more quickly shed and the chance of wake collection is reduced.

1. Introduction

Increasing quantity and toxicity of the pollutants in our atmosphere have increased the requirement for accurate predictions of the washout of pollutants by precipitation. The washout rate of particulate material below cloud level is the integrated product of the flux densities of the raindrops, their cross sections, and their scavenging efficiencies. The flux densities of the drops are obtained from raindrop spectra. The scavenging efficiencies, however, must be measured or calculated separately.

The scavenging or collection efficiency of a raindrop is that proportion of the particles in its path which is collected. It is better defined as the product of the target efficiency and the retention efficiency, the retention efficiency being the proportion of those making contact which is retained.

Target efficiencies are predicted theoretically by substituting into Newton's second law the forces on a particle contained in air flowing past the raindrop. Using numerical methods, investigators have obtained target efficiencies in viscous and potential flow (e.g., Sell, 1931; Langmuir and Blodgett, 1945; Das, 1950; Herne, 1960). The efficiencies are usually presented as functions of the impaction parameter,

\[ K = \frac{2a^2 \rho U}{9 \mu} S, \quad (1) \]

where \( a \) and \( \rho \) are the particle radius and density, \( \mu \) is the viscosity of the air, and \( U \) is the terminal velocity of a raindrop of radius \( S \).

It is easy to be misled by the accurate computations carried out with modern computers. The fact is, however, that predicted target efficiencies can be no better than the flow fields from which they are derived.

The assumption is often made that the target efficiency for raindrops lies between the target efficiencies calculated for viscous and potential flows, as given by Langmuir's (1948) interpolation formula. However, flow patterns at intermediate Reynolds numbers contain wakes with vortex flow (Pearcey and Hill, 1957; Stewartson, 1956; Möller, 1938), and the effect of the wake on scavenging is open to speculation. And, where larger drops are deformed to some extent, one cannot assume that ideal flow fields about a sphere will even apply on the upstream side. It would seem, therefore, that one has considerable freedom in choosing from among available predictions.

This is an especially unhappy state of affairs since the chosen target efficiencies must then be multiplied by an unknown retention efficiency. The future of a particle as it intersects a drop surface could be rather uncertain in the light of electrical effects, drag by the airstream, and wettability characteristics.

1 This paper has been abstracted from a dissertation submitted to the Department of Atmospheric Sciences, University of Washington. The complete dissertation has been published as BW-79382, Rain Scavenging of Particulates, at Hanford Laboratories, Richland, Washington. It is available from the Clearinghouse for Federal Scientific and Technical Information, Port Royal Road, Springfield, Virginia 22151, price, $3.00. This work was performed under Contract No. AT(45-1)-1380 between the Atomic Energy Commission and General Electric Company.

2 Recently changed to Battelle-Northwest.
Some theoretical work showing a reduction in collection of nonwettable particles has been presented by McCully et al. (1956), Pemberton (1960), and enlarged by McDonald (1963), but this work has not been properly supported with observations.

Kinzer and Cobb (1958), with laboratory experiments, found the efficiency for drops 0.3 to 1.0 mm in diameter to be greater than that predicted using potential flow, and felt that there must be some mechanism other than electrical effects at work. The peak scavenging efficiency was observed with 0.4-mm diameter drops.

Walton and Woolcock (1960), with laboratory tests with freely falling drops, found that 0.5-mm drops seemed to scavenge more efficiently than 2.6-mm drops, but felt that this could be attributed to the fact that the large drops were falling at much less than terminal velocity. They next suspended their raindrops from vertical capillaries and moved the particulate cloud upwards past the drops. The 0.5-mm drop was no longer the more efficient. It is probable that normal wakes will not occur in this situation.

It is indeed difficult to approximate natural states in a laboratory. Particularly objectionable is the collection of bulk rain samples, even when drop-size distributions are available. One cannot choose a representative drop size for the rain without introducing assumptions concerning the efficiency versus drop size curve. The same type of assumptions are made when a roughly monodisperse tracer is used but not sized after scavenging.

There are, in fact, so many uncertainties that successful prediction of particulate washout in the atmosphere would be fortuitous. There is immediate need for detailed measurements of the collection efficiencies with respect to both the particle size and drop size for at least one tracer. It was decided that the diffusion sampling course and tracer technique used by Atmospheric Physics of Hanford Laboratories could also be utilized in scavenging research. This paper describes the special rain samplers utilized, the experiments conducted, and the collection efficiencies obtained in the natural atmosphere for microscopic zinc sulfide particles.

2. Raindrop samplers developed for this research

Of the many rain sampling methods, that of Gunn (1949) seemed best able to sample individual raindrops. Gunn's instrument consisted of a moving sheet of blueprint paper. The size of a spot left on the paper was calibrated against the size of the drop producing it. Similar samplers utilizing filter paper dusted with water-soluble dye have been used.

A difficulty with using blueprint paper is its light sensitivity; difficulties with dyed filter paper are those of preparation and fragility. There was desired a low-priced paper that had strength and flexibility, and that provided a permanent or semipermanent record of waterdrops.

A number of papers now used instead of blueprint paper fulfill the requirements. One which has minimum capillary action was chosen for use (Ozalid 10S5Z, manufactured by Ozalid, a division of General Aniline and Film Corporation).

This paper consists of a sulfite paper base coated with a thin layer of polyvinyl acetate to present a very smooth surface. This layer is treated with diazonium salts, stabilizers, couplers, etc., which are partially water soluble.

When fully developed using ammonia fumes, the paper becomes black with white spots where waterdrops have dried. The best contrast for sizing work is obtained, however, by using a partial development while the spots are still wet. Spots 0.2 mm in diameter, corresponding to 0.13-mm drops, are easily identified with the naked eye (see Fig. 1).

Under a minimum tension, only drops larger than 1.2 mm show splash lines radiating outward from the spot. Small droplets from this type of splash can be later associated with the parent drop, as a rule.

The calibration procedure was patterned after that of Gunn and Kinzer (1949), with some modifications (Engelmann, 1962). Ten to 30 drops and about as many spots were sized to provide each of the 23 points from which the calibration curve was obtained. The least-squares fit, shown in Fig. 2, was

\[ D = 0.435V^{0.74} \]  

(2)
Consideration was given to the errors arising from variations in the paper, and in the development, bleaching, and drying times. The standard deviation of the spot diameter was found to be about 10 per cent for 2.15-mm drops, and about 20 per cent for 0.55-mm drops. Inasmuch as the variance of the spot size also contained the variance in the size of the drops used for calibration, it appears that the total error in sizing a raindrop is less than 10 per cent.

The cover of the sampler shown in Fig. 3 slides in two directions to expose successively two 11×17-inch sheets of the paper. It yields a large amount of data and is quite useful when a time average is not required. A continuous rain sampler is shown in operating condition in Fig. 4. The material covering the samplers is towelling. Towelling, even when wet, was found to virtually eliminate the splashing of drops. The sampler consists of a box housing a chart drive, a hot air system for drying the droplets that fall through the entrance hole, and a developing section.

It is hard to conceive of a sampler that doesn’t split raindrops at the edge of its entrance. The edge of the sampler entrance is coated first with a common household cement and then, before the cement hardens, it and the towelling near the entrance are dusted with a highly soluble dye. In addition, blotting paper impregnated with the dye is fastened to the underside of the cover around the sampler entrance.

Most of the drops that have broken on the sampler edge will leave characteristic spots; often the spots are elliptical with the major axis not parallel to the wind.

3. Field experimental design and conduct

The experiment designed is essentially the following. A particulate tracer is released from a ground source 200 m upwind from an 88-ft pole (Fig. 5). On the pole are exposed cellulose acetate or cellulose nitrate filters used to determine the concentration of each of the sizes of particulates in the plume. Artificial rain falls through the plume just downwind from the sampling pole. The rain is size-separated by the wind, and the consequences of this will be seen later.

Sufficient numbers of drops are individually collected over the time that the plume concentration is sampled to enable one to compare a mean drop content to a mean concentration in the plume. Further, sufficient numbers of drops are collected at the same time that one may compare the collections of drops of different sizes.
The tracer used is the zinc sulfide crystal manufactured by U. S. Radium Corporation as No. 2210. It fluoresces with green light when excited by ultraviolet light. It has approximately a log-normal size distribution with a geometric mean diameter of about two microns. The 2210 was enriched in large particles for some scavenging studies by blending with No. 2330. The latter is also a zinc sulfide crystal.

The tracer is suspended in agitated water to which a small quantity of sodium lauryl sulfate is added as a wetting agent. The suspension is atomized into a hot (550°C) atmosphere using an insecticidal fog generator. These water droplets are initially less than 200 microns in diameter, and calculations show (Engelmann, 1963) that in relative humidities less than 80 per cent they will evaporate before reaching the scavenging site. The final proof that the plume droplets have evaporated rests with the particles found in the collecting rain. If these are larger than the bulk of those released, the evaporation must be nearly complete.

Available for the more recent experiments was an experimental “live-sampler” capable of detecting minute quantities of the tracer in the atmosphere. This sampler was developed by Mr. M. O. Rankin of Hanford Laboratories. In operation, the air containing the zinc sulfide is drawn past an ultraviolet light and then into a dark chamber where a phototube measures the phosphorescence. The output is satisfactory for determining constancy of the plume during a scavenging experiment.

All tests used essentially the same techniques, although as the research progressed the equipment and the experiments became more refined and complete. The final procedure is described in the following paragraphs.

While the particulate tracer is being mixed in water, the equipment at the sampling pole is readied. The pumps providing vacuum to the filters on the pole are started. The “live-sampler” and the continuous rain sampler are actuated and given a time mark simultaneously.

When the wind direction appears satisfactory the generation is begun, the artificial rain turned on, and totalizing anemometers are read. When the plume has arrived and the live-sampler shows it to be constant, the slide samplers are exposed in order, starting with the one nearest the source. Personnel take care that rain does not splash from them or equipment onto the samples. A set of five samples is obtained in about one minute. The sample boxes are then turned end-for-end so that the second sheet is ready for exposure, and the procedure is repeated if the meteorological conditions persist.

After a period of about five minutes, the vacuum is removed from the filters as the record is marked, and the generation is ended. The anemometers are read, the artificial rain is turned off, and the continuous sampler is allowed to run its sample through the developing stage. After the test, the samples are laid out on a table covered with fresh wrapping paper, cut up, labelled, and mounted between new 3 × 4-inch glass lantern slides.

4. Microscopic system and procedure

It is necessary to size the drop images and to count and to size the particles within each image. In general, the lowest possible magnification is used. This is because the task of counting and, especially, of sizing particles is most fatiguing and time consuming, and becomes increasingly so as the magnification increases. Drop images are sized at 35× or 100×. Particles are located with incident ultraviolet, and sized at 250× with added transmitted white light from below. Sizing is done by comparison of the areas of particles to the areas of circles in a graticule.

In the case of a filter, there are so many particles that it is necessary to select a sample. To avoid biasing the result, a narrow strip passing through the center of the filter is counted or sized, with acceptance of all particles in the strip and all touching one edge, but with rejection of all particles touching the other edge of the strip. It was determined that the size distribution of particles was essentially constant across the filter.

5. Enumeration of tests conducted

Test R3. Test R3, using the continuous rain sampler, was conducted on 25 August 1961. The sampler was mounted on a truck which was driven forward and backward during the test in order to sample a range of drop sizes. The plume was contained within the height of the filters (as in all tests reported here), and the particulate content of the drops was good.

Contrary to expectations, the plume concentration varied, and, therefore, some one size of drop should have been liberally sampled throughout the test. This was not done, of course, with the truck driving forward and backward. However, the test was salvaged by combining the collection of drops in the diameter range of 0.6 to 1.2 mm.

These drops have nearly the same impactation parameters, as seen in Table 1. Values in this table are based on Gunn and Kinzer’s (1949) terminal velocity measurements.

Tests R7, R8, R9 and R10. Several tests were conducted in a stable atmosphere between 0500 and 0730 PST on 18 September 1962. These tests, R7, R8, R9 and R10, were designed to ascertain whether there was higher scavenging efficiency for the 0.4-mm drop.

Four tests were conducted of two “takes” each. A “take” consisted of exposing a sheet of sampling paper on each of the three samplers. Since a few minutes were consumed before the second set of sheets was exposed, it is not safe to assume that the two “takes” in each test were obtained with the same plume concentration.
Table 1. Impaction parameter, \( K \), for selected raindrop and zinc sulfide diameters.

<table>
<thead>
<tr>
<th>Drop diameter (mm)</th>
<th>2</th>
<th>6</th>
<th>8</th>
<th>10</th>
<th>14</th>
<th>18</th>
</tr>
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<tr>
<td>0.1</td>
<td>0.27</td>
<td>2.46</td>
<td>4.37</td>
<td>6.80</td>
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<td>22.1</td>
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<td>0.2</td>
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<td>5.83</td>
<td>9.07</td>
<td>17.9</td>
<td>29.5</td>
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<tr>
<td>0.3</td>
<td>0.40</td>
<td>3.55</td>
<td>6.32</td>
<td>9.83</td>
<td>19.3</td>
<td>32.0</td>
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<tr>
<td>0.4</td>
<td>0.41</td>
<td>3.68</td>
<td>6.56</td>
<td>10.21</td>
<td>20.1</td>
<td>33.2</td>
</tr>
<tr>
<td>0.5</td>
<td>0.42</td>
<td>3.75</td>
<td>6.67</td>
<td>10.38</td>
<td>20.4</td>
<td>33.8</td>
</tr>
<tr>
<td>0.6</td>
<td>0.42</td>
<td>3.75</td>
<td>6.67</td>
<td>10.37</td>
<td>20.4</td>
<td>33.8</td>
</tr>
<tr>
<td>0.7</td>
<td>0.42</td>
<td>3.73</td>
<td>6.64</td>
<td>10.33</td>
<td>20.4</td>
<td>33.6</td>
</tr>
<tr>
<td>0.8</td>
<td>0.41</td>
<td>3.72</td>
<td>6.62</td>
<td>10.30</td>
<td>20.3</td>
<td>33.5</td>
</tr>
<tr>
<td>0.9</td>
<td>0.41</td>
<td>3.71</td>
<td>6.61</td>
<td>10.28</td>
<td>20.2</td>
<td>33.4</td>
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<tr>
<td>1.0</td>
<td>0.41</td>
<td>3.67</td>
<td>6.53</td>
<td>10.16</td>
<td>20.0</td>
<td>33.0</td>
</tr>
<tr>
<td>1.2</td>
<td>0.39</td>
<td>3.52</td>
<td>6.26</td>
<td>9.74</td>
<td>19.2</td>
<td>31.7</td>
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<tr>
<td>1.4</td>
<td>0.37</td>
<td>3.36</td>
<td>5.98</td>
<td>9.31</td>
<td>18.3</td>
<td>30.3</td>
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<td>1.6</td>
<td>0.36</td>
<td>3.21</td>
<td>5.72</td>
<td>8.90</td>
<td>17.5</td>
<td>29.0</td>
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<tr>
<td>1.8</td>
<td>0.34</td>
<td>3.08</td>
<td>5.48</td>
<td>8.53</td>
<td>16.8</td>
<td>27.7</td>
</tr>
<tr>
<td>2.0</td>
<td>0.33</td>
<td>2.95</td>
<td>5.26</td>
<td>8.18</td>
<td>16.1</td>
<td>26.6</td>
</tr>
</tbody>
</table>

Test R13. Test R13 was conducted from 1901 to 1909 PST on 3 January 1963. Although the relative humidity was a marginal 70 per cent, the particle sizes found in the raindrops were somewhat larger than in a later test, R15, which was conducted in low humidity. The 2210–2330 blend of tracers was first used in this test.

Although there was a high concentration of pigment on the filters and in the raindrops, indicating that the plume was “on course,” there were fewer raindrops collected by the continuous sampler than desired. However, sufficient particles were collected to permit comparison of the particle sizes in the drops to those in the plume.

Test R15. Test R15 was made from 2109 to 2114 PST on 12 March 1963, using the 2210–2330 tracer blend. The test was successful for all purposes except that of determining efficiency versus drop size, the slide samplers having been exposed during a period of low tracer concentration. The drops sampled by the continuous sampler were predominantly in the size range 1.2- to 2.0-mm diameter, in which the impaction parameter is reasonably constant.

6. Location and characteristics of particles in raindrop images

It is a curious circumstance that the particles collected by a raindrop bear some semblance of organization, even after the evaporation of the raindrop and development of its image. A substantial portion of the particles are typically located in the center region of the image. Although not by any means an infallible rule, “center groups” or “families” have been observed in image after image. A “classical” example of such a group has been chosen and sketched for Fig. 6. For droplets smaller than about 0.7 mm, the spot area and the number of particles collected were generally too small to encourage such classification.

When the images have been elliptical, these particle groups have been located at times near the center, but somewhat closer to one end of the spot. This evokes a picture of a drop which strikes the sampling paper at an angle, leaving the leading fluid and its particles on the paper at the point of contact. This picture places the collected particles on the lower surface of the drop.

As the drop spreads upon the paper, its surface will tend to remain intact, due to the surface tension, with divergence of the fluid taking place at the top of the drop. Since the final spot diameter is more than three times the drop diameter, the particles on the original surface of the lower drop hemisphere will be found in the center region of the spot. The particles near the equator and in the upper hemisphere of the drop may be mostly located outside this center area.

Particles were frequently found in physical contact with each other, usually in pairs. There were also occasionally numerous particles found in contact. The reasonable assumption was made that particles in contact in the image were in contact when scavenged. “Pairs” and “trios” and agglomerates were accordingly treated as single particles of irregular shape.

The possibility that agglomerates break up following the collection does exist, but this apparently is infrequent as evidenced by the number of agglomerate particles well separated from all other particles in the same image, and even alone in the image in the case of the smaller drops. A deliberate attempt to break up and relocate moderate-size groups of particles by moving a cover slide was not successful.

The best indication that particles do not leave the drop on impact was provided in pilot scavenging experiments made while investigating dry dust dispersal. Tracers 2210, 2330 and a cadmium sulfide tracer, 1757, were dispensed as a dry dust in light natural rains.

![Fig. 6. Magnified raindrop image showing grouped particles.](image-url)
Hundreds of particles were collected by each of the drops, but with only one drop of the many reviewed was a significant number of particles found outside the image.

7. Calculation of efficiencies

Mathematical definitions of the scavenging efficiencies. The average number of particles in the cylinders swept by raindrops of cross section A is

$$\frac{A}{FG} \int [H/E(k)]dz.$$  (3)

Here, H is the number of particles collected on a filter, E(k) is the correction for the anisokinetic error of the collection (where k refers to the inertial parameter of the particle-filter system), and FG is the volume of air sampled in the time G.

The scavenging efficiency is the ratio of the average number of particles collected by the drops to the number given by (3), or

$$E(D,d) = \frac{(N_r - B b)G}{A} \int f[H/E(k)]dz.$$  (4)

In this equation, N is the total number of particles found in a raindrop of diameter D, B is the number of those resulting from dry deposition on the sample, and r, b and f are the proportions of Nr, B and H in a given size range centered on particle diameter, d. Both E(k) and f are dependent upon wind speed, and, therefore, upon elevation, and so are placed within the integral. Because H represents an average of the concentration over time, the bar over (Nr - Bb)/A stands for an average over the same time interval. E(D,d) will be called “absolute efficiency” for brevity.

If E(D,d) is divided by E(D0,d), where d is constant but D is allowed to vary from some reference drop size, D0, one obtains

$$E(D/D_0) = \frac{(N_r - B b)A_0}{(N_r - B b)D_0 A},$$  (5)

which is the “relative efficiency” with respect to drop size.

Computation of absolute efficiency. The raw data needed to calculate Eq. (4) appears in Engelmann (1963). The continuous samples were roughly divided into 15-sec intervals for the counting. The values of r and b were assumed to be constant through a test. The particle size distributions from each test were plotted on logarithmic probability paper, fitted with a straight line, or lines, by eye, and the smoothed distributions used to obtain r and b.

The drop sizes utilized from tests R3, R13 and R15 were limited to small ranges of the impact parameter. However, the number of drops collected was not the same for each time interval. To avoid biasing the efficiency with the cloud concentration existent when the greatest number of drops was collected, N/Db was computed for each time period, and then averaged again over the time of the test. Values of H were calculated for each filter by summing the particles in annuli, as obtained by counting a strip across each filter.

The integral \( \int H dz \) was graphically determined over four height intervals in R3 and R15, and over two intervals in R13. The particles on a filter near the midpoint of each height interval were sized to obtain a representative value of f. Corresponding wind speeds were estimated through interpolation of the wind profile, and for these speeds, values of f(E(k) were taken from the anisokinetic error as measured in the wind tunnel.

These values of f(E(k) were assumed to be representative of their height intervals, were multiplied by \( \int H dz \), and the products summed to approximate the desired integral.

Tests R3 and R15 thus provided the absolute collection efficiencies of two sizes of drops on zinc sulfide particles from 3 to 15 microns in diameter. The absolute efficiencies could then be estimated for other sizes of drops by means of the more easily measured relative efficiency.

Computation of relative efficiency with respect to drop size. The wind separated the artificial rain of tests R7 through R10 so well that the average size of the spots could be used to determine \( \bar{A} \) for each sample, and the quantity \( \bar{N}/\bar{A} \) substituted for \( (N/A) \) in Eq. (5).

The particle size distributions for each drop size and background were plotted on logarithmic probability paper, straight line segments fitted to the points, and “smoothed” distributions taken therefrom for use as r and b. This provided a relative efficiency between the

<table>
<thead>
<tr>
<th>Test</th>
<th>Drop sizes (mm)</th>
<th>3.45</th>
<th>4.9</th>
<th>6.9</th>
<th>9.8</th>
<th>13.8</th>
<th>19.5</th>
<th>27.6</th>
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<td>R7T1</td>
<td>E(0.77)/E(1.13)</td>
<td>0.76</td>
<td>0.76</td>
<td>0.90</td>
<td>0.99</td>
<td>1.57</td>
<td>1.62</td>
<td>1.34</td>
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<tr>
<td>R8T1</td>
<td>E(0.50)/E(0.70)</td>
<td>3.13</td>
<td>4.20</td>
<td>3.74</td>
<td>2.88</td>
<td>2.10</td>
<td>1.05</td>
<td>0.47</td>
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<td>R8T2</td>
<td>E(0.70)/E(0.81)</td>
<td>0.78</td>
<td>0.68</td>
<td>0.78</td>
<td>1.02</td>
<td>1.26</td>
<td>2.09</td>
<td>3.17</td>
</tr>
<tr>
<td>R9T2</td>
<td>E(0.66)/E(0.85)</td>
<td>1.08</td>
<td>1.23</td>
<td>1.31</td>
<td>1.37</td>
<td>1.28</td>
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<td>0.97</td>
</tr>
<tr>
<td>R10T1</td>
<td>E(0.42)/E(0.62)</td>
<td>1.66</td>
<td>1.72</td>
<td>1.54</td>
<td>1.45</td>
<td>1.20</td>
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<td></td>
<td>E(0.62)/E(0.88)</td>
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<td>0.92</td>
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<td>1.42</td>
<td>1.41</td>
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<tr>
<td></td>
<td>E(0.42)/E(0.64)</td>
<td>1.20</td>
<td>1.37</td>
<td>1.58</td>
<td>1.64</td>
<td>1.59</td>
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<td>1.12</td>
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</table>
two or three drop sizes in each test for each of the particle size class ranges, as given in Table 2.

A family of curves was then constructed for efficiency versus drop diameter (each curve for a different particle diameter). These curves passed through the measured absolute efficiencies of R3 and R15, and were such that the relative efficiencies of Table 2 were generally satisfied. Some deviations from the curves had to be accepted, and the very high apparent efficiency of the 0.5-mm drop in R8T1 was rejected.

Errors. The variance of the ratio of two variables can be approximated by the propagation of error formula. A 95 per cent confidence interval about \( \langle N_1/N_2 \rangle \) can then be estimated from twice the square root of the variance, using the Gaussian curve statistics.

The resultant confidence intervals are broad enough to permit doubt as to the certainty of many of the ratios. The values for R9T2 seemed to be established, however, and these show the smaller drops to be the more efficient. In addition, although individual tests may be of rather dubious accuracy, they are consistent with each other, and thereby they increase confidence in the higher efficiency of the smaller drop.

There were also formed the three cumulative percentage size distributions (unsmoothed) of the combined gross drop contents in tests R7–R10, for the drop sizes 0.36–0.50, 0.62–0.81 and 0.88–1.13 mm diameter. The Kolmogorov–Smirnov test for goodness of fit rejects the hypothesis that any two of these distributions are alike at about the 2 per cent level of significance. It was concluded that the particle sizes collected are truly different, with the collection of the smaller drop possessing a larger mean size of particle.

Considerable care was exercised in the counting, sizing, and reduction of the data, and errors from these sources are probably held to 10 per cent, with many errors being minimized in the averaging processes. However, the error in measuring and then in applying the anisokinetic correction, \( E(\bar{k}) \), may easily cause the calculated scavenging efficiencies to be in error by 20 per cent of their values, especially for the larger particles. The efficiencies reported here have benefited from more recent estimates of \( E(\bar{k}) \) than were available to Engelmann (1963). Consequently, the following Figs. 7 and 8 are somewhat changed from those given there.

8. Efficiency versus raindrop size and parameter \( \alpha' \theta \)

In Fig. 7 appear the absolute efficiencies measured for tests R3 and R15, and those calculated for other drop sizes on the basis of the relative efficiencies. Each curve in the figure applies to a particular zinc sulfide particle diameter, as labelled. Since, for a given drop size, the impaction parameter depends only on \( \alpha' \theta \), the curves have also been labeled with these values. For example, the curves for 4.1 density zinc sulfide particles should apply to 1.0 density water particles of about twice the diameter, the value of \( \alpha' \theta \) being the same for both cases. The units of micron\(^3\) g cm\(^{-3}\) for \( \alpha' \theta \) will be consistently used in this paper.

Plotted are also the laboratory measurements of Kinzer and Cobb. The most frequent water particle in their cloud was about 13 microns diameter \( (\alpha' \theta = 42) \), with a range of 7.8 to 28 microns. Langmuir’s prediction for \( \alpha' \theta = 42 \), using his interpolation formula, appears on the figure for comparison. Also plotted are two of the measurements made by Walton and Woolcock with small drops falling at near-terminal velocity through clouds of methylene blue particles. The values of \( \alpha' \theta \) for these two measurements were 49 and 789.

Fig. 7. Scavenging efficiencies versus drop diameter and \( \alpha' \theta \).

Fig. 8. Scavenging efficiencies for zinc sulfide versus \( K \).
The zinc sulfide data consistently indicate a higher efficiency for 0.4-mm drops. The data of Kinzer and Cobb show a peak in collection efficiency in the vicinity of 0.4-mm drop diameter. Test R10T1 also shows a peaking at 0.4 mm, but only for small values of \( \alpha \) for this is the only test in which drops smaller than 0.4 mm were adequately sampled.

9. Measured efficiency versus impaction parameter

In Fig. 8 are given Langmuir's predictions for potential and viscous flows as functions of \( K \). Also entered on the figure are the results of tests R3, R13, and R15.

Even though there were too few drops sampled during R13 to allow the calculation of an absolute scavenging efficiency, there were sufficient particles to yield a size distribution, \( r \). Division of this size distribution of the scavenged particles by that of the particles on the filters, after correction for anisokinetic error, reveals the collection efficiency with particle size, or with the parameter \( K \). In order to plot these data on the figure, it was assumed that the absolute efficiency of R13 and R15 were the same for 6.91-micron particles.

For comparison, the results are also given for 2210 and 2330 from McCully et al. (1956) who determined collection efficiencies by releasing tracers from aircraft in natural rain, and collecting bulk samples at the ground. These data, therefore, represent some sort of average over drop size. These tests were made using tracers of two different size distributions, U. S. Radium Corporation's 2210 and 2330, and the results showed that the tracers were scavenged very differently in their small area of overlap (8 to 10 microns).

The collection efficiency for the 2210 was found to be on the order of 0.2, and for 2330 about 1.5. This was attributed to a difference in retention resulting from a difference in wettability. The tracers, however, are both zinc sulfide and both have copper as an impurity. The only difference in the manufacture of the two is the temperature at which they are fired, so that these tracers should have the same wettability. McCully's data could thus be interpreted as showing a rapid increase in efficiency in the vicinity of \( K = 10 \). There are very few large particles in tracer 2210, and error is therefore more likely in this region.

10. Explanation of the observed efficiencies

The predominant features of the scavenging efficiencies reported here are

1) the high efficiencies for the 0.4-mm diameter drop,
2) efficiencies greater than 1.0 for particles larger than 13 microns, and
3) a more rapid increase in efficiency with particle size in the general region of 10 microns diameter.

The author proposes that these features can be adequately explained with the classical inertial impaction of particles on the leading side of the drops, and wake effects, as follows.

Those particles which nearly reach the raindrop as it passes gain further opportunity for contact when fluid convergence and weak electrical attractions carry the particles into the wake vortex. In the case of small raindrops, this vortex remains attached to the drop as it falls, and provides a "large" time interval for collection of the particles on the trailing or upper side of the drop. In the case of large drops, the vortex is shed regular before the particles can impact on the trailing side. (For all sizes of drops, disruption of the wake by turbulence may leave a smaller time available for collection than found in the laboratory.)

While the vortex is attached, particles therein fall with respect to the drop, and may be attracted toward the drop by electrostatic forces. The collection of the heavier and larger particles is thereby favored.

The number of particles subjected to collection is increased if the raindrop carries an electrical charge, since all the particles are subjected to an attractive force regardless of the fluid flow. Even very weak charges may be effective in drawing into the wake those particles that have narrowly missed the drop. The 1.5 efficiency observed by McCully may be that efficiency which can be obtained with inertial and wake effects coupled with the weak electrical charges that occur in natural rainfalls.

It is not necessary to invoke wettability differences to explain these results, since the increase in efficiency toward 1.5 was observed with tracer 2210, alone, as well as with 2210 and 2330 in combination. Of course, this does not exclude the possibility that the retention efficiency may be less than 1.0, and the target efficiency correspondingly greater than the observed efficiencies.

The wake-effect argument has further support in that the data for zinc sulfide agree with that taken with water particles by Kinzer and Cobb, at least in the high efficiency with the 0.4-mm drop. Then, too, a contribution in the wake has been mathematically demonstrated by Pearcey and Hill for smaller drops, and actually observed by Telford et al. (1955), Rosinski and Nagamoto (1961), Magarvey and Geldart (1962), Woods and Mason (1965), and, perhaps, even by Walton and Woolcock.

Möller's research on larger spheres in water has shown separation or shedding of the wake vortices at a Reynolds number of 450, corresponding to a raindrop diameter of 1.35 mm. There was some elevation of the number by the trough in his experiments. The Reynolds number where shedding occurs from a cylinder is about 75, as inferred from photographs published in Goldstein (1938) and Tietjens (1934). This corresponds to a raindrop of 0.5- or 0.6-mm diameter. These diameters do not conflict with the location of the apparent maximum efficiency in Fig. 7.
11. Future research

The data come from only a few tests in artificial rain, and it would be well to obtain data in natural rainfalls using additional tracers. There may be significant changes in electrical charge, and there may exist differences in retention efficiency dependent upon particle “wettability” and charge, or upon drop size. Although greater accuracy and proof of theory may come from laboratory measurement, measurement made in the field with natural rain removes much doubt as to the applicability of the test results to later prediction.

Particularly advantageous would be a portable testing system, which could be taken to sites with rain or snow, and easily oriented with the wind direction. The scavenging power of snow appears to have been overlooked in the literature, and this is a serious omission. The measurements of efficiencies for the variety of individual crystals should, perhaps, be preceded by measurements of the bulk washout coefficient for various snow classifications and precipitation rates.

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