Experimental Investigations of Ice in Supercooled Clouds. Part II:
Scavenging of an Insoluble Aerosol

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ABSTRACT

An experimental study of aerosol scavenging by ice growing in supercooled clouds was conducted with a continuous flow cloud chamber. Techniques for detecting insoluble (latex) submicron particles in individual ice crystals were developed. The effects of microphysical parameters on the scavenging process were examined quantitatively. Measurements of the aerosol scavenging rates were documented as functions of cloud temperature, liquid water content, and the diameters (0.109 μm and 0.551 μm) of the nearly monodisperse aerosol particles. Scavenging data were acquired at temperatures of −6°, −8°, −11.5°, and −14°C. The liquid water contents of the supercooled clouds were varied from −0.3 to 6 g m⁻³, while the maximum dimensions of the ice crystals ranged from about 50 to 300 μm.

The scavenging data agree with some previously published theoretical and experimental results and expand the empirical database available for understanding the mechanisms of scavenging. It was found that the presence of liquid water reduced the aerosol removal rates, particularly for crystals growing in the habit transition region near −8°C. It is hypothesized that the retardation effect is due to enhancement of the thermophoretic forces arising from more rapid vapor deposition and latent heat release at higher liquid water contents. The scavenging efficiency at a given liquid water content, however, was not found to depend significantly on the growth habit of the ice crystal. The data, particularly regarding the dependence of the scavenging rates on liquid water content, appear to resolve an important conflict in the literature regarding the relative roles of thermophoresis and diffusiophoresis in the scavenging of submicron particles by ice crystals growing in supercooled clouds.

1. Introduction

Detailed knowledge of the microphysical behavior of mixed-phase clouds is important for understanding not only ice evolution and precipitation development, but also the atmospheric budgets of trace chemicals and aerosol particles. Since precipitation has been found to play important roles in the removal of atmospheric aerosol particles, we expect cloud–aerosol interactions to affect the spatial and spectral distributions of pollutants in the atmosphere, hence air quality and regional climates. Although clouds are generally recognized for their ability to cleanse the atmosphere, many of the mechanisms in the overall scavenging process remain uncertain, particularly those involving ice.

The methods and approaches for investigating the problem of aerosol scavenging by ice particles are analogous to those for studying the scavenging of particles by water drops. A good summary of the concepts involved and the results of earlier studies has been given by Pruppacher and Klett (1978, section 12.7). Here, we focus on ice as the collecting body and distinguish below-cloud from in-cloud scavenging. Below-cloud scavenging of aerosol particles, during which the ice crystals experience subsaturated conditions, has been studied experimentally by a number of authors (e.g., Sood and Jackson 1970; Knutson et al. 1976; Murakami et al. 1985a,b,c; Sauter and Wang 1989; Mitra et al. 1990; Andrew and Sanders 1991). Some of these experiments were conducted in a stagnant and aerosol-laden chamber, and natural snow crystals were often used. Scavenging information was acquired by examining the residues of the ice crystals through microscopy or microchemical analysis. The effectiveness of aerosol collection, as expressed by the scavenging efficiency, was determined as a function of the size and habit of the ice crystal, its fall distance, and the characteristics of the aerosol particles and other environmental conditions.

Aerosol scavenging by ice in unsaturated conditions has also been modeled numerically by various authors (Wang and Pruppacher 1980; Martin et al. 1980a,b; Miller and Wang 1989; Zhang and Pitter 1991). Typically, the problem of aerosol scavenging has been treated as a fluid dynamical problem. If the aerosol particles were small enough, less than about 0.1 μm
diameter, so that particle inertia could be ignored, the scavenging was treated as a convective–diffusive process. In some cases, analytic solutions of the convective–diffusive equation were found when the shape of the collector was relatively simple. For larger particles, on the other hand, the flow field around the scavenging crystal was computed first, then the trajectories of the aerosol particles were computed in order to determine if the aerosol particles would intercept the ice crystal surface.

Some consistency has been found between the available experimental data and theoretical results (Martin et al. 1980b; Miller and Wang 1989; Mitra et al. 1990). However, completely satisfactory agreement has not been reached for a number of reasons. One of the reasons is the difference in crystal size and shape. In experimental studies, the ice crystals employed were usually large (greater than 1 mm) and exhibited the complicated shapes arising from natural clouds. In the theoretical studies, often limited by computational resources, only small ice crystals (size much less than 1 mm) with highly simplified shapes were modeled. Therefore, direct and close comparisons are usually not available for most crystal size ranges. Nevertheless, the magnitudes of the collection efficiencies and their trends with the particle or crystal size were found to be reasonably consistent between the findings of the experiments and theory.

Another problem that arises when trying to verifying theoretical concepts is the large scatter that typically exists in the experimental data. Explanations for such scatter, sometimes spanning more than an order of magnitude, have been attempted by several authors, but a satisfactory explanation is still needed. Some cases nevertheless exist in which large differences between theoretical results and experimental data emerge. Andrew and Sanders (1991), for instance, reported that the rates of collection of NaCl particles (about 4 μm in diameter) by small ice crystals were found to be much greater than those determined by the theoretical calculations of Martin et al. (1980b). The difference here may be partially explained by the fact that the study by Andrew and Sanders (1990) considered the NaCl particles to be dry, whereas the particles should perhaps have been treated as haze droplets.

Electric effects on aerosol scavenging by ice have also been explored both theoretically and experimentally (e.g., Martin et al. 1980a; Murakami et al. 1985b). It was found that electric effects can significantly enhance scavenging in electrically active atmospheric clouds. A source of electric charge separation on a growing ice crystal was hypothesized and the potential charge effect on scavenging was explored by Zhang and Pitter (1991).

Some very large efficiencies of aerosol scavenging by ice have been found in some field observations (e.g., Graedel and Franey 1975; Magono et al. 1979). However, such observational data serve mainly as qualitative indicators. The mechanistic interpretations and quantitative comparisons with theory are made difficult due to the lack of control over and complete information about the environmental conditions. For example, the change in aerosol concentration at a fixed location during the initial stage of a snowfall was used by Graedel and Franey (1975) for the calculation of a washout coefficient without an adequate way of separating out the effects of other meteorological processes. In the studies by Magono et al. (1979), morphological analyses of the residues of individual snow crystals with electron microscopes were used to acquire the scavenging information without knowing the relative contributions from the different scavenging mechanisms (e.g., nucleation, riming, and interstitial particle collection). No rigorous justification of the detection method was given, even though the sample processing procedure itself may cause surface morphological changes. Also, the growth histories of the ice crystals (e.g., growth time) were not known well enough for the calculation of scavenging efficiencies.

By contrast with the studies of below-cloud scavenging, relatively few detailed investigations of aerosol scavenging by ice during crystal growth have been reported. Overall, in-cloud scavenging has been found to be a major contributor to the aerosol scavenging process (Murakami et al. 1983), possibly due to riming or the collection of interstitial aerosol particles. The specific mechanisms responsible for in-cloud scavenging are not at all clear, however. Martin et al. (1980b) extended their calculations of aerosol scavenging efficiencies by ice to water-saturated conditions and found the scavenging efficiencies to be less than those in ice-ununsaturated conditions, in accord with the suggestion of Slinn and Hales (1971) that the net phoretic force arising from thermophoresis and diffusio-phoresis weakens the scavenging for growth situations.

An alternative mechanism of aerosol scavenging by ice in supercooled clouds was proposed by Vittori and Prodi (1967) and Vittoni (1973, 1984). According to these authors, the dominating influence that thermophoresis normally exerts on particle collection by growing crystals is suspended during the brief time that supercooled cloud drops pass by a sedimenting ice crystal. In essence, the vapor field is able to respond rapidly to the sudden influx of water vapor from each evaporating cloud drop, but the temperatures of the water droplet and the relatively massive crystal cannot change significantly during the small times available. Any aerosol particles “caught” in the intervening space between the crystal and the droplet during the transient “flushes” of water vapor toward the crystal surface are expected to experience exceptionally strong diffusion-phoretic (Stefan flow) forces toward the crystal and only weak thermophoretic forces away from the collecting body. The experimental findings of Vittori and Prodi (1967) have been offered as evidence for the im-
portance of Stefan flow in the scavenging of aerosol particles in mixed-phase clouds.

From the arguments and analysis of Vittori (1984), we should expect that the scavenging efficiency of ice crystals should be enhanced roughly in proportion to the frequency of the transient drop-passage events. For a given droplet size distribution, then, the enhancement should scale with the liquid water content of the supercooled cloud in which the crystal is growing. Such a dependence of aerosol collection efficiency on liquid water content is not expected to prevail if only the traditional, steady-state phoretic processes are operative during the vapor growth of ice. It is therefore reasonable to expect that a careful set of experimental measurements of aerosol collection rates at widely varying liquid water contents could lead to a resolution of these two opposing viewpoints.

The primary objective of the present study is to quantify the rates of submicron aerosol scavenging by ice in supercooled clouds as functions of the particle diameter, crystal size, liquid water content, and temperature under controlled environmental conditions. The experimental results are intended to be used to evaluate plausible scavenging mechanisms and so resolve the conflict that has existed in the literature for many years. The data are documented in a way that allows them to be applied more or less directly to cloud models for studying aerosol budgets on larger scales. Section 2 describes the experimental methods and procedures. The experimental data are then presented in section 3. The physics of aerosol scavenging is discussed in light of the current scavenging data in section 4. Section 5 summarizes and concludes this research effort.

2. Experimental methods

a. Experimental apparatus and procedures

The current set of experiments was conducted with a continuous-flow cloud chamber, details of which have been given by Song and Lamb (1993; Part I of this series). The cloud chamber allowed the scavenging experiments to be carried out under controlled and well-characterized environmental conditions. As explained in Part I, the components of the supercooled clouds—the liquid drops, the ice crystals, and the aerosol particles—are generated external to the main wind tunnel where crystal growth and aerosol capture took place. The special design of the flow system allowed a population of ice crystals to grow up to about 450 μm in maximum dimension at temperatures in the range of −3° to −20°C.

The experimental data were intended to be compared with past model results and the proposal by Vittori (1973) that transient phenomena may augment scavenging in supercooled clouds. Unfortunately, a detailed theoretical study of aerosol scavenging by ice in supercooled cloud conditions has not been available for direct comparison in this situation, so the experimental data have to be compared with the available model results for scavenging in unsaturated conditions. Thus, the scavenging experiments were carefully designed to bring the laboratory studies as close as possible to the scavenging conditions in the model simulations. Since data are most needed in the “Greenfield” gap region, two aerosol particle diameters, 0.109 μm (“0.1 μm”) and 0.551 μm (“0.5 μm”), were chosen. The sizes of typical ice crystals in the experiments were a few hundred micrometers (see Part I). To compare the scavenging data among different temperatures, the growth duration was fixed at 100 s. However, the crystal size varied with temperature because of the inherent dependence of growth rate on temperature. The concentrations of aerosol particles in the scavenging experiments were typically $6 \times 10^3$ cm$^{-3}$ for the 0.1-μm latex particles and $1.8 \times 10^3$ cm$^{-3}$ for the 0.5-μm latex particles. The concentration of ice crystals was in the range of 100 to 1000 L$^{-1}$. The ice concentration was controlled to be large enough to yield a sufficient number of crystal samples and small enough to avoid growth competition among the ice crystals.

The aerosol particles (0.1 μm and 0.5 μm diameter) used in the experiments described here were produced by atomization (TSI 3076) from an aqueous suspension of latex particles (Seradyn, Inc.). The particle stream was electrically neutralized (TSI 3012) and then blended with the supercooled cloud in the mixer of the cloud chamber (see Fig. 2 of Part I). The total number concentration of aerosol particles in the wind tunnel was monitored by a condensation nucleus counter (TSI 3022).

After their growth and exposure to the aerosol, the ice crystals were collected in a clean environment so as to avoid contamination and possible misinterpretation of the results. During the crystal growth portion of the experiments, a weak flow of humidified argon was introduced into the collection box and allowed to exit at the neck position near the base of the distribution chamber in order to keep cloud from settling into the vicinity of the collector. During actual collection of the ice crystals, the argon flow in the collection box was increased to compensate for the extra flow of cloud air into the neck region (see Part I). The relatively large fall speeds of the ice crystals allowed them to fall through the cloud–argon interface and into the collection box where they could be caught by the ice collector. All small particles (interstitial aerosol particles and cloud droplets) were kept well away from the collector by the updraft of argon into the neck of the chamber.

Any significant electrical charges on the aerosol particles and cloud droplets were eliminated by the use of bipolar neutralizers (TSI 3012 and TSI 3054). Special tests were performed in order to assess the charge on the ice crystals and assure ourselves that the scavenging data apply to near-neutral conditions. For this test, the ice crystals were allowed to fall through a strong, hor-
izontally directed electric field (4 kV/cm). No detectable deviation of the fall path was observed over a fall distance of 10 cm, so we estimate that the charge on the crystals (mass 0.1 μg) was smaller than about 1 fC. Thus, no significant amounts of charges were found on the ice crystals generated in the cloud chamber, and we judge the uncertainty due to charge effects in the current experiments to be small (see Song 1991).

The temperature in the wind tunnel, the liquid water content of the supercooled cloud, and growth time of ice crystals were monitored or measured as described in Part I. The ice crystal shape and size were documented with an optical stereo microscope after they were collected. Figure 1 shows the physical arrangement used for sample collection and documentation.

b. Sample processing

Various techniques have been used in the past to detect the aerosol particles scavenged by individual ice crystals (e.g., Sood and Jackson 1970; Murakami et al. 1985a; Sauter and Wang 1989; Goodman et al. 1989). The techniques can be classified into two major kinds: detection of the residues of individual ice crystals and detection of individual aerosol particles in the replica of an ice crystal. The first technique is tedious and makes it difficult to unambiguously associate a particular residue with the right ice crystal. The second technique is subject to contamination from the replication process. Here, we describe the procedure, a variation of these two basic kinds, used to retrieve the scavenging data in the current research.

After the ice crystals were collected, the ice collector was brought quickly to an adjoining cold environment that was made of an open foam box purged from below with clean, cold air (Fig. 1c). There, the ice collector was dismantled and the sample holder retrieved and covered with a thin piece of glass such that the ice crystals were “sandwiched” for protection while being photographed. The substrate onto which the crystals fell had been precoated with a thin plastic layer (polystyrene). The ice crystals were then melted into drops, by replacing the cover glass with a warmer piece of glass, and rephotographed in order to determine the masses of the ice crystals (Fig. 1b).

Soon after the melt drops formed, they were briefly exposed to the vapor of the plastic solvent, the aerosol particles being well protected by the water from the effects of the solvent. A plastic replica of the drop edge would thus remain on the substrate after the drops evaporated. A morphological analysis by a scanning electron microscope (SEM) readily revealed the aerosol particles on the substrate since the replicas of the drop outlined the boundaries of the region of the substrate belonging to the individual ice crystals. Figure 2 shows photographs of such particles within the boundaries of the drops formed from the melting of the ice crystals. In Fig. 2a the mean diameter of the latex spheres was 0.109 μm, whereas in Fig. 2b the mean diameter of the latex spheres was 0.551 μm. The arcs in the picture are parts of the drop replicas. The sizes of the drop replicas also provide information about the masses of the ice crystals. This technique was found to be very useful for processing samples containing small aerosol particles, especially when such particles might be easily destroyed or obscured during a true crystal replication process.

c. Data analysis

The latex particles contained in the individual ice crystals constitute the primary evidence for the scavenging that took place during the vapor growth of the crystals. The particle counts obtained from the SEM photographs were combined with other data to provide an estimate of the scavenging kernel, the parameter needed in the continuity relationship for calculating the concentration of particles in clouds (Pruppacher and Klett 1978, p. 380). This subsection describes the conceptual basis for estimating the magnitude of the kernel from the experimental data as well as the adjustments made to the experimental procedures to compensate for two naturally occurring experimental problems: particle clustering during atomization and false counting by the condensation nucleus counter due to the presence of nonlatex particles.
Fig. 2. SEM photographs of replicas, of drops from ice crystals, and latex aerosol particles scavenged by the ice crystals. The growth habits were broad-branch plate, growth durations were 100 s, and the liquid water content was 2.0 g m$^{-3}$. (a) Aerosol particles were 0.109-μm latex spheres and aerosol concentration was $6 \times 10^5$ cm$^{-3}$. (b) Aerosol particles were 0.551-μm latex spheres and aerosol concentration was $1.8 \times 10^3$ cm$^{-3}$. 
The total number of latex particles scavenged by each crystal was used, along with the measured aerosol concentration in the cloud chamber and the experimental run time, to determine the scavenging kernel. The kernel $K$ that represents a suitable average of the aerosol scavenging that took place in the cloud chamber under a particular set of conditions was calculated by the following equation:

$$K = \frac{\Delta N_s}{n_o \Delta t}. \quad (1)$$

Here, $\Delta N_s$ is the number of aerosol particles collected by the ice crystal over the experimental time interval $\Delta t$ when the concentration of aerosol particles was $n_o$. However, the microphysical characteristics of the experimental aerosol unfortunately disturb the otherwise straightforward application of Eq. (1) to the determination of the scavenging kernel. The dispersion, or size variation of the aerosol particles, introduces an error because any given particle may be composed of several latex spheres. Such a “cluster,” when caught by the ice crystal, represents a single scavenging event, yet it is counted as the number of latex particles that it contained. In addition, not all of the particles generated by the atomizer necessarily contain latex particles, even though they are counted by the condensation particle counter. Such factors, discussed here in some detail, need to be understood and accounted for.

The number of individual latex spheres (of diameter 0.1 or 0.5 μm) actually found in the residue of an ice crystal is the sum of all the spheres from all particle clusters. This total number $\Delta N_s$ of separately counted latex spheres can therefore be represented mathematically by the summation

$$\Delta N_s = \sum_{j=1}^{\infty} j \Delta N_j, \quad (2)$$

where $\Delta N_j$ is the number of clusters each containing $j$ latex spheres. The number of such clusters collected during the experiment cannot be determined directly, but it can be related mathematically to the size-specific scavenging kernel $K_j$ through the relationship

$$\Delta N_j = K_j P_j n_o \Delta t, \quad (3)$$

where $P_j$ is the probability of finding a cluster containing $j$ spheres within the aerosol population. For a nearly monodisperse distribution of particles, as used in our experiments, there exists a small value $j_e$ (less than 10) beyond which the chances of finding such a large cluster are extremely small. Furthermore, since the diameters of the clusters increase weakly with increasing $j$ (as $j^{1/3}$), the magnitude of $K_j$ varies relatively little with cluster size for all practical purposes (Martin et al. 1980b). We may therefore express the scavenging kernel as

$$K = \frac{\Delta N_s F}{n_o \Delta t}, \quad (4)$$

where

$$F = 1 \left/ \sum_{j=1}^{j_e} j P_j \right. \quad (5)$$

is the correction factor that is needed to compensate for the clustering of spheres that naturally occurs during the aerosol generation. The factor $F$ was obtained empirically from measurements of the size distribution of the aerosol particles collected on Nuclepore filters (0.2-μm pore diameter).

The clustering of latex spheres in the aerosols generated by atomization may be controlled to some extent by varying the dilution of the liquid suspension used in the atomizer. In dilute solutions, the cluster-forming tendency of the latex spheres becomes small, but then the tendency to generate “empty” aerosol particles, those that contain no latex spheres at all, is relatively large. These empty particles invariably contain the residue of the original solution and so appear as valid counts in the condensation nucleus (CN) counter (sensitive to particles greater than about 0.01 μm in diameter). Dilution thus introduces a counting error that operates in the sense of making the true latex particle concentration $n_o$ too large and the scavenging kernel calculated from Eq. (4) too small. We therefore correct for this effect by defining the factor $R = n_o / n_{CN}$ and calculating the kernel from

$$K = \frac{\Delta N_s F}{n_{CN} \Delta t R}. \quad (6a)$$

As with $F$, the correction factor $R$ was determined empirically through filter sampling of the aerosol and simultaneous measurement of the total particle concentration $n_{CN}$ with the CN counter. Data and additional details have been provided by Song (1991).

Equation (6a) simultaneously corrects for the effects of clustering and false counting, both of which depend on the dilution of the aqueous suspension used in the experiments. Fortunately, the dependence of $F$ on the solution dilution ratio opposes that of $R$. In fact, the magnitudes of $F$ and $R$ must become equal to each other at some intermediate dilution ratio. In the current set of experiments, the solution dilution ratio was thus carefully adjusted to force the magnitudes of $F$ and $R$ to be equal ($0.8$). The need for any adjustments in the data themselves was thus circumvented, and the formula

$$K = \frac{\Delta N_s}{n_{CN} \Delta t} \quad (6b)$$

was used operationally to determine the scavenging kernel. The average scavenging efficiency was then calculated from
where \( V_\infty \) is the terminal fall speed and \( S \) is the horizontal cross-sectional area of the ice crystal. An average value for \( S \) was obtained from the photographic records of the ice crystals after collection. Values for \( V_\infty \) were estimated from the literature (Martin et al. 1980b; Miller and Wang 1989). Each data point presented in this paper represents the average of about 20 columnar crystals or 15 planar crystals from any given batch.

3. Experimental results

The effect of the supercooled droplets on the scavenging of submicron particles by ice was determined by taking the scavenging data under similar experimental conditions except for the liquid water content. The updraft velocity in the wind tunnel was set to be about 10 cm s\(^{-1}\) for these runs. Figures 3 to 5 show the scavenging efficiencies plotted as functions of the liquid water content for the growth temperatures of \(-6^\circ\), \(-8^\circ\), and \(-14^\circ\). The growth and scavenging duration in all these cases was the same, 100 s. Most of these data were taken using latex aerosol particles with diameters of 0.1 \( \mu m \), although some data for 0.5 \( \mu m \) diameters are also shown in Fig. 5. The scavenging efficiencies from the theoretical computations by Martin et al. (1980b) for planar-type ice crystals and by Miller and Wang (1989) for columnar-type ice crystals are also plotted in these figures. The current data are plotted as solid symbols on the right-hand side, while the theoretical results are plotted as open symbols on the left-hand side. The vertical axis is the scavenging efficiency, and the horizontal axis is linear in the total ambient water content (vapor and liquid phases only). Leftward of the vertical axis, the abscissa represents the degree of undersaturation with respect to ice. The spacing between the two zeros on the horizontal axis represents the supersaturation with respect to ice due to water saturation. Rightward from the second zero, the horizontal axis represents the liquid water content in the supercooled cloud. A strong decreasing trend of the experimental scavenging efficiency with the liquid water content appears in Fig. 4 for the short columnar ice crystals grown at \(-8^\circ\), whereas there was no apparent dependence of the scavenging efficiency on the liquid water content at the other two temperatures, \(-6^\circ\) (Fig. 3) and \(-14^\circ\) (Fig. 5). The experimental data suggest that the liquid water content has effects on aerosol scavenging under some but not all conditions. At \(-6^\circ\) and \(-14^\circ\), the experimental scavenging efficiencies appear to be similar to the theoretical values extrapolated to water saturation.

The habits of the crystals varied predictably with the growth temperature. For ice crystals grown at \(-6^\circ\) (Fig. 3), the crystal habit was long column. The average crystal length was 90 \( \mu m \) and the average width was 40 \( \mu m \). For ice crystals grown at \(-8^\circ\) (Fig. 4), the habit was short solid column with an average length of 60 \( \mu m \) and an average width of 50 \( \mu m \). For ice crystals grown at \(-14^\circ\) (Fig. 5), the shape was broad-column with an average diameter of 200 \( \mu m \). For comparison with the experimental results, we chose the "diameter" of the ice crystals used by Martin et al. (1980b) to be 225.6 \( \mu m \), and the shape to be an oblate spheroid with an axial ratio of 0.05. This set of results was chosen for the comparison because the diameter and the shape are closest to the plate-type crystals found in the current data at \(-14^\circ\). In Figs. 3 and 4, we chose the columnar crystals represented as prolate spheroids with length 90 \( \mu m \) and width 40 \( \mu m \) that were used by Miller and Wang (1989). This shape and
Fig. 5. Scavenging efficiency as a function of the liquid water content. The left-hand side is the theoretical results from Martin et al. (1980b) for oblate spheroid with 226 μm in diameter and 0.05 in axis ratio; aerosol size is 0.1 μm. The right-hand side is the experimental data; the growth temperature was −14°C; shape is broad-branch plate; mean diameter is 200 μm; the growth time was 100 s. Two different aerosols with sizes 0.1 μm and 0.5 μm were used.

Fig. 6. Scavenging efficiency as a function of crystal diameter. The growth temperature was −14°C; the liquid water content was maintained between 2 and 3 g m⁻³.

Fig. 7. Scavenging kernel as a function of crystal diameter. The growth conditions was the same as those in Fig. 6.

The size were similar to the long columns produced at −6°C, but differed somewhat from the ice crystals produced at −8°C. Nevertheless, these same theoretical results are shown in Fig. 4 for contrast.

Scavenging data were also taken for 0.5-μm latex particles at −14°C, as plotted in Fig. 5, while the other conditions were kept the same as those for the 0.1-μm latex particles. Overall, the scavenging efficiencies for 0.5-μm particles are only slightly larger than those for 0.1-μm particles. In general, the collection of 0.5-μm latex particles is not drastically different from that of 0.1-μm latex particles.

The dependence of the scavenging efficiency on crystal size was examined by performing a series of experiments at one temperature (−14°C) with similar liquid water contents. The scavenging efficiencies are plotted as a function of the mean crystal diameter in Fig. 6, while the corresponding scavenging kernels are plotted in Fig. 7. Each data point again represents an average obtained from about 15 ice crystals in a given growth batch. The variations in the average crystal diameter were produced by varying the growth duration over a range from 60 to 150 s. The shapes of the ice crystals were broad-branch plate. The size of the aerosol particles used was 0.1 μm. The flow rate in the wind tunnel was adjusted from run to run according to the length of the growth time to accommodate the growth of large ice crystals. The liquid water content was kept between about 2 and 3 g m⁻³ in all cases. The theoretical results from Martin et al. (1980b) are also plotted for comparison. In the theoretical calculations, the diameter of the aerosol particles was 0.1 μm, the shape of the simulated ice crystal was oblate spheroid with an axial ratio of 0.05, and scavenging was for a water-saturated environment (droplet effects were not considered). Both the experimental and theoretical scavenging efficiencies show decreasing trends with the crystal diameter, although the measured trend is slightly stronger. Both the experimental and theoretical scavenging kernels show increasing trends with the crystal diameter.

In order to present an overall dependence of aerosol scavenging on the growth habit of the ice crystals, data were taken as functions of the growth temperature. In
each case, the growth time was 100 s and the liquid water content was between 1 and 2 g m\(^{-3}\). Again, the particles collected were the 0.109-\(\mu\)m latex spheres. Since the growth rates and habits of the ice crystals are highly dependent on the growth temperature, the differences among the scavenging data for different temperatures could be due to either crystal shape or size. The measured scavenging efficiencies are plotted in Fig. 8 and vary surprisingly little with the growth habit under these conditions. The scavenging efficiency appears to be largest at \(-11.5^\circ\text{C}\) and smallest at \(-14^\circ\text{C}\).

4. Discussion

The current data allow one to examine plausible mechanisms of aerosol scavenging by ice crystals grown in supercooled clouds. In particular, the data taken over a broad range of liquid water contents allow one to examine the hypothesis of Vittori (1973) regarding the effects of diffusiophoresis. In Vittori's hypothesis, scavenging should be enhanced by the "vapor flush" associated with the passage of each cloud droplet. If this effect is active in scavenging, the efficiency should increase with the liquid water content, a surrogate measure of drop number concentration (since the size distribution was relatively constant; see Part I) and thus the frequency of drop-passage events. The experimentally determined scavenging efficiency was, however, never found to increase with the liquid water content. From this evidence alone, it can be concluded that the scavenging mechanism proposed by Vittori was ineffective in scavenging and that other mechanisms apparently dominate the mechanics of collection. Comparisons between the present scavenging data and past results may help one to identify the true scavenging mechanisms. Most prior experimental data do not encompass in-cloud scavenging in a way that permits a proper assessment of aerosol scavenging by ice growing in supercooled clouds, so we emphasize a close comparison between the current data and previous model results.

a. Comparison with model results

Our experimental data and the theoretical scavenging efficiencies were found to be of similar magnitudes near liquid water saturation in the two cases where the best comparisons can be made (long columns, Fig. 3; and plates, Fig. 5). In the other case considered here (short columns, Fig. 4), a comparable theoretical model was not available and the experimental scavenging efficiencies were found to decrease substantially as the liquid water content increased.

These characteristics of the data are supported by some theoretical results. As indicated by prior theoretical calculations (e.g., Slinn and Hales 1971; Martin et al. 1980b), thermophoresis dominates diffusiophoresis for particles with radii less than about 2 \(\mu\)m. Thermophoretic forces in effect "push" aerosol particles toward an ice crystal during evaporation and away from them during vapor deposition. As a result, the net phoretic force augments scavenging under subsaturated (below cloud) conditions and weakens the scavenging under supersaturated (in cloud) conditions. Such is the basis for the downward trend with increasing moisture content shown by the theoretical results plotted in Figs. 3 to 5. In our experiments, the crystal growth was found to be enhanced by the presence of cloud droplets (Song and Lamb 1993), particularly in the case of compact, solid columns grown at \(-8^\circ\text{C}\). Such growth enhancement should be accompanied by the enhanced release of latent heat. Therefore, the effect of thermophoresis should increase with the liquid water content, and the scavenging efficiency should decrease. Such a decreasing trend in scavenging efficiencies with the liquid water content was indeed found for short solid column crystals (Fig. 4). At the same time, however, any change of crystal size or habit due to the liquid water may also have had an effect on the scavenging, so we present Fig. 9, a plot of the dependence of the scavenging kernel on the liquid water content using the same set of data as presented in Fig. 4. We still find a strong decreasing trend with the liquid water content even though the crystal size enhancement tends to increase the scavenging kernel. Of particular interest, the magnitude of the growth enhancement due to the increasing liquid water content was found to be largest for those crystals (short solid columns) that exhibited the most pronounced decrease in scavenging efficiency. This decreasing trend in both scavenging efficiency and kernel could well be due to the enhanced latent heat released by the enhanced growth and the development of larger temperature gradients near the crystal surface.

The scavenging efficiency and kernel were each plotted as functions of the crystal diameter in Figs. 6 and 7 for broad-branch plates. A decreasing trend of the scavenging efficiency was found in Fig. 6, whereas
an increasing trend of the scavenging kernel was found in Fig. 7. This set of results was found to be entirely consistent with the theoretical results of Martin et al. (1980b) for ice crystals represented as thin oblate spheroids at water saturation, which were also plotted in Figs. 6 and 7. The slightly stronger decreasing trend found in the experimental scavenging efficiencies than in the computed cases could be attributed to several factors, including differences in crystal shape and in the environmental conditions. The trend of decreasing scavenging efficiency with the size of the ice crystal was explained by Martin et al. (1980b) as the effect of the deflecting flow upstream of an ice crystal. For crystals up to 400 μm in crystal size, the increasing effect of the deflecting flow reduces the scavenging efficiency as the diameter of the ice crystal increases. The consistency between the theoretical and experimental scavenging data on the size of ice crystals indicates that the explanation given by Martin et al. (1980b) is reasonably good.

The dependence of the scavenging efficiency on temperature (or growth habit) appears to be quite small. The scavenging efficiency at −11.5°C is only slightly larger than at the other temperatures. The current data support the reports by Sood and Jackson (1970) and Knutson et al. (1976) that the habits of ice crystals do not make a significant difference in the scavenging efficiency.

Scavenging data for 0.5-μm aerosol particles were also taken for the ice crystals grown at −14°C over a period of 100 s. In Fig. 5, the scavenging data for 0.5-μm aerosol particle were presented. The environmental conditions for these experiments were the same as for the 0.1-μm particles. The scavenging efficiencies at different liquid water contents were found to be slightly larger, which is consistent with the results reported by Murakami et al. (1985a). The model results by Martin et al. (1980b), as well as by Wang (1992), show a complicated dependence of scavenging efficiency on the particle size for plates of certain sizes. The right combination of the hydrodynamic flow and particle inertia apparently produces a minimum in scavenging effectiveness. Our experimental data are not able to show such subtle scavenging behavior.

b. An examination of scavenging mechanisms

Even though the theoretical results are not in exact quantitative agreement with the present experimental data, the overall behavior of aerosol scavenging by ice in supercooled clouds has been found to be consistent with the results of theory based on steady-state phenomena like those considered originally by Slinn and Hales (1971). Therefore, it is likely that the scavenging mechanisms operating in supercooled clouds are essentially the same as those operating in unsaturated conditions. In both subsaturated and supersaturated environments, thermophoresis dominates diffusiophoresis, so the net phoretic force weakens the Brownian/inertial scavenging of submicron particles as the moisture supply increases. The general agreement between the experimental data and the traditional computational results suggests that the model physics applied by Martin et al. (1980b) and Miller and Wang (1989), for example, are realistic even though real crystal shapes and the detailed physical processes may be much more complicated.

The current scavenging data suggest that the supercooled droplets themselves do not enhance scavenging. In particular, the enhanced diffusiophoretic effect proposed by Vittori (1973) to arise from the transient "flushes" of vapor from passing droplets does not appear to produce any measurable increase in the scavenging efficiency. In order to understand how the "vapor flush" effect may augment crystal growth, but not aerosol scavenging, the physics of the process needs to be considered in some detail. According to the analysis by Murakami et al. (1985b), the settling of ice crystals increases the scavenging rates by up to two orders of magnitude, which is consistent with the idea that scavenging depends on a continuous flux of aerosol particles around the falling ice crystals. The capture of an aerosol particle by an ice crystal due to the "vapor flush" effect can happen only when a droplet is physically near the scavenging ice crystal and an aerosol particle is in the intervening gap between the droplet and the ice crystal. However, during the short transit time that a droplet passes by an ice crystal, the chance of finding even one aerosol particle near the droplet and subject to the "vapor flush" effect may be vanishingly small.

This reasoning can be treated semi-quantitatively in order to estimate the contribution of the "vapor flush" effect to the scavenging kernel and efficiency. The number of aerosol particles so collected by the ice crystal in a given time interval can be estimated from the product of the number of droplets that pass by the ice crystal and the average number of particles in the gap.
When a droplet passes by an ice crystal, the rectangular volume affected by the “vapor flush” can be approximated as \( V_f = 2\pi R^2 \delta \). Here, \( \delta \) is the distance between the droplet and the crystal surface at the moment of closest approach, \( r \) is the droplet radius, and \( R \) is the crystal radius. With the liberal assumption that every particle that happens to be in the affected volume is scavenged, the number of particles collected due to each droplet passage is \( N_c = V_f n_d \). The total number of droplets passing by the ice crystal in time \( t \) can be calculated by \( N_d = n_d SV_{es} t \). Here, \( S \) is the cross-sectional area of the crystal, \( V_{es} \) is the terminal velocity of the crystal, and \( n_d \) is the droplet concentration. Thus, the total number of particles contributed by the “vapor flush” effect is \( \Delta N = N_t \times N_d \), or

\[
\Delta N = 2\pi \delta SV_{es} n_d n_d.
\]

The scavenging efficiency can then be estimated by dividing \( \Delta N \) by \( SV_{es} n_d \), the product of the geometric sweepout volume and the aerosol concentration [Eqs. (1) and (7)]. That is,

\[
E = 2\pi \delta n_d.
\]

If we use the assumption that the separation distance \( \delta \) is the same as the droplet radius \( r \) and apply the typical values of \( r = 5 \mu m, R = 50 \mu m, \) and \( n_d = 1 \times 10^3 \) cm\(^{-3} \), the scavenging efficiency is found to be \( 3 \times 10^{-6} \), a value more than two orders of magnitude smaller than any measured in our experiments. Even if the drop–crystal separation distance \( \delta \) is expanded to be comparable with the crystal dimensions, we fall far short of the measured efficiencies. Therefore, under all reasonable conditions, the contribution of the so-called vapor flush effect to the aerosol collection is negligible. In essence, the diffusiophoretic mechanism of Vittori (1973) fails because the probability that an aerosol particle is present in the right place during the short time that the individual cloud drops are close to the crystal is exceedingly small.

Nevertheless, the presence of the cloud droplets does contribute to other effects. As discussed before, the continual passage of supercooled droplets by the ice crystals may enhance the depositional growth of compact crystals and eventually lead to riming. Scavenging is likely to be affected indirectly through changes in the sizes or habits of the ice crystals and through enhancement of the thermophoretic effect by release of additional latent heat. Such qualitative arguments should of course be substantiated through detailed numerical models [e.g., those of Martin et al. (1980b) or Miller and Wang (1989)].

Even though a direct comparison between current and past experimental data is not possible, some similarity and consistency can be found. The analyses from most experimental data for large natural snow crystals tend to suggest that simple interception is the dominant scavenging mechanism (Knutson et al. 1976; Murakami et al. 1985; Mitra et al. 1990). A general consistency was found by Martin et al. (1980b) between their theoretical data for small ice crystals and the experimental data from Knutson et al. (1976) for large ice crystals. A general consistency between theoretical results and experimental data under similar scavenging conditions was also reported by Miller and Wang (1989). Therefore, the current experimental data and the scavenging data for natural snow crystals are in agreement in the sense that they are mutually consistent with independent theoretical computations. The current findings furthermore seem to support the results reported by Knutson et al. (1976) that there is no significant dependence of scavenging efficiency on the growth habit.

c. Scatter in the scavenging data

Large scatter, often order magnitudes of variation, appears to be typical of ice scavenging data. Large scatter was also found among individual ice crystals in the current data. For compact crystals, the standard error was found to be as large as the mean, and, for broad-branch plates, the standard error is even larger. Explanations for the large scatter have been offered by several authors, including Murakami et al. (1985a,b,c), Knutson et al. (1976), Sauter and Wang (1989), and Mitra et al. (1990). In their explanations, scatter was attributed to the variations in the sizes and shapes of the ice crystals. However, the dependence of scavenging on habit and size does not appear to be large enough to explain the scatter. In fact, the results reported by Knutson et al. (1976) and Sood and Jackson (1970) indicate that the shape had no statistically significant effect on their scavenging efficiencies. The size dependence predicted by Miller and Wang (1989) also cannot explain the scatter in the scavenging data reported by Sauter and Wang (1989). In the experimental studies by Mitra et al. (1990), some large snow aggregates were found to be effective scavengers, and a “filtering” effect was attributed to the effective scavenging. However, in most past experiments, ice crystals were selected carefully before they were used. Large aggregates should be singled out by this selection process.

Another possible contribution to relatively large scavenging efficiencies and the large scatter in such data is the tumbling motion of ice crystals. There might be several modes of tumbling behavior. As our laboratory-grown ice crystals (growth temperature −14°C) were allowed to fall into the collection box of the cloud chamber, we have often observed that the crystals spin as if they were “pinwheels.” Under the stereo optical microscope, some dendritic ice crystals are seen to have two hexagonal structures grown together along slightly different crystallographic directions. Such three-dimensional structures could readily cause such horizontally self-rotating motion. Tumbling behavior in general may differ very much from one ice particle to another even though their sizes and growth habits are
similar. This argument is consistent with the experimental data in the way that the scatter is dependent on the growth habit. Much less scatter appears in the data obtained from columns than those of broad-branch plates, for instance. Such tumbling motion has significant implications in several areas of interactions between ice crystals and other kinds of particles: aerosol scavenging, riming, and aggregation. Therefore, it deserves further investigation in the future.

There are other plausible explanations for the large scatter in scavenging data, including the methods of data acquisition and processing. The data provided by Wang and Pruppacher (1977) for drop scavenging show very small error bars. Quite small variations are also presented in the data reported by Leong et al. (1982) for a study of scavenging by drops. Indeed, scavenging by liquid drops should be more predictable and reproducible than the scavenging by ice because of the simpler geometry and hydrodynamical interactions involved. However, one should notice that in the data provided by Wang and Pruppacher (1977) and Leong et al. (1982), each data point was derived from the accumulation of large numbers of drops. The rationale to do so may be that the technique for detecting scavenged aerosol particles required such, and that the scavenging for these drops was assumed to be the same because these drops were made to be about the same. It is not at all impossible that the scavenging by these individual drops also differed significantly from each other, although the overall result might not be sensitive to such differences because of the averaging done. It is also possible that ice crystals have often been treated individually because of the uniqueness of each one and because they are more readily handled by techniques such as plastic replication. It may be taken for granted that their scavenging properties should be individually documented. This could result in the large scatter presented in some ice scavenging data. In the current scavenging data, motivated by these arguments, each data point was derived as an average from a population of ice crystals grown in the same growth batch. Compared to the typical large scatter in the past scavenging data, the scatter in the current scavenging data is therefore small. We thus recommend that some method of averaging a population of ice crystals be employed in future experimental investigations.

5. Summary and conclusions

A laboratory study of aerosol scavenging by ice in supercooled clouds has been conducted. Special techniques were developed for detecting the aerosol contents of individual ice crystals, although it was found helpful to average the data from many crystals. The continuous-flow cloud chamber allowed crystal growth and scavenging to occur under realistic and controlled conditions. Scavenging data were documented as functions of temperature, the liquid water content, and the sizes of the aerosol particles and ice crystals. Scavenging was found to be similar for particles of 0.1-μm and 0.5-μm diameters. The trends and magnitudes of the scavenging efficiency and kernel as a function of diameter were found to be similar to some previous computations, and the dependence of the scavenging efficiency on temperature was found to be small. The effects of the liquid water content on scavenging were found to be most pronounced for ice crystals with the habit of short solid columns grown at −8°C. In fact, scavenging was found to be weakened by the presence of the liquid water content in the same situations in which the crystal growth was appreciably enhanced. Otherwise, the effects of the liquid water content on scavenging could lead either to slight enhancement, due to an increase in the crystal size, or to a reduction due to enhanced thermophoresis.

Through comparisons with some of the available theoretical results, the dependence of the scavenging rate on various environmental parameters, including the sizes of the ice crystals and the aerosol particles, as well as the liquid water content, was found to be generally consistent with the traditional models based on steady-state fluxes (small particles) and trajectories (large particles). Scavenging apparently results from combining the effects of particle inertia and interception, as indicated by Martin et al. (1980b), while thermophoresis plays a role in reducing the scavenging efficiency under growth conditions.

The experimental data provide indirect, albeit strong, evidence for the effects of phoretic forces in scavenging. The “vapor flush” mechanism proposed by Vittori (1973) was found not to be important for aerosol scavenging under atmospheric conditions, although this process may be responsible for the enhanced growth of rapidly falling compact crystals. In such cases, the main effect of the liquid water on the scavenging could be a general weakening of the scavenging due to the enhanced thermophoresis set up by augmented growth and surface heating. However, the liquid water content may also indirectly enhance the scavenging kernel through the increase in the crystal size.

These new understandings about the aerosol scavenging by ice could be applied to other scavenging conditions. For example, in the case of riming, the scavenging of interstitial aerosol particles may be less than in nonriming situations due to the effect of the surface warming. It will be helpful to improve our understanding of aerosol scavenging by ice in supercooled clouds by studying the processes systematically in detailed numerical models, such as those reported by Martin et al. (1980b) and Miller and Wang (1989). In order to assess the overall effect of the scavenging process under a range of atmospheric conditions, it is recommended that the scavenging data be utilized in cloud models with detailed microphysics.

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REFERENCES


——, ——, and ——, 1980b: A theoretical determination of the efficiency with which aerosol particles are collected by simple ice crystal plates. J. Atmos. Sci., 37, 1628–1638.


