

## An Experimental Study of the Scavenging of Aerosol Particles by Natural Snow Crystals

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### ABSTRACT

An experimental study of the scavenging of aerosol particles of mean radius  $0.75\ \mu\text{m}$  by natural snow crystals of a few millimeters is carried out. Aerosol particles are spherical indium acetylacetonate particles generated by a modified La Mer generator. Snow crystals are obtained during natural snowfalls. Shapes of snow crystals include needles, columns, broad-branched crystals, stellar crystals, and hexagonal plates. Aerosol particles are dispersed into an aerosol chamber and snow crystals fall through the chamber to scavenge aerosol particles. The collection efficiency of aerosol particles by snow crystals is found to decrease with increasing crystal size for all shapes. This can be explained by the relative strength of the inertial force of particles and the hydrodynamic drag force created by the fall of the snow crystal. Larger crystals would create greater drags during the fall and force the aerosol particle to follow more closely to streamlines and hence reduce the collection efficiency.

### 1. Introduction

Removal of atmospheric aerosol particles by precipitation is one of the most important natural processes in cleansing the air. It is often referred to as *precipitation scavenging*. Both liquid (fog and cloud droplets, raindrops) and solid (cloud ice crystals, snow, graupel, hail) precipitation can perform the scavenging act. We shall call the former rain scavenging and the latter ice scavenging. The problem of rain scavenging of aerosol particles has been subjected to numerous studies and is better understood. Readers are referred to Wang and Pruppacher (1977) and Wang et al. (1978) for summaries of laboratory experimental and theoretical studies of rain scavenging.

Compared to rain scavenging, the process of ice scavenging of aerosol particles is inadequately studied. Some earlier theoretical studies have been carried out and are summarized in a companion paper (Miller and Wang 1989). The present paper will focus on the laboratory experimental studies. The earliest experimental study of ice scavenging was probably the one performed by Starr and Mason (1966) who used paper tissues cut in the shapes of snow crystals and dropped them through an aerosol chamber. The applicability of the results of this study to atmospheric ice scavenging is questionable as the paper flakes cannot duplicate the physical properties of natural snowflakes. Much improved experiments were later performed by Knutson et al. (1976) who conducted a series of experiments using natural snowflakes and artificial aerosol particles

in the Chicago area from 1968 to 1972. However, the possible importance of the phoretic effects (due to nonsaturation humidity) was not known at the time and the relative humidity was not monitored during the experiments. Consequently, the results were difficult to interpret. More recently, Murakami et al. (1985a,b) conducted a series of experiments in Japan using natural snow and artificial aerosol particles ( $0.1$  to  $6.0\ \mu\text{m}$  in diameter). Quite consistent results were obtained but the snow crystals were all of stellar shapes. Since there are many other shapes of snow crystals, it is desirable to determine their collection efficiencies as well. The present study was motivated by this need. The experimental setup and procedure, as well as discussion of results, are presented in the following sections.

### 2. Experimental setup and procedure

The experimental setup used in the present study is similar to that used by Wang and Pruppacher (1977) for studying the scavenging of aerosol particles by raindrops. However, the total dimension of the present setup is smaller due to the fact that snow crystals, unlike raindrops, require relatively short distance to reach terminal velocity. A schematic and relevant dimensions of the setup are shown in Fig. 1.

A modified La Mer generator was used to produce  $0.75\ \mu\text{m}$  radius indium acetylacetonate aerosol particles. These particles are nearly monodispersed and are of spherical shape as evidenced by electron micrographs taken of them after being precipitated out onto a glass slide by a thermal precipitator. The aerosol particles produced by the generator would be flushed into a  $1.21\ \text{m}$  long by  $0.155\ \text{m}$  inner diameter Plexiglas aerosol

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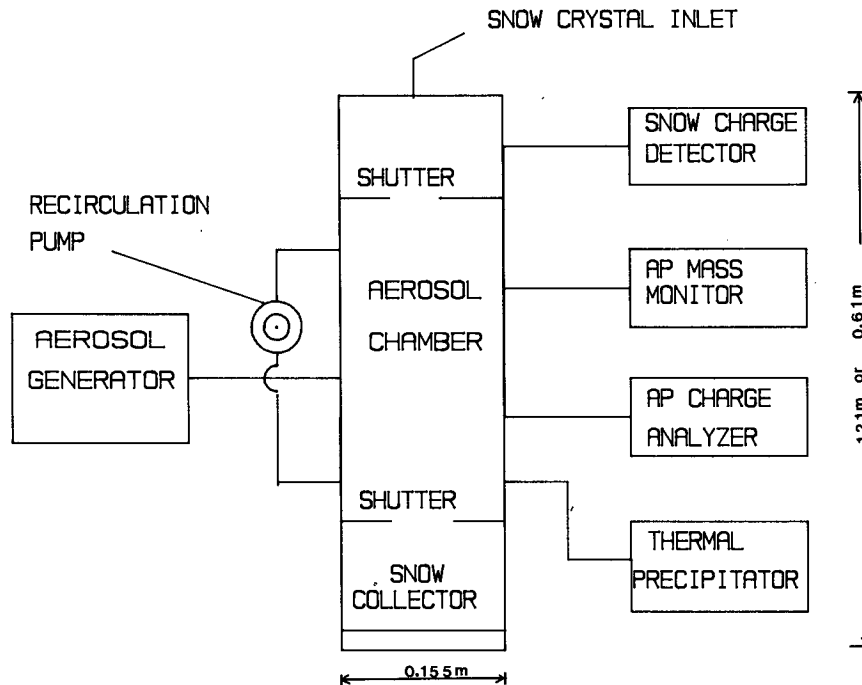


FIG. 1. Schematic of the experimental setup.

chamber fitted with mechanical shutters at both top and bottom. This chamber was built in three pieces. In several experiments the center piece was removed so that the chamber length was 0.61 m. At the time the chamber was being filled, a small air pump was run intermittently to circulate the aerosol in the chamber to ensure a uniform distribution. If the aerosol was to be electrically charged, it was first mixed with ions produced from a corona discharging unit. A mean charge of up to 26 electrons could be placed on each aerosol particle if so desired. Using a technique developed by Dalle Valle et al. (1954), we determined the mean charge of aerosol particles by allowing an aerosol stream to pass through two parallel cylindrical electrodes and measuring the angle of deflection. The potential difference between the two electrodes was maintained at 10 800 volts. Both the amount and the sign of aerosol charge can be determined in this way.

A typical experiment proceeded as follows. Prior or during a promising snowfall, the aerosol generator was turned on. After approximately an hour when the assembly had reached proper temperature (423 K), the aerosol particles were fed into the aerosol chamber. The mass concentration of aerosol particles in the chamber was measured by a TSI Model-3500 mass monitor. The particle size was measured directly under a scanning electron microscope. Typical aerosol concentration in the chamber was between  $10^3$  to  $10^4$  particles  $\text{cm}^{-3}$ . Relative humidity in the chamber was monitored by a dewpoint hygrometer. Next a rectan-

gular sheet of polyethylene was coated with 2–5% (by mass) solution of Formvar. This was quickly placed on a clean sheet of paper on top of an adjustable metal stand beneath the bottom aerosol chamber shutter. A cylindrical hollow piece of rigid cardboard extended approximately 20 cm below this shutter. This was sealed with plastic sheet and tape to keep it airtight once the stand with the Formvar-coated sheet was cranked up tight against it. At this point, both the top and bottom shutters were briefly opened. The chamber humidity was about 85% over ice surface due to some mixing of the original chamber air and the environmental air. Some loss of particles occurred during the opening and the concentration was diluted to 30–50% of the original. Since the concentration was monitored continuously during the whole experiment, its value at the time of scavenging could be determined.

When the snowflakes fell through the aerosol chamber, they first passed through an induction-ring electrometer connected to an oscilloscope. A short pulse would be induced when a charged snow crystal passed through the ring. From the amplitude of the pulse, the flake charge can be evaluated. Small charges were observed on snow flakes ( $10^{-4}$  esu or less). This seems to agree with measurements of Bauer and Pitter (1982) who indicated that 50–90% of unrimed snowflakes, including those several millimeters across, have negligible charges. In addition, in an experimental study done by Magono et al. (1974) in Sapporo, Japan, negligible charges on natural snow crystals were measured. Most

of the crystals retrieved in this study were both unrimed and small. Thus the small charges are probably not surprising.

After the shutters were closed, the polyethylene sheet was carefully removed and kept below freezing for at least 24 hours so that ice crystals were completely sublimated. Some crystals appeared to have melted during the collection process. As long as their size and shape could be determined, they were examined. When an appropriate snow crystal was identified, it and the surrounding polyethylene were carefully cut out. The crystal size was measured and the shape recorded and photographed. Originally, neutron activation analysis was employed to determine the aerosol mass on the snow crystal but the results were unsatisfactory. It was then decided to count the number of particles on a snow crystal directly under a scanning electron microscope. The indium acetylacetonate particles were fairly easy to distinguish from other debris or markings on the crystal due to their size and spherical shape. Overcounting or undercounting of aerosol particles was not deemed to be a serious problem. Several flakes were counted twice, the second time being several days to weeks after the initial count. The dual count gave consistent results. Once the number of aerosol particles on an individual snow crystal was known, the collection efficiency was determined by the following formula:

$$E = n/(ALC), \quad (1)$$

where

- $E$  collection efficiency,
- $n$  total number of aerosol particles on crystal,
- $A$  cross-sectional area of crystal,
- $L$  length of aerosol chamber (either 0.61 m or 1.21 m), and
- $C$  aerosol concentration.

Collection efficiencies determined in this manner are given in section 3.

### 3. Results and discussions

A total of 72 crystals was analyzed and their collection efficiencies determined. Of these, the shapes of 17 are irregular and are excluded. The results of the remaining 55 crystals are given in the following.

Since snow crystals have complicated shapes, it is impossible to describe both the size and shape *simultaneously* by a single parameter. Therefore, in presenting the experimental results, we choose to present collection efficiencies as a function of crystal size only. For columnar crystals, we use the length as the size parameter. For planar crystals, the diameters are used. Other parameters such as crystal cross-sectional area, total surface area, Pasternak-Gauvin length (total surface area/perimeter length), and crystal Reynolds

number were tried as the independent variable but no significant difference was observed.

Figures 2–6 show the measured collection efficiencies versus crystal size. The data points show a fair amount of scatter. This may be due to many factors. First of all, the shapes of ice crystals in one category are actually not the same. For example, of all crystals categorized as broad-branched, many have branches larger than the rest. The shapes of branches are also different. Similarly, crystals in the category of hexagonal plates may not be ideal hexagons. Some needles and columns have one end larger than the other. Some stellar crystals have branches broken off. With these nonideal size-shape situations, it is not surprising to see some scatter, moreover, the aerosol chamber was exposed to the environmental air which might have caused a temperature gradient with temperature lower near the wall than in the center of the chamber. This could cause some nonuniformity in the aerosol concentration.

Despite the scatter, the results show clear trends. For a fixed crystal habit, the collection efficiency decreases with increasing collector size. In order to understand this behavior in a clearer way, it is best to recall the definition of the collection efficiency for nonspherical collectors (Wang 1983):

$$E = K/K^* \quad (2)$$

where  $K$  is the collection kernel and  $K^*$  the geometrical collection kernel; i.e., the volume swept out by the collector per unit time. For a fixed collector shape,  $K^*$  increases with increasing size. In general,  $K$  also increases with size. The decreasing trend of  $E$  indicates that the increase of  $K$  with size is less than that of  $K^*$ . A detailed explanation of this fact can be provided by the mathematical model of Miller and Wang (1989) and Martin et al. (1980, 1981). A qualitative expla-

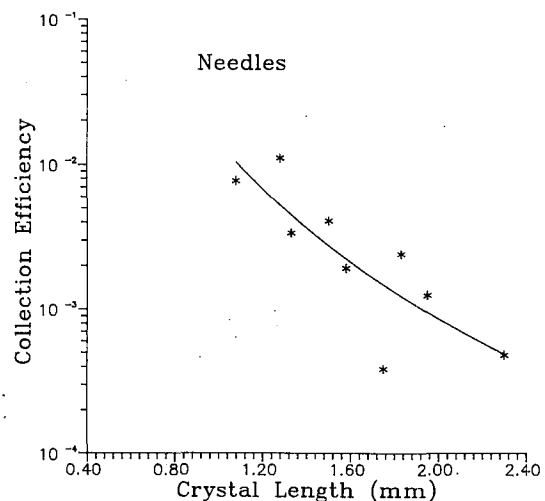


FIG. 2. Collection efficiency vs crystal length for needles.

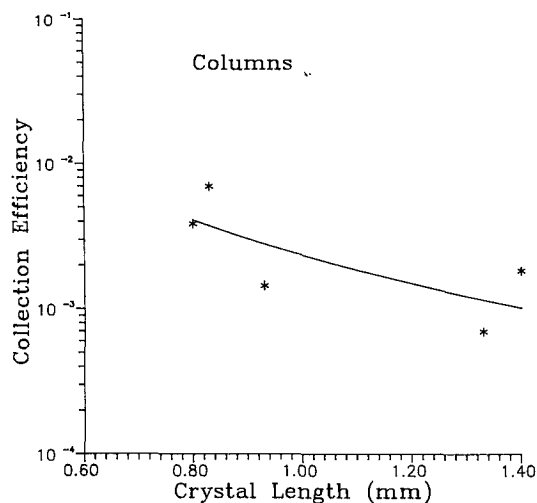


FIG. 3. Collection efficiency vs crystal length for columns.

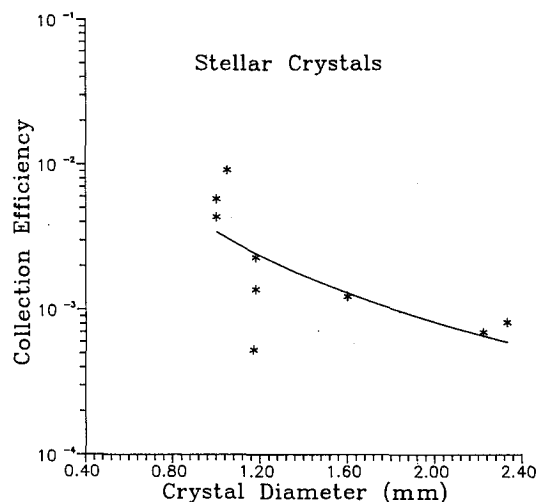


FIG. 5. Collection efficiency vs crystal diameter for stellar crystals.

nation is given as follows. For the particle size ( $r \sim 0.75 \mu\text{m}$ ) considered here, the most important collection mechanism is the inertial impaction which depends on the relative strength of the inertial force of the particle and the hydrodynamic drag force created by the falling motion of the crystal. Since the particle size is fixed in the present study, the decrease of collection efficiency with increasing crystal size must be due to the increasing drag force. Larger crystals of the same type have higher fall speed. But the increase in fall speed is not enough to increase the induced inertia (which scales with fall speed divided by size). Thus the effect of particle inertia relative to particle drag decreases, causing the particle to follow the streamlines more closely. Consequently, the particle is less likely

to collide with the crystal and to decrease in the collection efficiency results.

The foregoing discussion can be further clarified by looking at the dimensionless equation of motion of the particle:

$$d\mathbf{V} = \mathbf{z}/\text{Fr} - (\Delta v/\mathbf{V})/\text{Sk} \quad (3)$$

where  $\mathbf{z}$  is the vertical unit vector,  $\mathbf{V}$  the vector particle velocity, and  $\Delta v = \mathbf{V} - \mathbf{U}$  the instantaneous particle velocity relative to the local air velocity  $\mathbf{U}$ ;  $\text{Fr}$  and  $\text{Sk}$  are the Froude number and Stokes number and are defined by

$$\text{Fr} = U_\infty^2/gD \quad (4)$$

$$\text{Sk} = \rho_p d_p^2 U_\infty / 18\mu D \quad (5)$$

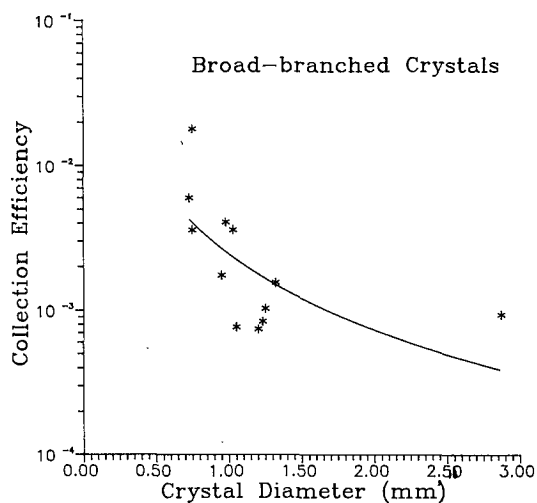


FIG. 4. Collection efficiency vs crystal diameter for broad-branched crystals.

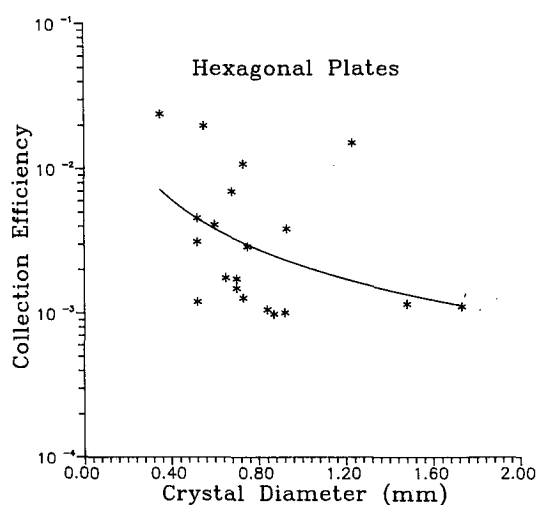


FIG. 6. Collection efficiency vs crystal diameter for hexagonal plates.

where  $U_\infty$  and  $D$  are the fall speed and the diameter of the crystal,  $g$  the gravitational acceleration,  $\mu$  the dynamic air viscosity, and  $\rho_p$  and  $d_p$  the density and diameter of the aerosol particle. For constant particle size and environmental conditions,  $Sk$  is proportional to the ratio of the crystal terminal velocity to its dimension; i.e.,  $Sk \propto U_\infty/D$ . Since fall speeds generally increase with the crystal size as  $U \propto D^n$ , where  $n < 1$ , then  $Sk$  decreases with increasing crystal size. For capture of particle on the upstream side of the crystal, the collision efficiency decreases monotonically with  $Sk$ .

The Froude number scales particle inertia relative to gravity, and helps explain changes in vertical forces that affect front and rear capture. For example, the increasing influence of gravity (decreasing  $Fr$ ) may cause collision efficiencies to decrease on the upstream side of the collector. Changes in  $Fr$  depend on whether  $n > 0.5$  or  $n < 0.5$ ; since  $Fr \propto U_\infty^{2n}/D$ .

For these experiments a typical value of  $Sk \sim 0.02$  suggests that the particle inertia is small compared to drag, whereas a typical value of  $Fr \sim 10$  indicates that particle inertia is large compared to gravity. Even though drag dominates the equation of motion, so that particles tend to follow streamlines, inertia and gravity can still be important in the low collision efficiency problem. It is only because of such forces (and geometrical interception) that the efficiency is nonzero.

It is convenient to fit the experimental data by some empirical relations. The following power equations represent the best fits for the data points in Figs. 2–6 ( $E$  = efficiency,  $L$  = crystal length in mm,  $D$  = crystal diameter in mm):

$$E = 1.42 \times 10^{-2} (L)^{-4.05}, \quad 1.0 < L < 2.3 \text{ mm (Needles)} \quad (6)$$

$$E = 2.35 \times 10^{-3} (L)^{-2.49}, \quad 0.8 < L < 1.4 \text{ mm (Columns)} \quad (7)$$

$$E = 2.45 \times 10^{-3} (D)^{-1.75}, \quad 0.7 < D < 3.0 \text{ mm (Broad-branched)} \quad (8)$$

$$E = 3.41 \times 10^{-3} (D)^{-2.07}, \quad 1.0 < D < 2.4 \text{ mm (Stellar)} \quad (9)$$

$$E = 2.12 \times 10^{-3} (D)^{-1.17}, \quad 0.3 < D < 1.8 \text{ mm (Plates)}. \quad (10)$$

Sometimes it is desired to categorize the crystal habits into two general categories, namely, columnar and planar ice crystals. Figures 7 and 8 show the collection efficiencies for these two broad categories. The columnar category consists of needles and columns, while the planar category consists of broad-branched, stellar, and hexagonal plate crystals. The decreasing trend of  $E$  with increasing crystal size is still preserved. The empirical fits for the data points in Figs. 7 and 8 are

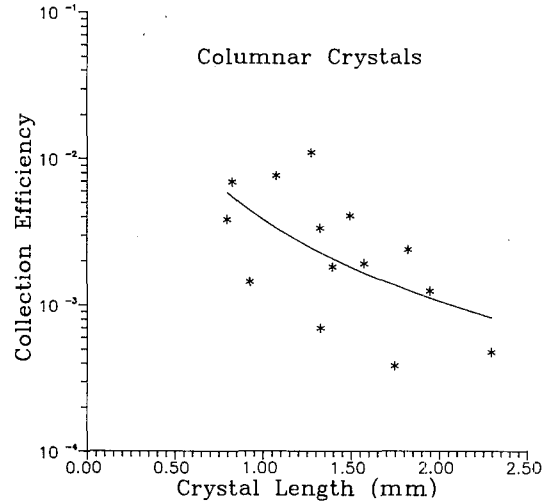


FIG. 7. Collection efficiency vs crystal length for columnar crystals including needles and columns.

$$E = 3.87 \times 10^{-3} (L)^{-1.86}, \quad 0.8 < L < 2.3 \text{ mm (columnar)} \quad (11)$$

$$E = 2.27 \times 10^{-3} (D)^{-1.28}, \quad 0.3 < D < 3.0 \text{ mm (planar)}. \quad (12)$$

Note that since all the above relations are empirical fits, the applicable collector size range must be observed when using them. Efficiencies generated by them outside the specified size range may not be reliable.

The typical values of collection efficiency in Figs. 2–8 are between  $10^{-2}$  and  $10^{-3}$ . These values are characteristics of the collection efficiency in the atmosphere for collectors of a few millimeters in size scavenging submicron size aerosol particles. Such  $E$  values are also typical for drop-particle scavenging (Wang and Pruppacher 1977).

The effect of aerosol charge on the collection efficiency was also investigated. Since snow crystals in the present study had negligible charge, the only effective electrostatic force would be the image force due to the charges on the aerosol particles. The results obtained with electrically charged aerosol particles (also included in Figs. 2–6) do not show significant difference from the uncharged aerosol. This indicated that the image force did not play an important role in the scavenging process. This is consistent with the finding of Wang and Pruppacher (1980) who concluded that image force is unimportant in the scavenging of aerosol particles by water drops in the presence of an external electric field.

Note that the insignificance of the electric effect in the present study is due to the negligible charges on the snow crystals. In other situations where snow crystals may carry substantial amount of charges, the electric effect may become important. We have inadequate knowledge of the electric charges on ice crystals in

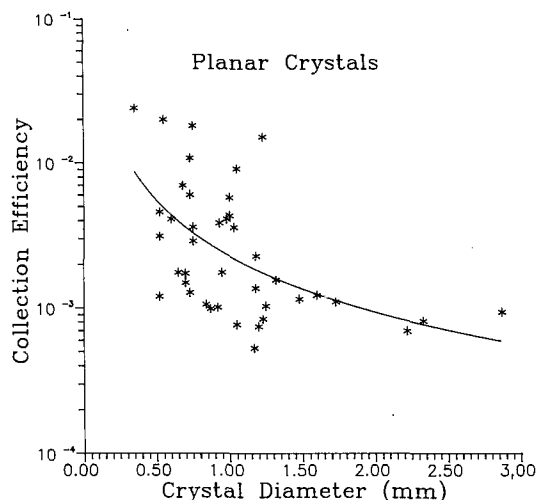


FIG. 8. Collection efficiency vs crystal dimension for planar crystals including broad-branched, stellar, and hexagonal pale crystals.

thunderclouds and snowstorms. If these crystals are significantly charged, the electric effect may be expected to play an important role in the scavenging of particles in these clouds.

#### 4. Conclusions and outlook

In the foregoing discussion, we have presented experimental results of snow crystals scavenging aerosol particles of fixed size. It is shown that the collection efficiency decreases with increasing crystal size. This can be explained by the dominance of drag force over the particle inertia. The model results in the accompanying paper show a general agreement in the order of magnitude and trend with the experimental data.

Some improvements in the future are suggested here. It is felt that more experiments should be conducted to collect more data to minimize the scatter. It is also desirable to vary the size of aerosol particles, especially the smaller size end, so that other effects such as Brownian motion and phoretic effect can be studied. The case of charged snow crystals scavenging charged aerosol particles should be studied. If possible, the thickness and velocity of the snow crystals should be measured. Finally, the present study is limited to snow crystals of simple shapes and a few millimeters in size. The results are applicable to the scavenging of particles by light snow and ice crystals in clouds. The important case of scavenging by aggregates which dominate in medium and heavy snow should be investigated in the future.

Presentation of experimental data of nonspherical particles continues to be a problem. More research will be necessary to find a suitable method. Recently, Wang (1982), Wang and Denzer (1983), and Wang (1987)

have developed some simple mathematical expressions to characterize simultaneously the shape and size of nonspherical particles. It may be possible to develop new methods of presentation based on these formulas.

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#### REFERENCES

- Bauer, C. F., and R. L. Pitter, 1982: Investigations of rimming and snowflake electrification in stratiform-clouds. *Prep. Cloud Phys. Conf.*, Chicago, 568–569.
- Dalle Valle, J. M., C. Orr and B. L. Hinkle, 1954: A new method for the measurement of aerosol electrification. *Brit. J. Appl. Phys., Suppl.*, **3**, 5198–5206.
- Grover, S. N., H. R. Pruppacher and A. E. Hamielec, 1977: A numerical determination of the efficiency with which aerosol particles collide with spherical water drops in air due to hydrodynamic, phoretic, and electrical forces. *J. Atmos. Sci.*, **34**, 1655–1663.
- Knutson, E. O., S. K. Sood and J. D. Stockham, 1976: Aerosol collection by snow and ice crystals. *Atmos. Environ.*, **10**, 395–402.
- Magono, C., T. Endoh, T. Harimaya and S. Kubota, 1974: A measurement of scavenging effect of falling snow crystals on the aerosol concentration. *J. Meteor. Soc. Japan*, **52**, 407–416.
- Martin, J. J., P. K. Wang and H. R. Pruppacher, 1980: A theoretical determination of the efficiency with which aerosol particles are collected by simple ice crystal plates. *J. Atmos. Sci.*, **37**, 1628–1638.
- Miller, N. L., and P. K. Wang, 1989: A theoretical determination of the efficiency with which aerosol particles are collected by falling columnar ice crystals. *J. Atmos. Sci.*, **46**.
- Murakami, M., K. Kikuchi and C. Magono, 1985a: Experiments on aerosol scavenging by natural snow crystals. Part I: Collection efficiency of uncharged snow crystals for micron and submicron particles. *J. Meteor. Soc. Japan*, Ser. II, **63**, 119–129.
- , —, and —, 1985b: Experiments on aerosol scavenging by natural snow crystals. Part II: Attachment rate of 0.1  $\mu\text{m}$  dia. particles to stationary snow crystals. *J. Meteor. Soc. Japan*, Ser. II, **63**, 130–135.
- Starr, J. R., and B. J. Mason, 1966: The capture of airborne particles by water drops and simulated snow crystals. *Quart. J. Roy. Meteor. Soc.*, **92**, 490–499.
- Wang, P. K., 1982: Mathematical description of the shape of conical hydrometeors. *J. Atmos. Sci.*, **39**, 2615–2622.
- , 1987: Two-dimensional shape and size characterization of polygonally symmetric particles. *J. Colloid Interface Sci.*, **117**, 271–281.
- , and H. R. Pruppacher, 1977: An experimental determination of the efficiency with which aerosol particles are collected by water drops in subsaturated air. *J. Atmos. Sci.*, **34**, 1664–1669.
- , and —, 1980: The effect of an external electric field on the scavenging of aerosol particles by cloud and small rain drops. *J. Colloid Interface Sci.*, **75**, 286–297.
- , and S. M. Denzer, 1983: Mathematical description of the shape of plane hexagonal snow crystals. *J. Atmos. Sci.*, **40**, 1024–1028.
- , S. N. Grover and H. R. Pruppacher, 1978: On the effect of electric charges on the scavenging of aerosol particles by cloud and small raindrops. *J. Atmos. Sci.*, **35**, 1735–1743.