6.5 MEASUREMENTS OF THE DEPOSITION COEFFICIENT FOR SMALL CIRRUS-LIKE ICE CRYSTALS

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1. INTRODUCTION

The composition and evolution of cirrus clouds in the upper troposphere has been the subject of many recent in-situ, remote sensing, and modeling studies. Such research has greatly advanced the understanding of cirrus dynamics and microphysics, but important gaps still remain. One uncertainty pertains to how efficiently excess vapor is taken up by growing cirrus particles. A measure of this incorporation efficiency is given by the so-called “deposition coefficient”, or mass accommodation coefficient α_m. This coefficient, defined as a ratio of the number of molecules incorporated into an ice crystal lattice to the total number of impinging molecules, reflects the still-uncertain mechanisms that act at the crystal surface to preclude the successful incorporation of some molecules. It is the relative magnitudes of the deposition coefficients over the basal and prism faces of an ice crystal that explain the diverse habits seen in atmospheric ice crystals.

This coefficient has also found extensive use in theoretical models of cloud particle growth (e.g., Pruppacher and Klett, 1997, p. 597). For these models to simulate crystal growth rates successfully, they must account for the transport of vapor to the surface as well as incorporation. The two “resistances” to crystal growth presented by vapor diffuse and surface kinetics act in series and vary in magnitude as functions of several parameters. In particular, the habit of any ice crystal may be affected by the deposition coefficient, but the mass growth rate is limited by surface kinetics only when this resistance is very low, the particle is very small (less that 50 μm maximum dimension), or the total pressure is quite low (small diffusion resistance). The cirrus environment satisfies the second two conditions, but a realistic value for the deposition coefficient on small ice crystals is still very much in question.

Unfortunately, measurements of deposition coefficients suffer from an apparently contradictory record. In fact, reported values of α_m from ice range from 0.001 to unity (see Haynes et al., 1992, and references therein). In our opinion, a deposition coefficient of unity for ice is unlikely because the complex morphology of pristine ice crystals could not be explained, and experimental evidence abounds that both the basal and prism faces of ice crystals exhibit variable and inefficient growth at relatively high temperatures [Lamb and Scott, 1974; Nelson and Knight, 1998; Fukuta and Takahashi, 1999; Nelson, 2001]. However, most of the experimental data rely on large crystals, some on substrates, grown at temperatures above those typical of cirrus clouds. Of the few measurements made at relevant temperatures, Isono and Iwai [1969] measured crystal growth rates near -75 °C and determined α_m to be about 0.06, while Choularton and Latham [1977] measured the deposition coefficient to be 0.001 at -37 °C, and Haynes et al. [1992] found values above 0.6 over a broad range of very low temperatures (below -90 °C). These apparently contradictory values could be explained by the diverse experimental conditions employed and by an as-yet unclarified dependence on environmental variables, such as temperature, pressure, saturation ratio, and even the defect structure of individual particles [Gierens et al., 2003; Bailey and Hallett, 2004]. In any case, it appears that most previous measurements of the deposition coefficient have been made at saturation ratios, temperatures, or crystal sizes that are not representative of real cirrus particles.

Numerous recent atmospheric observations have uncovered surprisingly large ambient supersaturations (sometimes at much as 100% above ice saturation), as well as exceptionally large concentrations of very small ice crystals [Kahn et al., 2003; Gierens et al., 2004; Gayet et al., 2004; Jensen et al., 2005a]. Gierens et al. [2003] modeled the effect that low deposition coefficients for small ice crystals could have on cirrus evolution and concluded that if a strong kinetic growth resistance was occurring in cirrus, it would help to explain discrepancies between cirrus cloud models and findings from in-situ observations. In fact, many numerical models of cirrus formation presume that vapor deposition to chemically clean ice proceeds with high efficiency [Heymsfield and Sabin, 1989; Boehm et al., 1999; Gierens et al., 2003; Jensen et al., 2005b]. It has been established that if the deposition coefficient for small ice crystals is less than about 0.2, the microphysical properties of cirrus clouds are sensitive to the efficiency with which the ice takes up excess vapor [Lin 2002]. Jensen et al. [2005b] found that deposition coefficients on the order of 0.05 would cause unrealistically large concentrations of small ice crystals, but Gierens et al. [2003] performed detailed calculations based on the particle-growth model of Pruppacher and Klett [1997, p. 597] and became convinced that mass accommodation coefficients lower than 0.01 offer a compelling resolution to the apparent conundrum.

2. EXPERIMENTAL SYSTEM

In the research reported here we offer new measurements on ice particles that were grown in at cirrus temperatures. During independently repeated experiments, tiny water droplets were levitated, homogenously nucleated to ice, and observed as they grew and evaporated in response to varying saturation ratios.

The conception, design, construction, and characterization of this system are described in detail in the thesis of Magee (2006). This engineering work built on the efforts of Shaw (1998) and Shaw et al. (2000) and has resulted in a system of considerable complexity.

The laboratory system is centered about a quadrupole electrodynamic levitation cell which confines a small particle (typically an ice crystal) within a vertical wind tunnel. The particle is suspended in a vertical airstream through a combination of drag and electrodynamic forces (Fig. 1). The temperature of the ambient airflow is controlled by circulating baths to a minimum temperature of approximately -70 °C, as measured by a Type-T thermocouple to a precision of 0.03 °C. Supersaturation of the air flow is generated as pre-humidified air passes over the warmed surface of an ice coil just before encountering the levitation cell (Fig. 1). Ice-supersaturations approaching liquid water saturation can be achieved, although modest undersaturation occurs in the default state. Variable saturation ratios can be measured to within 5% (although the differential resolution is +/- 0.1% of the saturation ratio) by a technique employing thermocouple measurement of the final flow temperature and a model of mass and energy balance. This method of humidity calculation was

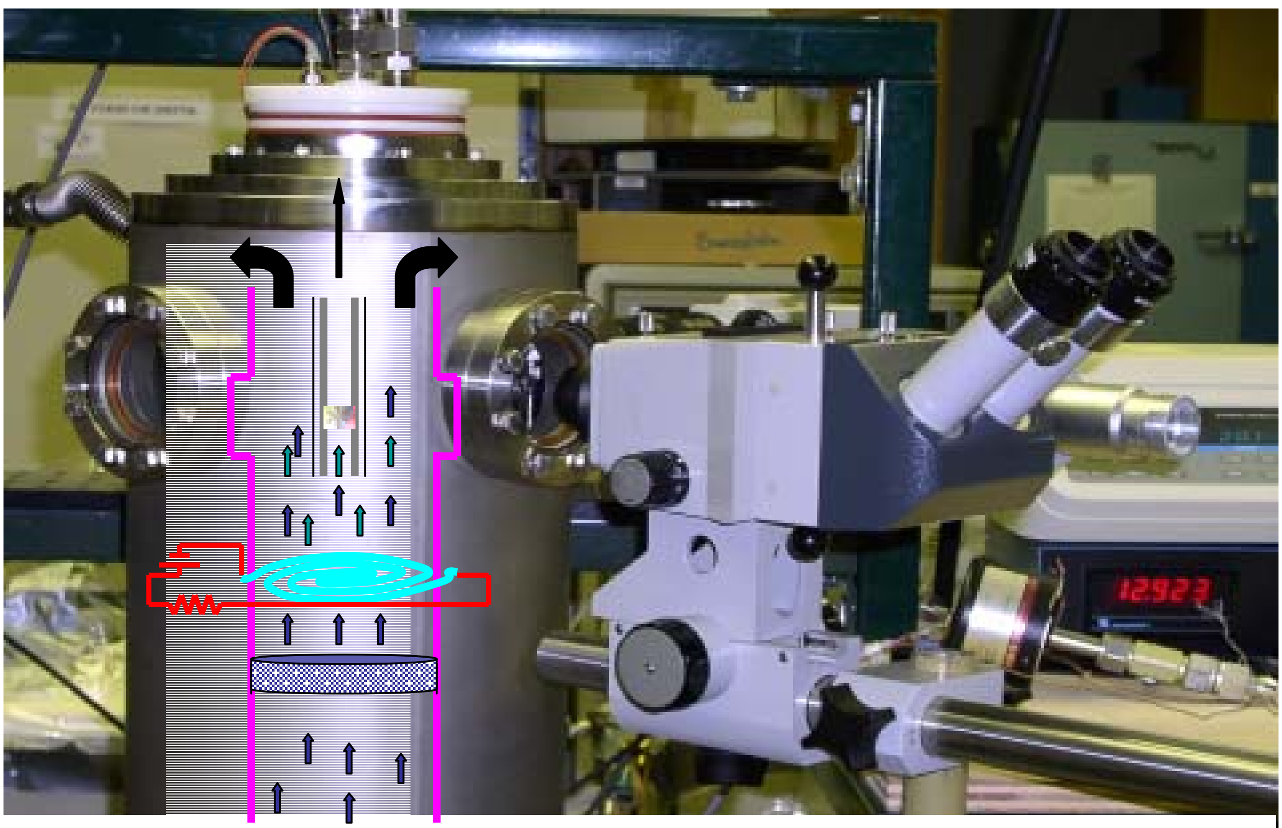


Figure 0. Photograph of exterior chamber with cut-away of environmental chamber (magenta). The in-chamber ice bed is cross-hatched and the ice-coated coil is shown in blue.

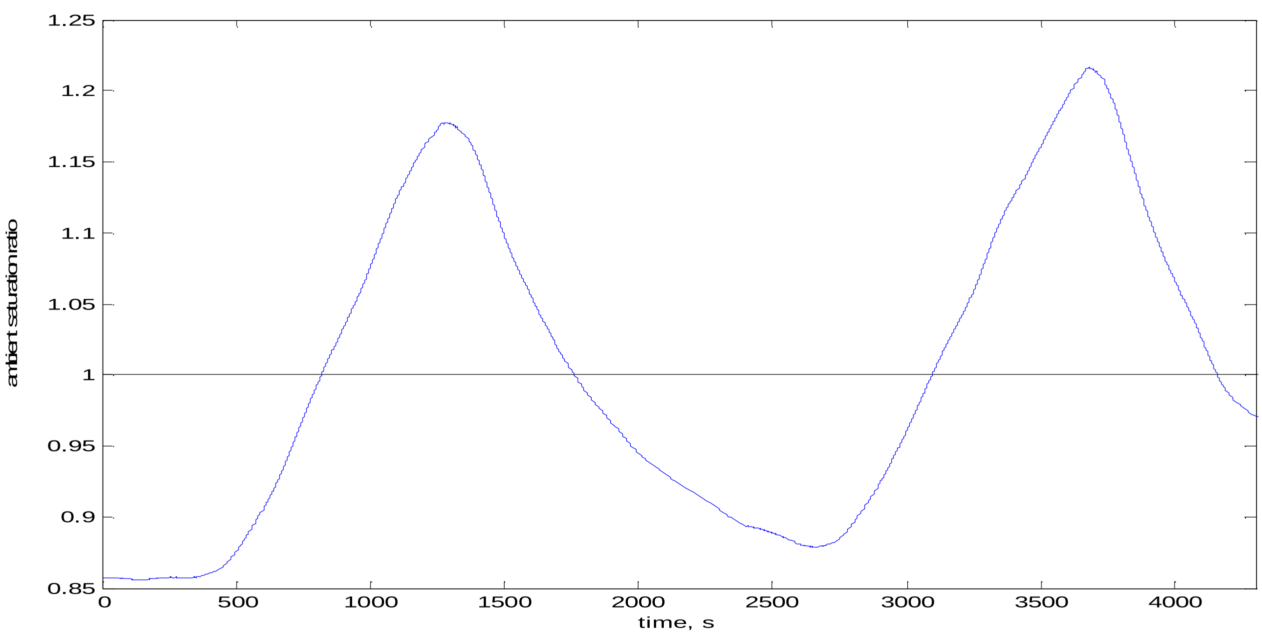
tested against the humidity calculated through an independent calibration (Magee 2006). In this calibration, the measured sizes of levitated H_2SO_4 solution drops were compared to the equilibrium sizes of the solution drops for the calculated ambient saturation ratio (Carslaw et al. 1995).

The conditions surrounding the growing ice crystal were monitored by an array of thermocouples, flow meters, a manometer, as well as the frequency and voltages of the levitation electrodes. Because the ice particles observed in these experiments were very small (~5 µm radius), it was not possible to use optical techniques. However, a technique was developed that exploits the boundaries of stable levitation to determine changes in the charge-to-mass ratio of a levitated particle (Magee, 2006). The domain of stable levitation is well defined by the Mathieu function for our five-electrode geometry (Hu and Makin, 1991 a;1991b), so the charge-to-mass ratio of the levitated object at the stability boundary (referred to as a "springpoint") can be accurately determined. While a growing or evaporating particle was levitated, the AC frequency was repeatedly adjusted to match this stability boundary, providing a record of particle mass change throughout each run. This technique was tested against particle sizes determined independently from Mie-scattering techniques (Xue et al. 2005), and the Mathieu analysis was found to give mass measurements more precise and far more sensitive than do Mie-scattering techniques. Based on the multiple-technique comparisons and the available frequency resolution, the measured changes in particle mass were accurate to within +/- 1% (Magee 2006).

3. RESULTS

Measurements on small ice crystals were made over the course of several months using independent water samples (distilled and dionized). The data presented here trace the mass of the ice particles as they were subjected to cyclic variations in ambient humidity, while the nominal temperature was held approximately constant. The data generally cover one or more hours of levitation for each crystal.

The first experiment was conducted in a vertical flow (5.2 l/min) of pre-conditioned purified air (Balston) with an initial temperature of -50.2 °C. A charged water droplet was injected into the levitation cell and examined for about 80 minutes. The initial saturation ratio with respect to ice was measured to be 0.86, and it was then driven to a maximum saturation ratio of 1.18 (Fig. 2). The supersaturation coil was turned off and the ambient environment relaxed to a subsaturated



state before supersaturation was reestablished. Measurement of particle mass was made at approximately two-minute intervals by measuring the Mathieu stability boundary. As expected, the ice particle was observed to lose mass when the measured saturation ratio was below 1.00 and increased in mass at all values above 1.00. However, when incorporating the measured physical parameters into a standard model of ice-particle growth, it became apparent that the particle was growing and evaporating far more slowly than predicted. Only by including a large kinetic resistance to vapor deposition could the measured behavior be modeled successfully. During the experimental initiation phase, the particle required α_m = 0.0025 and thereafter was well described by α_m = 0.006 +/-0.0015 . Figure 3 displays the varying size of the measured particle along with the deposition coefficient which was required to model the data.

The second experiment was conducted at an initial temperature of -40 °C with an identical airflow profile. In this experiment, the ice particle was levitated for about 1 hour and was exposed to an ambient saturation profile beginning at 0.92, rising steadily to 1.14, relaxing to 0.93, and then increasing again to 1.12. The particle responded to the ambient vapor field as expected, but once again it was clear that a very strong kinetic resistance was necessary to explain the sluggish growth and evaporation. The initial evaporation followed α_m = 0.002 and α_m = 0.006 +/- 0.002 during the remainder of the experiment (Table 1).

The third experiment took place at a nominal temperature of -60 °C, and followed the same pattern as the first two experiments. The only variation resulted from only one cycle of variation in ambient humidity. In this case the initial saturation ratio was smaller (0.72) and the maximum saturation was greater (1.44), but this still represents a smaller vapor density deficit or excess (compared to the first two experiments) because of the reduced temperature. Once again, the response of the crystal to the measured vapor profiles required a very small deposition coefficient: α_m = 0.004 fit the behavior of the crystal during the entirety of the experiment.

4. DISCUSSION

The small deposition coefficients that are implied by these measurements are surprising indeed. They indicate that over a range of temperatures typical for cirrus clouds, small ice particles make very inefficient use of excess water vapor. This interpretation of the data is clearly dependent on the precision of the measurements and the application of the particle-scale model, so it is appropriate to question the robustness of the implication for strong kinetic resistance.

The particle-scale model itself is based on the traditional capacitance model of Pruppacher and Klett (1997, pg. 597). This model may be inappropriate for crystals with large aspect ratios and complex habits, but the crystals in our experiments began as frozen spheres and were

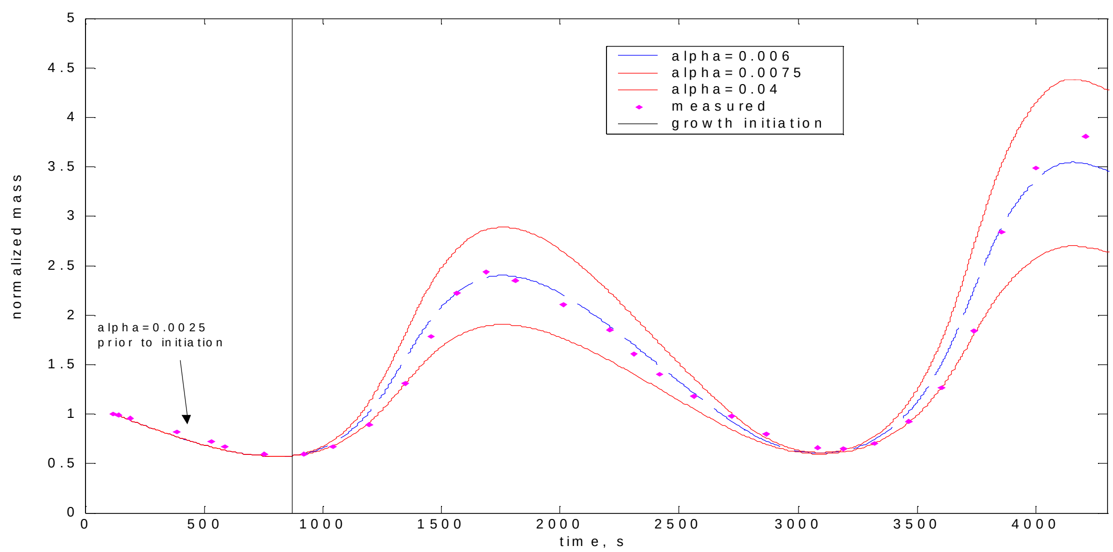


Figure 1. Measured variation in mass over the course of the first experiment at -50 °C (diamonds). The solid li indicate the modeled particle mass based on a range of plausible deposition coefficients.

likely to maintain a compact shape. Furthermore, this particle model has been shown to give an accurate description of particle evolution under conditions where kinetic influences are less important (Chen and Lamb, 1994).

It is also appropriate to question whether uncertainties associated with the measurements are sufficient to cast doubt upon the findings. The particles were small (~5 cm initial radius), so the model is very sensitive to low kinetic resistances. Also, the model sensitivity associated with the imprecise measurement of saturation ratio (+/- 3%) or particle mass (+/- 1%) pales in comparison to the influence of an uncertain kinetic resistance. Certainly, it was not possible to determine the deposition coefficient to within better than +/- 25%, but even this degree of uncertainty firmly places the values below most of those in the literature.

It has been suggested that chemical impurities or effects of the levitation electric field could account for the small observed growth rates. However, the ice particles, unlike those expected in the atmosphere, were relatively pure (originating from distilled and deionized water) and were grown in filtered air. If surface impurities contributed to the inefficient processing of incident vapor molecules, it would seem that this effect would only be magnified in real cirrus clouds. It is also unlikely that the electrical characteristics of the levitation device could have significantly affected the particle behavior. The levitation chamber does create regions of moderate electric fields in order to confine the particles, but the net potential gradient approaches zero at the exact location of the levitated particle. Furthermore, measurements by Bacon et al. [2003] on electrodynamically levitated ice crystals offer evidence that the moderate electric fields associated with this technique do not affect the net mass transport.

With little information beyond particle mass, it is not possible to speculate on the specific mechanisms associated with the strong kinetic resistance. The results are certainly surprising in the context of other measurements of the deposition coefficient, but it should be noted that experiments under these specific conditions have not been conducted. It is likely that the deposition coefficient is a function of several thermodynamic and crystallographic parameters. The kinetic resistance could well depend upon the crystal size, ambient temperature, degree of vapor excess, and possibly upon the crystal defect structure. It is quite plausible that the deposition coefficient could vary significantly from one crystal to the next within the same cirrus cloud.

The experiments here were intended to simulate the environment experienced by nascent cirrus crystals as closely as possible. The "wall-less" levitation technique approaches this goal quite closely, and the conditions surrounding these ice particles are very close to those found in a cirrus environment with the notable exception of total pressure. Engineering constraints required that the levitation occur at atmospheric pressure, rather than at the ~100-400 hPa values typically found in real cirrus. We expect that a lower total pressure would serve to increase the near-surface ambient saturation ratio, which could contribute to a modest increase in the kinetic incorporation

Table 1. The mass accommodation coefficient (α_m) used in the model calculations for three different ice particles levitated at different temperatures. The particles were levitated on different days, using independent water samples for the initial drop.

Ambient Temperature (ºC)	***Initial drop radius (μm)***	***α_m during initialization phase***	***α_m during primary phases***
-40	7.8 +/- 1.2	0.002 +/- 0.002	**0.006 +/- 0.002**
-50	5.3 +/- 1.2	0.0025 +/- 0.002	**0.006 +/- 0.015**
-60	7.0 +/- 1.2	0.004 +/- 0.002	**0.004 +/- 0.002**

efficiency. However, the lower pressure would conversely act to strengthen the relative influence of the kinetic resistance to growth compared to the resistance offered by diffusion. New experiments are planned that will allow measurements to be made at low pressure.

5. CONCLUSIONS

The experimental technique of merging electrodynamic levitation with a vertical wind tunnel worked well for this study of ice-particle growth at temperatures characteristic of some cirrus clouds. Levitation, especially when used in a "wall-less" flow configuration, keeps the particle isolated from wall influences and within a narrow field of view. The upward flow of cold gas through a packed bed of ice and a warmed ice coil permits the appropriate manipulation of gas properties and the development of calibrated supersaturations. Future studies will exploit these novel features of the system and explore the factors that limit the growth efficiency of small ice particles.

While we do not believe our findings define the full range of possibilities, these measurements offer some of the strongest evidence to date that small deposition coefficients are experienced by real cirrus crystals. Because of the role of vapor diffusion, even very small deposition coefficients would not strongly affect the growth characteristics of middle-sized and large cirrus crystals (maximum dimension > 50 μm). However, these results suggest that the development of recently nucleated ice particles might have difficulty growing.

The experimental results of this study appear to agree with the hypothesis of Gierens et al. (2003) that a kinetic resistance at the surface of atmospheric ice crystals could help to explain some of the apparent discrepancies between in-situ measurements and cirrus cloud models. It was not possible to define a single value or trend for deposition coefficients of different crystals under different conditions, but all measured crystals required a deposition coefficient less that 0.01 for successful simulation. Although a unique deposition coefficient has not been found that would be appropriate for direct application to all cold-cloud modeling, it is clear that simulations of cirrus clouds must not ignore surface-kinetic resistance of small ice particles.

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