Scavenging of Aerosol Particles by Precipitation[1]

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ABSTRACT

Airborne measurements have been made of aerosol particle size distributions (>0.01 μm) in aged air masses, in the plumes from several coal power plants and a large Kraft paper mill, and in the emissions from a volcano, before and after rain or snow showers. These measurements have been used to deduce the precipitation scavenging collection efficiencies of aerosol particles ranging in size from ~0.01 to 10 μm diameter.

Despite large variations in the nature of the aerosol particles and the precipitation, the scavenging collection efficiencies as a function of particle size showed marked similarities, with some well-defined maxima and minima values. The measurements agree well with theoretical calculations for aerosol particles > 1 μm, but for the submicron aerosol particles the scavenging collection efficiencies are generally much higher, and the region of very low scavenging efficiencies (the "scavenging gap") much narrower, than current theories predict. Some possible explanations for these discrepancies are suggested.

1. Introduction

The scavenging of particles in the air by precipitation is one of the major processes by which the atmosphere is cleansed and a balance maintained between the sources and sinks of atmospheric aerosol particles. It has been estimated that in temperate latitudes precipitation scavenging accounts for 70–80% of the mass of the aerosol particles removed from the troposphere (SMIC, 1971).

Particles may be scavenged by precipitation due to inertial impaction, electrical and phoretic effects, Brownian motion, and by serving as cloud condensation nuclei. Consequently, the scavenging collection efficiency (E) varies markedly with the size (D_p) of the aerosol particles. In the last few years considerable progress has been made in theoretical studies of these scavenging mechanisms which have led to predictions of the functional dependence of E on D_p (e.g., Grover *et al.*, 1977; Wang *et al.*, 1978; Leong and Beard[2]). However, corresponding field measurements have been lacking. In this paper we describe field studies of the scavenging of "natural", man-made and volcanic aerosol particles by precipitation from which scavenging collection efficiencies have been deduced.

2. Experimental procedures

Measurements were made aboard the University of Washington's Douglas B-23 research aircraft [see Hobbs *et al.* (1979) for full description of this facility]. The size spectra of aerosol, cloud and precipitation particles were measured with six instruments which, in overlapping steps, covered a size range from 0.01 to 4500 μm diameter (Table 1). Measurements from these instruments were integrated, processed and displayed by a minicomputer aboard the aircraft. In addition to these automatic probes, a metal foil impactor provided imprints of precipitation particles > 300 μm and a Formvar replicating system provided information on the types of precipitation.

Two different sampling and processing techniques were used for the aerosol particles. Those to be sized by the electrical mobility and Royco 90° light-scattering instruments, which cover a size range from 0.01 to 10 μm, were first passed through a nearly isokinetic entry port into a large (28 ℓ) electrically-neutralized rubber bag (this permitted acquisition of essentially point air samples). These air samples were then passed through a diffusion dryer and from there into the two aerosol particle size-measuring instruments. The aerosol particles to be measured by the Royco forward-scattering probe (1.7–40 μm) were sampled isokinetically, dried in a heated chamber, and then sized in the Royco counter.

Most of the measurements to be described in this paper were obtained in substantial particulate

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[1] Contribution No. 534.

[2] Leong, K. H., and K. V. Beard, 1978: The hydrodynamic influence on phoretic scavenging of aerosol particles by cloud drops. *Preprints Conf. Cloud Physics and Atmospheric electricity*, Issaquah, Amer. Meteor. Soc., 49–52.

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TABLE 1. Instruments used to measure the size spectra of airborne particles and precipitation.

Instrument	Technique for measurement	Size range (μm)
Thermal Systems, Inc., Model No. 3030	Electrical mobility analysis	0.01 to 1 (in 11 size intervals)
Royco Inc., Model No. 202 (modified)	Single-particle 90° light-scattering	0.3 to 10 (in 15 size intervals)
Royco Inc., Model No. 245 (modified)	Single-particle forward light-scattering	1.7 to 40 (in 15 size intervals)
Particle Measuring Systems, Model No. ASSP-100	Single-particle forward light-scattering	2.8 to 66 (in 15 size intervals)
Particle Measuring Systems, Model No. OAP-200X	Single-particle diode occultation	20 to 300 (in 15 size intervals)
Particle Measuring Systems, Model No. OAP-200Y	Single-particle diode occultation	300 to 4500 (in 15 size intervals)

plumes. In these cases, the size distributions of the aerosol particles were made at a point along the centerline of the plume just before and just after the plume was intercepted by a rain or snow shower. The number of occasions on which measurements of this type can be obtained are quite rare. Consequently, the measurements to be described were obtained over a period of three years as suitable opportunities presented themselves during the course of other airborne studies. In the case of measurements of the scavenging of "natural" aerosol particles, the aircraft was flown around the perimeters of showers in order to obtain particle size spectra measurements in the air upwind and downwind of the showers. In both types of measurements the presence of vertical wind shear was needed in order to separate the scavenged air from the precipitation. The scavenging time was determined by either the wind shear between the height at which the plume (or natural aerosol) was sampled and the cloud height, or by observation of the time required for a shower to traverse across a visible plume.

Measurements of raindrop size distributions were usually obtained during a pass through the precipitation (after measuring particle size distributions in the undisturbed air and prior to measurements of the scavenged air). The rain rate was calculated from the resulting averaged size distribution with the terminal velocities corrected for air density.

3. Results

The conditions under which suitable measurements were obtained are listed in Table 2. Fig. 1 shows an example of the spectrum of aerosol particle sizes measured in the plume of a coal-fired electric power plant before and after the plume was scavenged by a rain shower, and Fig. 2 shows similar measurements for natural aerosol particles. Although the

TABLE 2. Conditions under which measurements were obtained.

Date	Source of scavenged aerosol particles	Location	Nature of cloud and precipitation scavengers	Scavenging time (s)	Average rain rate (mm h^{-1})
13 May 1974	Kraft-process paper mill	Port Townsend, Washington	Cumulonimbus with low cloud base; rain shower.	960	7
25 March 1976	Natural	Near Centralia, Washington	Cumulonimbus with medium cloud base; rain shower.	480	18
10 May 1976	Coal-fired power plant	Near Centralia, Washington	Stratocumulus precipitation with an orographic component; rain.	(a) 400 (b) 265	10
1 July 1976	Natural	Near Miles City, Montana	Cumulonimbus cluster, high cloud base, lightning discharges; heavy rain showers.	(a) 1800 (measurements taken at 3 km MSL) (b) 1100 (measurements taken at 2.5 km, MSL)	9
21 April 1977	Emissions from a volcanic maar	Near King Salmon, Alaska	Cumulonimbus with low cloud base; graupel shower.	300	12
29 June 1977	Coal-fired power plant (2100 MW)	Near Farmington, New Mexico	Isolated cumulonimbus with high cloud base; short period of light rain showers.	600	8

particle size spectra for these two cases are quite different, their responses to scavenging are similar. Both show minimal differences between the scavenged and the unscavenged aerosol particles in a narrow size region around 1 μm diameter and both show marked scavenging effects for particles smaller and greater than 1 μm.

Shown in Fig. 3 are the percentages of aerosol particles of various sizes removed by precipitation for all the cases we have studied. The measured size distributions of the precipitation scavengers for each of these cases are shown in Fig. 4.

The concentration of aerosol particles $X(D_p)$ with diameters between D_p and $D_p + dD_p$ at time t after scavenging is related to the concentration $X_0(D_p)$ at $t = 0$ by

$$X(D_p) = X_0(D_p) \exp[-\Lambda(D_p)t], \qquad (1)$$

where $\Lambda(D_p)$ is the (size-dependent) scavenging rate.

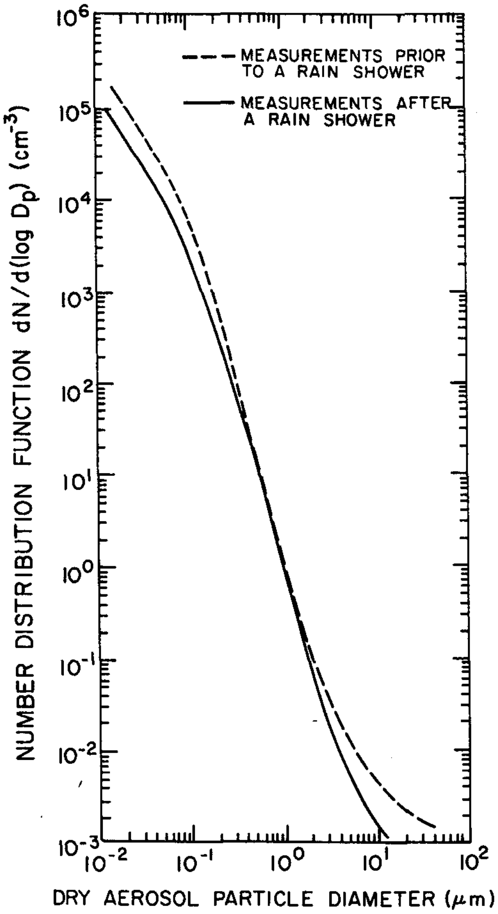

FIG. 1. Airborne measurements of the size spectra of aerosol particles in the plume from the coal-fired electric power plant at Centralia, Washington measured at 5 km (12 min travel time) downwind of the stack on 10 May 1976, before and after the plume was intercepted by a rain shower.

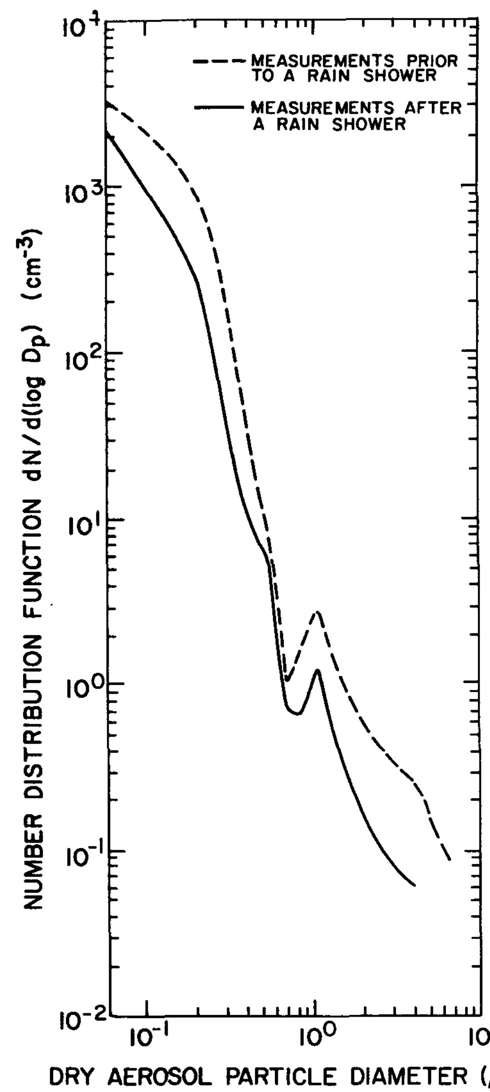

FIG. 2. As in Fig. 1 except for "natural" aerosol particles measured at an altitude of 2.5 km MSL near Miles City, Montana, on 1 July 1976.

The scavenging collection efficiency $E(D_p,D)$ of aerosol particles of size D_p by precipitation particles of size D is derived from

$$\Lambda(D_p) = \int_0^\infty A(D)E(D_p,D)V(D)N(D)dD, \qquad (2)$$

where $A(D)$ is the effective cross-sectional area of a precipitation particle, $V(D)$ its terminal fallspeed, and $N(D)dD$ the concentration of precipitation particles, with diameters between D and $D + dD$. If, as a first approximation, it is assumed that E is only a function of the aerosol particle size D_p, then it is given to reasonable accuracy by

$$E = \Lambda(D_p)[\Sigma A_i(D)V_i(D)N_i(D)\Delta D_i]^{-1}, \qquad (3)$$

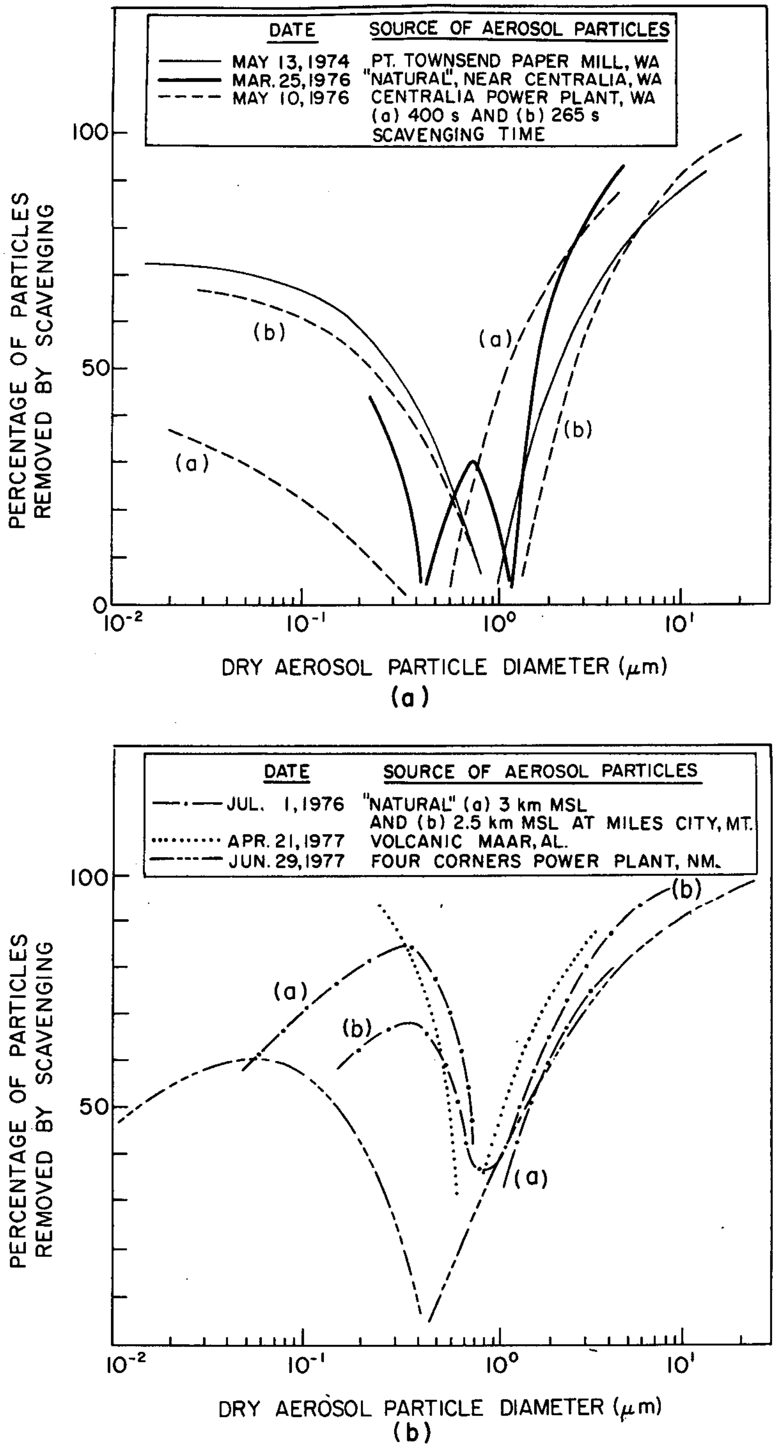

FIG. 3. Percentages of aerosol particles of various sizes removed by precipitation scavenging. See Table 2 for details on conditions under which measurements were obtained and Fig. 4 for size distributions and rainfall rates of the precipitation scavengers.

where i indicates a size interval for the precipitation particles.

We also tried a more elaborate scheme to determine E in which the precipitation particle size spectra were fitted to a Marshall-Palmer distribution and E was assumed to be distributed log-normal with respect to the size of the precipitation particles. [The studies of Wang *et al.* (1978) provide some support for the latter assumption for drops of radius ~ 100 μm.] The scavenging collection efficiency was then fitted (using a chi-squared minimization technique) to all of our data combined for each aerosol particle class over the spectra of precipitation particles. This fit showed E to be essentially independent of the size of the precipitation particles. However, when the assumption of a scavenging collection efficiency independent of precipitation particle size was applied separately to each of our six sets of data, those cases with high concentrations of small precipitation particles exhibited the higher E values. In view of the widely varying conditions under which our measurements were obtained, we will treat each data set individually with the assumption that E is independent of precipitation size within a particular data set.

The values of E so derived are shown in Fig. 5. It can be seen that, despite the wide variety in the nature of the particles scavenged and the scavengers themselves, the scavenging collection efficiency curves show marked similarities, with a minimum in E near 1 μm (we will call this the "scavenging gap").[3]

A scavenging collection efficiency with the general shape of the curves shown in Fig. 5 should have a strong impact on the development of the size distribution of atmospheric aerosol particles. Thus, outside of the range of the scavenging gap, particles should be efficiently scavenged by precipitation, but within the scavenging gap removal of particles from the air by precipitation is very inefficient. Consequently, we should expect to see relatively high concentrations of particles with dry diameters of ~0.5–1 μm. This size range corresponds reasonably well with that in which peaks in concentrations are regularly observed in atmospheric aerosol—the so-called accumulation mode (Willike and Whitby, 1975).

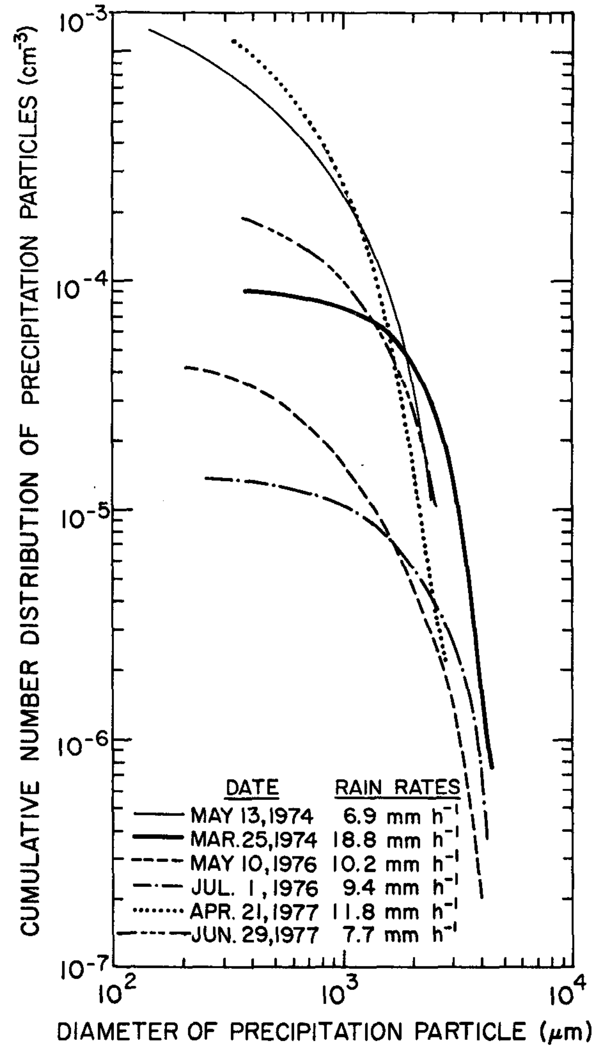

FIG. 4. Size distributions and rainfall rates of the precipitation scavengers for the data shown in Fig. 3.

4. Comparisons with previous studies and discussion

Shown in Fig. 6 for comparison with our measurements are a number of theoretical calculations, and one set of laboratory measurements, of the scavenging collection efficiency as a function of the diameter of the scavenged aerosol particles.

For particles ≥ 1 μm in diameter, where inertial impaction dominates scavenging, our measurements are in good agreement with the theoretical results of Dana and Hales (1976) and Wang *et al.* (1978) and the laboratory measurements of Lai *et al.* (1978). As far as the right-hand edge of the scavenging gap is concerned, our measurements place this at a *dry* aerosol particle diameter between 0.5 and 2 μm. Wang *et al.*'s theoretical calculations place this edge of the scavenging gap between 0.5 and 2 μm for monodispersed scavenging drops 84 and 620 μm in diameter, respectively, and Leong and Beard[2] place it at ~8 μm for scavenging drops 80 μm in diameter (Fig. 6). Laboratory measurements by Lai *et al.* (1978), also shown in Fig. 6, indicate the right-hand edge of the scavenging gap to be near 0.6 μm. Our measurements (Fig. 5) show the scavenging gap to be only a few tenths of a micrometer wide. The theories of Wang *et al.* for 84 μm scavenging drops and Leong and Beard for 80 μm scavenging drops predict a similarly narrow scavenging gap, as do Lai *et al.*'s laboratory measurements (Fig. 6). However, for the larger scavenging drops the theories

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[3] Errors in the derived values of E due to uncertainties in the measurements of the size spectra of aerosol particles and precipitation scavengers are ~15%; other sources of error (impossible to quantify) may be greater. Since the principal feature of the E curves is their strong dependence on the size of the aerosol particles, removal processes that are not strongly size dependent (e.g., advection) appear to have little effect on our measurements.

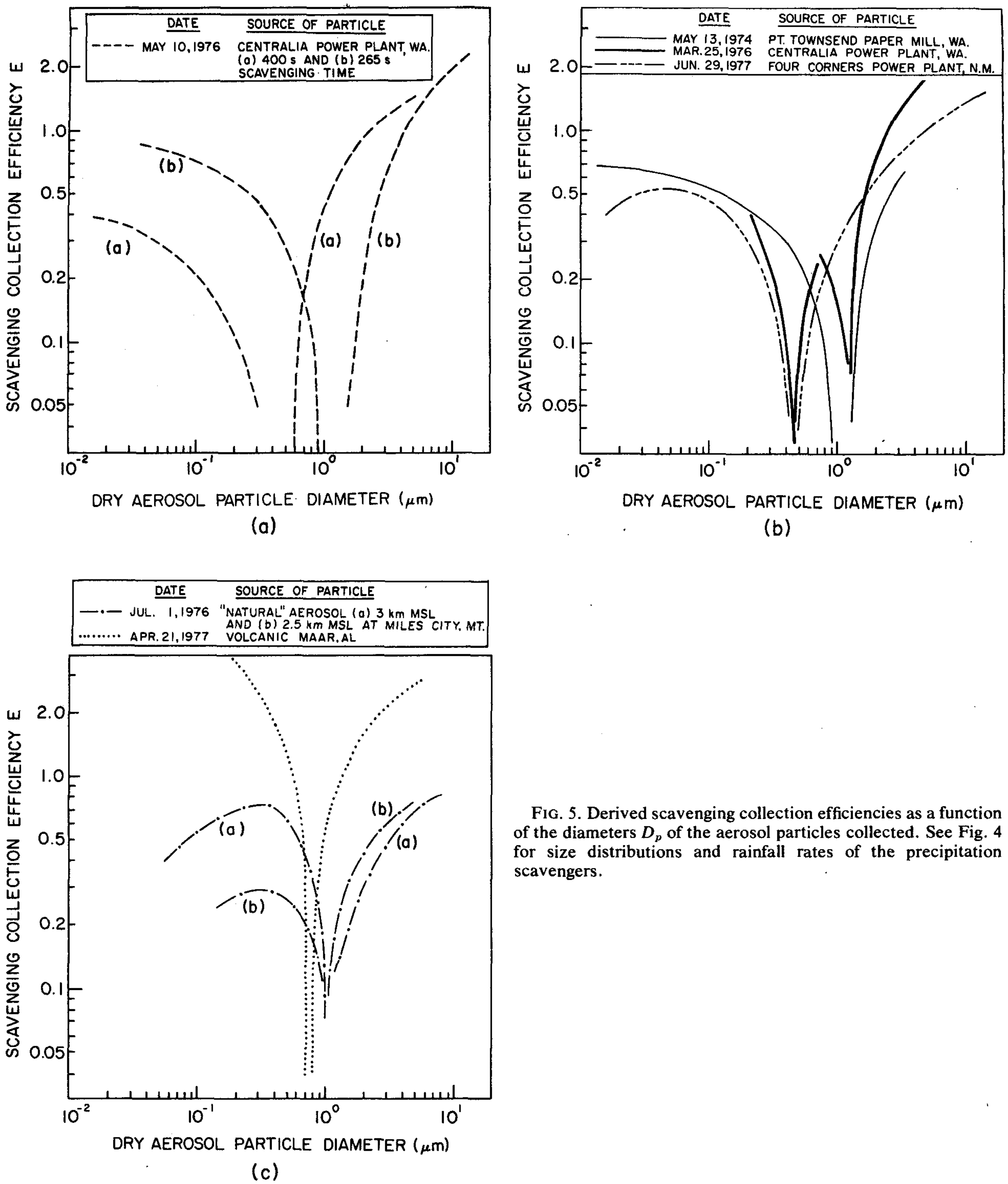

FIG. 5. Derived scavenging collection efficiencies as a function of the diameters D_p of the aerosol particles collected. See Fig. 4 for size distributions and rainfall rates of the precipitation scavengers.

predict a much broader scavenging gap (e.g., the left-hand branches of the E curves for Wang *et al.*'s results for 620 μm drops and Dana and Hales' result for 500 μm drops are too far to the left to be plotted on Fig. 6).

The principal apparent discrepancy between our field measurements and the theoretical predictions is that our derived values for E for submicron aerosol particles are generally about an order of magnitude larger than the theories predicted for scavenging by

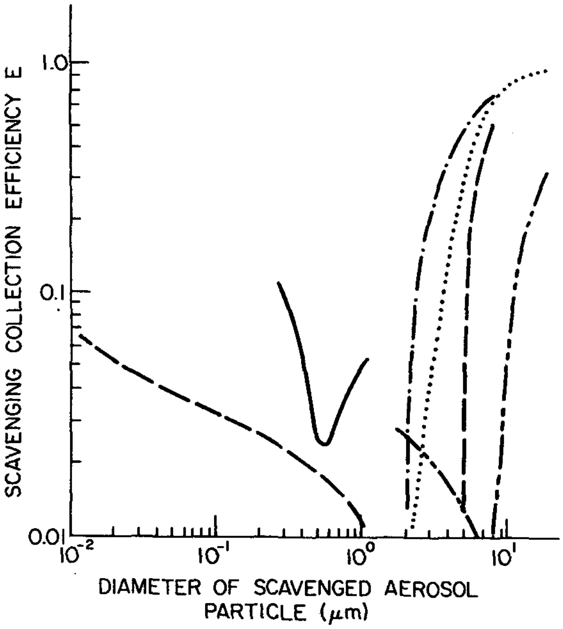

FIG. 6. Scavenging collection efficiencies predicted theoretically by Wang *et al.* (1978) for scavenging drops 84 μm (– – –) and 620 μm (–·–·–) in diameter, by Leong and Beard[2] for 80 μm in diameter (— — — —) and by Dana and Hales (1976) for 50 μm diameter drops (·····). The solid line shows the results from laboratory measurements by Lai *et al.* (1978) for scavenging drops 620 μm in diameter (——).

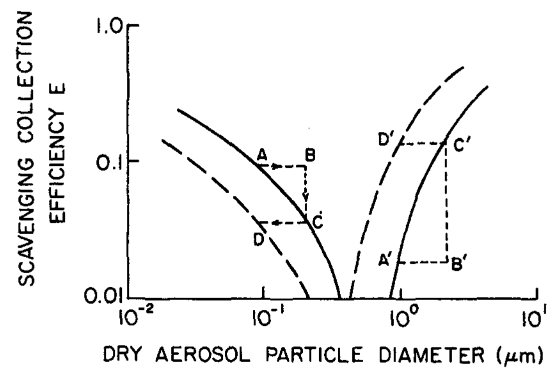

FIG. 7. Schematic showing the effects of deliquescent growth on collection efficiency. The solid curve shows E as a function of the dry size of the aerosol particles for the case when the particles are dry when they were scavenged in the atmosphere. The dashed curve shows E as a function of the dry size of the aerosol particles for the case when the particles are scavenged in the atmosphere after they have deliquesced and increased in size. The lettered and arrowed lines indicate how the dashed curve is constructed from the solid curve.

Brownian and phoretic effects alone. In fact, outside of the narrow scavenging gap, our derived E values lend support to Slinn's (1974) contention that E could be assumed to be close to unity for submicron particles.

Some of the differences between our measurements and the theoretical predictions might be due to the fact that we measured the size of the aerosol particles after they had been dried, whereas if the particles deliquesced, they would have scavenged at larger sizes in the atmosphere.[4] Fig. 7 illustrates schematically the effects of deliquescent growth on the scavenging collection efficiency. In the hypothetical case shown in this figure, an aerosol particle at A with a dry diameter of 0.1 μm increases to ~0.2 μm diameter (point B) due to deliquescence in the atmosphere, and the efficiency with which it is collected is given by point C. This would give rise to point D on a curve of E versus the dry aerosol particle diameter. However, a 1 μm diameter particle at A′ which increases in size by deliquescence to ~2 μm (point B′) would be collected with an efficiency given by point C′. Thus, deliquescent growth enhances the scavenging collection efficiency of those aerosol particles which grow to sizes that place them to the right of the scavenging gap, but it decreases the collection efficiencies of aerosol particles which grow to sizes that place them to the left of the scavenging gap. Hence, for a reasonable range of subcloud relative humidities, small aerosol particles might grow large enough to be efficiently removed by inertial impaction. This might explain in part the relatively high values of E shown in Fig. 5

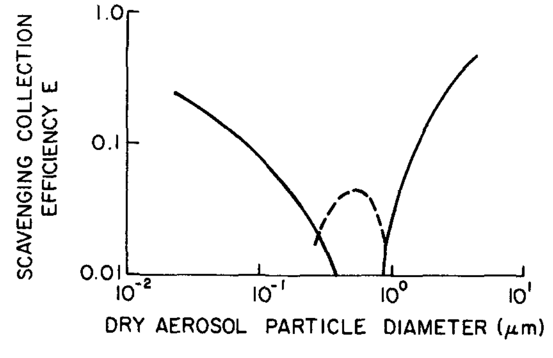

FIG. 8. Schematic showing the effect of deliquescent growth on the scavenging collection efficiency for aerosol particles in which the fraction of deliquescent material decreases with increasing particle size, becoming near zero for aerosol particles ~ 1 μm in diameter. The solid curve shows the scavenging collection efficiency without deliquescent effects; the dashed curve shows the *additive* effect of deliquescent growth.

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[4] There is strong evidence that the submicrometer atmospheric aerosol generally contains a significant soluble component of ammonium sulfate and acid sulfates (e.g., Wagonner *et al.*, 1976): the submicrometer particles in the plumes from coal-fired power plants are largely acid sulfates (Stevens *et al.*, 1978), the effluents from volcanoes contain large numbers of small soluble particles (Hobbs *et al.*, 1977, 1978; Cadle *et al.*, 1978; Stith *et al.*, 1978), and there are numerous hygroscopic particles in the plumes from paper mills (Hindman *et al.*, 1977). Hence, the submicron aerosol particles on which we obtained measurements probably deliquesced near cloud base or in the presence of precipitation.

for aerosol particles with dry diameters down to ~0.2 μm.

The peak values of E at $D_p = 0.3\text{–}0.4$ μm and $D_p = 0.8$ μm which were measured on 1 July 1976 and 25 March 1976, respectively (Fig. 5), might be explained in part if the aerosol particles contained a decreasing fraction of deliquescent material with increasing particle size (as suggested schematically in Fig. 8). There is, in fact, some evidence for natural aerosol particles having such characteristics (Dzubay and Stevens, 1975; Kadowaki, 1976; Patterson and Wagman, 1977; Whitby, 1978; Charlson *et al.*, 1978).

While deliquescent growth is capable of explaining some features of our E curves, it cannot explain the large difference between the observed and theoretical values of E for $D_p \leqslant 0.1$ μm. This difference might be due to nucleation scavenging, particularly in the case of convective clouds since some of the scavenged air just below cloud base (where we made many of our measurements) may have previously been involved in cloud processes. In this case, soluble particles as small as 0.01 μm could have served as cloud condensation nuclei and the droplets that formed on them subsequently removed by the coalescence mechanism. The effect of such nucleation scavenging on our measurements would be to increase the value of E for aerosol particles with $D_p \geqslant 0.01$ μm. This mechanism also might have been responsible for the decrease in E observed on 29 June 1977 for $D_p \geqslant 0.05$ μm (Fig. 5) if decreasing numbers of particles < 0.05 μm served as cloud condensation nuclei. Some of the features of the E curves to the left of the scavenging gap observed on 1 July 1976 (Fig. 5) could also have been due in part to nucleation scavenging, since the measurements at 3 km MSL near cloud base show significantly greater values of E than those at 2.5 km MSL.

Nucleation scavenging also may have played a role in the one case (21 April 1977) of scavenging by solid precipitation shown in Fig. 5. In this case, the derived values of E exceed unity by substantial amounts for both the smallest and largest particles. Since the scavenged particles were effluents from an active volcano which contained several steam vents, they likely acted as cloud condensation nuclei, and were then scavenged very effectively by the heavy shower of graupel.

Finally, we note that our measurements generally agree with recent theory (Wang *et al.*, 1978) and laboratory measurements (Lai *et al.*, 1978) showing the scavenging collection efficiency to be a function of the size of the raindrops (especially to the left of the scavenging gap). Comparison of Figs. 4 and 5 shows that (with the exception of 10 May) the scavenging collection efficiencies for aerosol particles with diameters < 0.2 μm increase as the concentrations of small (<400 μm) raindrops increase.

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REFERENCES

Cadle, R. D., and E. J. Mroz, 1978: Particles in the eruption cloud from St. Augustine volcano. *Science,* **199,** 455–458.

Charlson, R. J., D. C. Covert, T. V. Larson and A. P. Waggoner, 1978: Chemical properties of tropospheric sulfur aerosols. *Atmos. Environ.,* **12,** 39–53.

Dana, M. T., and J. M. Hales, 1976: Statistical aspects of the washout of polydisperse aerosols. *Atmos. Environ.,* **10,** 45–50.

Dzubay, T. G., and R. K. Stevens, 1975: Ambient air analysis with dichotomous sampler and XRF spectrometer. *Environ. Sci. Tech.,* **9,** 663–668.

Grover, S. N., H. R. Pruppacher and A. E. Hamielec, 1977: A numerical determination of the efficiency with which spherical aerosol particles collide with spherical water drops due to inertial impaction and phoretic and electrical forces. *J. Atmos. Sci.,* **34,** 1655–1663.

Hindman, E. E., P. V. Hobbs and L. F. Radke, 1977: Airborne investigations of aerosol particles from a paper mill. *J. Air Pollut. Control Assoc.,* **27,** 224–229.

Hobbs, P. V., L. F. Radke and J. L. Stith, 1977: Eruptions of the St. Augustine volcano: airborne measurements and observations. *Science,* **195,** 871–873.

——, —— and ——, 1978: Particles in the eruption cloud from St. Augustine volcano. *Science,* **199,** 455–458.

——, D. A. Hegg, M. W. Eltgroth, and L. F. Radke, 1979: Evolution of particles in the plumes of coal-fired power plants—I: Deductions from field measurements. *Atmos. Environ.,* **12,** 935–951.

Kadowaki, S., 1976: Size distribution of atmospheric total aerosols, sulfate, ammonium and nitrate particulates in the Nagoya area. *Atmos. Environ.,* **10,** 39.

Lai, K.-Y., N. Dayan and M. Kerker, 1978: Scavenging of aerosol particles by a falling raindrop. *J. Atmos. Sci.,* **35,** 674–682.

Patterson, R. K., and J. Wagman, 1977: Mass and composition of an urban aerosol as a function of particle size for several visibility levels. *J. Aerosol Sci.,* **8,** 269–279.

Slinn, W. C. N., 1974: Precipitation scavenging: Some problems, approximate solutions, and suggestions for future research. *Precipitation Scavenging (1974),* R. G. Semonin and A. W. Beadle, Eds., ERDA Symposium Series, 1–60.

SMIC, 1971: *Report of the Study of Man's Impact on Climate.* The MIT Press, 308 pp.

Stevens, R. K., T. G. Dzubay, G. Russwurm and D. Richel, 1978: Sampling and analysis of atmospheric sulfates and related species. *Atmos. Environ.,* **12,** 55–68.

Stith, J. L., P. V. Hobbs and L. F. Radke, 1978: Airborne particle and gas measurements in the emissions from six volcanoes. *J. Geophys. Res.,* **83,** 4009–4017.

Waggoner, A. P., A. J. Vanderpol, R. J. Charlson, S. Larsen, L. Granot and C. Tragardle, 1976: Sulfate light scattering ratio as an index of the role of sulphur in tropospheric optics. *Nature,* **261,** 120–122.

Wang, P. K., S. N. Grover and H. R. Pruppacher, 1978: On the effect of electric charges on the scavenging of aerosol particles by clouds and small raindrops. *J. Atmos. Sci.,* **35,** 1735–1743.

Whitby, K. T., 1978: The physical characteristics of sulfur aerosols. *Atmos. Environ.* **12,** 135–159.

Willeke, K. W., and K. T. Whitby, 1975: Atmospheric aerosols: size distribution interpretation. *J. Air. Pollut. Control Assos.,* **25,** 529–534.