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Comparison of model results of collection efficiency of aerosol particles by individual water droplets and ice crystals in a subsaturated atmosphere

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Abstract

The aerosol collection efficiencies of water droplets and ice crystals are compared based on the concept of equivalent geometrical kernel K^* which is the geometrical sweep-out volume per unit time by the collector. It is thought that the comparison based on this quantity reveals the real difference of the aerosol collecting abilities of different collectors and sheds lights on the precipitation scavenging mechanisms. The collection efficiencies are taken from theoretical model results computed by us previously at relative humidities of 95% for water droplets, columnar and hexagonal plate ice crystals. It is shown that the efficiencies are rather insensitive to collector shape for aerosol particles smaller than 0.01 μm. The shape factor becomes more important for larger aerosol particles, especially in the Greenfield–Gap size range.

1. Introduction

One of the most efficient ways that aerosol particles can be cleansed from the atmosphere is via the cloud and precipitation process. In this process, particles either serve as cloud condensation nuclei (CCN) or ice nuclei (IN) to form water droplets or ice crystals, or become attached to water droplets and ice crystals by other mechanisms. This paper is mainly concerned with the latter. Furthermore, we shall discuss only the cases where the atmospheric condition is subsaturated as is the case for most parts of a cloud. Clouds and precipitation may consist solely of water drops, or solely of ice crystals, or both. All three cases exist in any season. Clouds in winter time may consist of liquid drops and clouds in

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summertime may contain ice particles. Thus a complete understanding of precipitation scavenging, as the above cleansing mechanism is commonly called, would necessarily involve the study of both rain and ice scavenging. The main question of interest in the present paper is the relative scavenging efficiencies of water drops and ice crystals. There are comments in the past that snow is a more efficient particle scavenger than rain (e.g., Carnuth, 1967; Magono et al., 1974; Murakami et al., 1985a, b). The comparisons were made based on the equivalent liquid water contents. For data derived from field observations, this probably the only way to compare the efficiencies. Recent theoretical studies and laboratory measurements (e.g., Slinn and Hales, 1971; Wang and Pruppacher, 1977; Wang et al., 1978; Martin et al., 1980a, b; Murakami et al., 1985a, b; Sauter and Wang, 1989; Miller and Wang, 1989) make it possible the comparison between efficiencies by individual collectors.

2. The basis of comparison

One question that has often been asked when the scavenging efficiencies of rain and snow are compared is: what is the basis for comparison? This is a valid question that should be answered before we proceed. The most straightforward method of comparison is the one based on the size of the collectors. This type of comparison is easily understood when the collectors have the same shape (e.g., both are spheres or hexagonal plates) but will lead to confusions when collectors are of different shapes. For example, a raindrop of the same diameter as a hexagonal ice plate will normally fall much faster than the plate. If the drop collects more aerosol particles, it may simply be that the drop has traveled a longer distance and hence has more chance to collect particles. This is also true for ice particles of the same size but of different shapes. Another method is the one already mentioned above, namely, make the comparison based on the equivalent liquid water contents or equivalent rainfall rates (e.g., see comments in Wang, 1992). While this may be useful for field observational data in view of the difficulty in separating various kinds of collector particles and mechanisms, it is to be noted that this method considers the integrated effect of the whole collector size (and often also shape) distribution. The real ability of each collector particle is blurred by that of other particles. Thus, in the present paper only the collection efficiencies of individual collectors will be compared. Such a comparison will be useful especially in the elucidation of the physical mechanisms of scavenging and their relative contributions in the collection and removal of atmospheric aerosol particles. A comparison of the collection efficiencies is to compare the "ability" of the collectors to collect particles, hence we should use a fair basis of the comparison. We propose here to use a quantity K^* which is the geometrical sweep-out volume per unit time for this purpose. This quantity has been introduced in Wang and Pruppacher (1980) and Wang (1983) and was called "geometrical kernel" in these papers. There are also suggestions that the term "geometrical sweep-out volume per unit time" should be used to avoid confusion with the term "scavenging kernel". The relation between K^* and the conventional scavenging kernel K (see, for example, Pruppacher and Klett, 1978) can be made clear from the following expression:

$$E = K/K^*$$

i.e., the scavenging kernel is simply the collection efficiency times K^*, or, alternatively, the collection efficiency is simply the ratio of K to K^*. Thus, for example, the K^* for a drop of radius a is

$$K^* = \pi a^2 V_\infty$$

while K^* for a columnar ice crystal of length L and radius a is

$$K^* = 2aLV_\infty$$

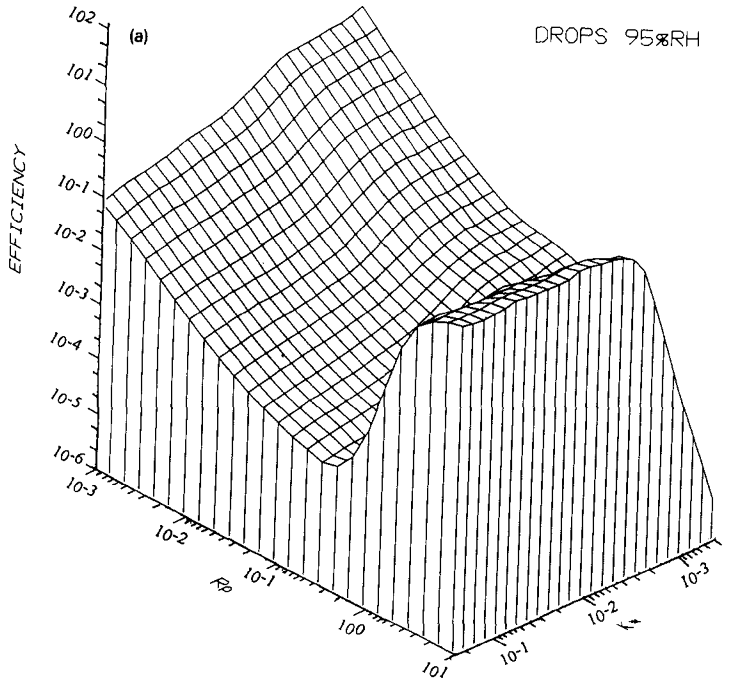
where V_∞ represents the terminal fall velocity of the collector. In this definition of K^*, we have assumed that all hydrometeors fall with their largest dimensions oriented horizontally. This assumption is valid for smaller hydrometeors which are being considered in the present paper. For larger hydrometeors, the orientation may change with time and the definition of K^* has to be modified. But this will not be considered here. The rationale of using K^* as the basis for comparison of collection efficiency is as follows. Since we are comparing collectors of different shapes, we should compare their efficiencies under the condition that they are given the same chance to be exposed to the same amount of aerosol particles per unit time. We shall assume that the aerosol concentration is uniform which is necessary for the fair comparison to be made. Under this condition, the same volume of air would contain the same amount of aerosol particles. Thus when we compare the collection efficiencies for collectors with the same K^*, we are exposing the collectors (even though of different shapes) the same amount of aerosol particles per unit time. The one collects more particles does so because it is really more efficient in getting particles. Hence using K^* as the basis for comparison is "fair". Apart from Wang and Pruppacher (1980), Podzimek (1987) also used the same technique for comparing the collection efficiencies.

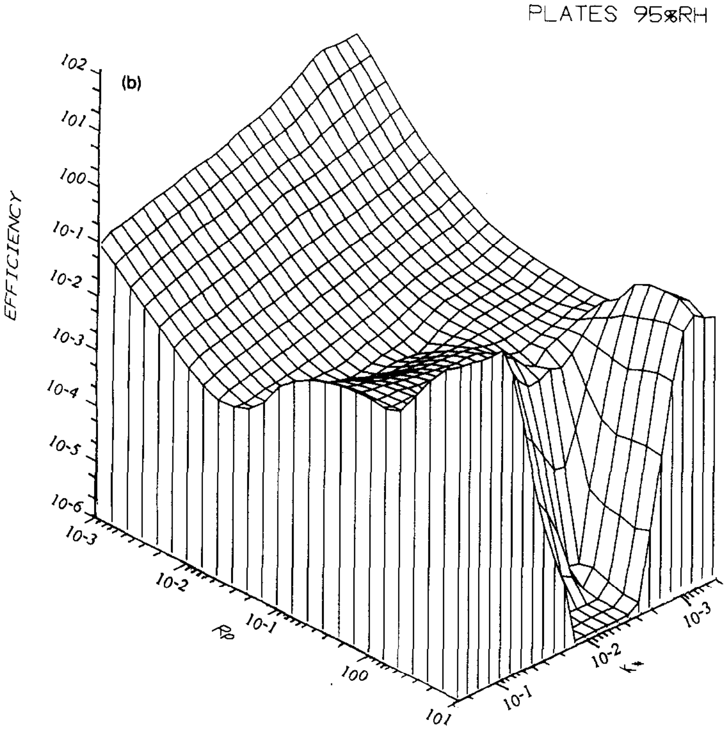
3. Data sources of collection efficiencies

The "data" used for the present comparison will be the results derived from model calculations based on the models developed by Wang et al. (1978) for drops, and Martin et al. (1980a, b), Wang (1985) and Miller and Wang (1989) for ice particles. Ideally, experimental data such as those obtained by Wang and Pruppacher (1977), Lai et al. (1978), Prodi (1976), Murakami et al. (1985a, b), Sauter and Wang (1989) and Song and Lamb (1992) should be the ones used for the comparison. However, at present the experimental data are not complete enough to form continuous data sets and the available sets do not overlap substantially in the range of K^*. This makes the comparison difficult. Hence it was decided to use the model results for such a comparison. Fortunately, the validity of these models has been confirmed by experimental measurements, thus it is felt that the comparison is meaningful. In the present comparison, the results of collection efficiencies calculated under the atmospheric condition of 95% relative humidity are used. These are taken from the results obtained in Wang et al. (1978) for drops, Martin et al. (1980a) for hexagonal ice plates, and Miller and Wang (1989) for columnar ice crystals. Note that the drop collection efficiencies are calculated under the condition of $p = 1000$ mb, $T = 20°C$, while that for the ice crystals are $p = 700$ mb, $T = -10°C$ for plates, and $p = 600$ mb, $T = -20°C$ for columnar ice crystals, respectively. Wang et al. (1978) have shown that the pressure and temperature conditions have little effect on the collection efficiency. Note also that here we define the RH as that with respect to saturation vapor pressures over plane water and plane ice surface, respectively, instead of the commonly used definition which uses the saturation over plane water surface as the standard. Thus, the condition RH = 95% is different for drops than for ice particles. But again the resulting differences of collection efficiencies using different definitions are typically only a few percent, hence would not alter the general conclusions to be discussed below.

4. Comparing the collection efficiencies of water drops and ice crystals

Figs. 1 and 2 show the collection efficiencies of water drops, hexagonal ice plates, and columnar ice crystals. In order to emphasize the differences between these efficiencies, the logarithmic scales are used. The range of the original values of E is from 10^{-6} to 1, Rp from 0.001 to 10 s, and K^* from 3.7×10^{-4} to 0.6 cm^3/s. These two figures are based on entirely the same data sets except that they are viewed from different angles. It is immediately seen that from Fig. 1 that the collection efficiencies for small Rp for water drops, ice columns, and ice plates are very similar to each other. This indicates that these three kinds of collectors are equally efficient in collecting aerosol particles of about 0.001 μm radius. The reason that the efficiencies are almost the same independent of collector habit and size is because that at this particle size range, the most important collection mechanism is the aerosol Brownian diffusion which depends mainly on the aerosol particle size. They are





therefore fairly insensitive to the habit of collectors. The collection efficiencies also decrease with increasing K^* for all three collectors at about the same rate. This is entirely due to the way the efficiency is defined in the first equation of Section 2. Here the collection kernel K is virtually the same but as K^* increases, E decreases. Thus, as all three collectors become larger, their efficiencies decrease because the number of particles they can capture remains pretty much the same even though the volumes swept by them per unit time increase. Subtle differences exist among the three collectors but the magnitudes are too small to be significant. Moving along the Rp-axis reveals more significant differences between the three collectors. If we look at the E for larger K^* (the left-hand-side of the axis), we see that while the efficiencies for drops and columns are similar in magnitude, that for plates are drastically different. First of all, the E for all three collectors initially decrease with increasing Rp due to the decrease in the Brownian collection. For drops and columns, however, the efficiencies reach a minimum near Rp = 1 μm, then increase with Rp as the inertial impaction becomes important. For plates, the minimum is located between 0.01 and 0.1 μm. Thereafter, E rises with Rp. It is also obvious that the E for plates over the Rp range between about 0.02 and a few μm are much higher than that of the drops and columns. For example, at Rp = 1 μm, the E of plates are typically at least about two orders of magnitude higher than that of columns and drops. This size range is the so-called Greenfield–Gap range. Thus, the plates are much more efficient in removing the Greenfield–Gap particles than do columns and drops. Fig. 1 is only clear for E in the larger K^* range. To see the other side of the story,

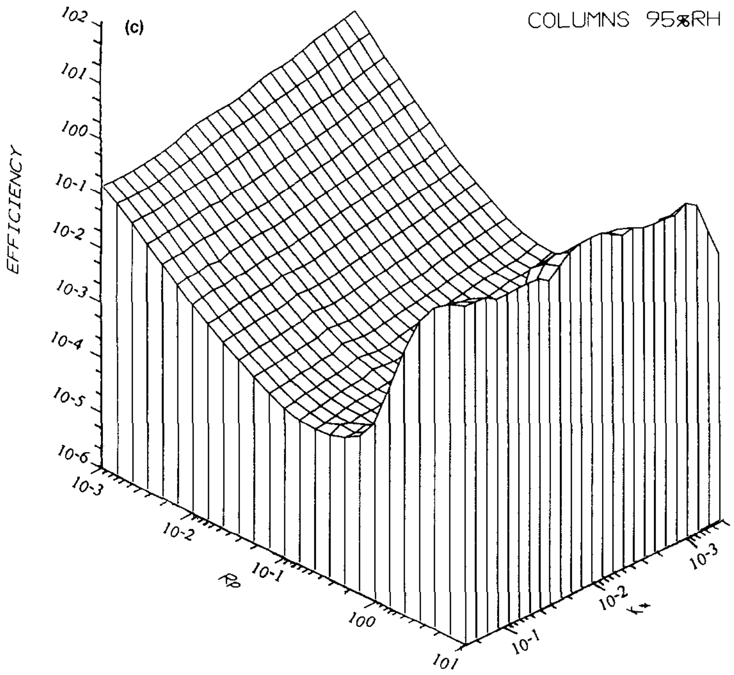


Fig. 1. Collection efficiencies of aerosol particles captured by (a) water drops, (b) planar and (c) columnar ice crystals at relative humidities of 95% as computed by Wang et al. (1978), Martin et al. (1980a) and Miller and Wang (1989), respectively. The particle radius Rp is in the unit of micron. The unit for K^* is cm^3/s.

we have to look at Fig. 2. Here we see that what we said in the previous paragraph is also true for smaller K^* range. Unlike the drops and columns, the E-surface of plates does not have a very deep gap along the Rp-axis. The gap seems to be especially deep (and narrow) for columns. For drops, the gap is more gradual, although there is a very deep "pit" near $K^* = 2.7 \times 10^{-3}$ which corresponds to drop radius of 42 μm which is not very easy to see in this type of diagram. Both the gaps of columns and droplets are much deeper in this side where the collector sizes are small than the other side. Thus, small columns and droplets are hardly effective in removing aerosol particles of about 1 μm size. Before reaching this gap region, all three collectors have relatively smooth E-surfaces, indicating that the variation of E with K^* is gradual for all three collectors. This remains the case for columns and drops throughout all K^* values. But that for plates are again very different. We see from both Figs. 1 and 2 that there is deep "crack" in E for plates if we move along K^*-axis. This crack is located over the plate size range between about 100 and 200 μm s. The efficiencies are rather small for these plates at Rp > 1. Indeed, if we examine the E-curves in Martin et al. (1980a, b), we see that there are local maxima of E at Rp about 1 μm for $N_{Re} = 1, 2$ and 5. E decreases with increasing Rp for these cases. This can also be seen more clearly in Fig. 2b if we move along the Rp-axis. This special collection behavior of plates is probably due to the strong pressure built-up near the stagnation point. Unlike columns and drops whose surface curvatures near the stagnation points remains constant, that of plates are fairly flat. This results in high pressure built-up over a larger area on plate surface than on drops and

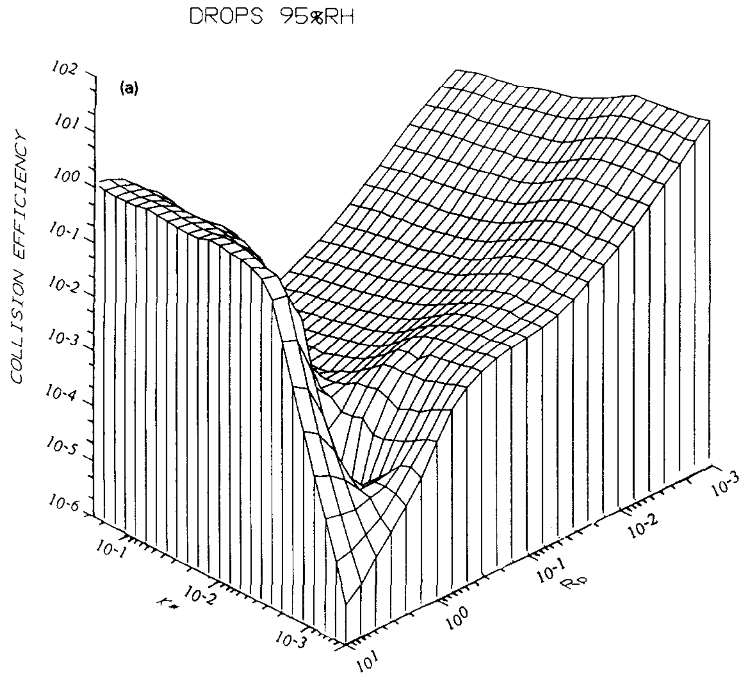


Fig. 2.

columns. This large high pressure region prevents easy penetration of aerosol particles to the surface. Thus particles may get deflected away from the surface, as having been pointed out by Pitter and Pruppacher (1974), Pitter (1977) and Martin et al. (1980a, b). This phenomenon is particularly efficient in reducing the collection efficiency if the plates are small enough such that the resulting relative velocities between them and the aerosol particles are so small that particles have sufficient time to be totally deflected. For smaller aerosol particles, the relative velocities will be larger and therefore this effect is not as important. Fig. 1 also shows that the collection efficiencies of drops at small K^* and larger Rp are much smaller than that of columns and plates. This is due to the fact that at small K^*, the terminal velocities of drops are not much different from that of the aerosol particles. The collector and collectee are thus falling at about the same speed and hence the low collection efficiency. It is also seen in Figs. 1 and 2 that at small K^*, columns have the highest collection efficiencies among the three collectors for $\mathrm{Rp} > 1\ \mu\mathrm{m}$. At large K^*, drops have higher efficiencies than both columns and plates.

5. Conclusions

In the above we have compared the calculated collection efficiencies of cloud droplets, ice plates, and ice columns. Several conclusions can be drawn from the above discussions: (1) The three collectors are about equally efficient in collecting very small ($\mathrm{Rp} < 0.01$)

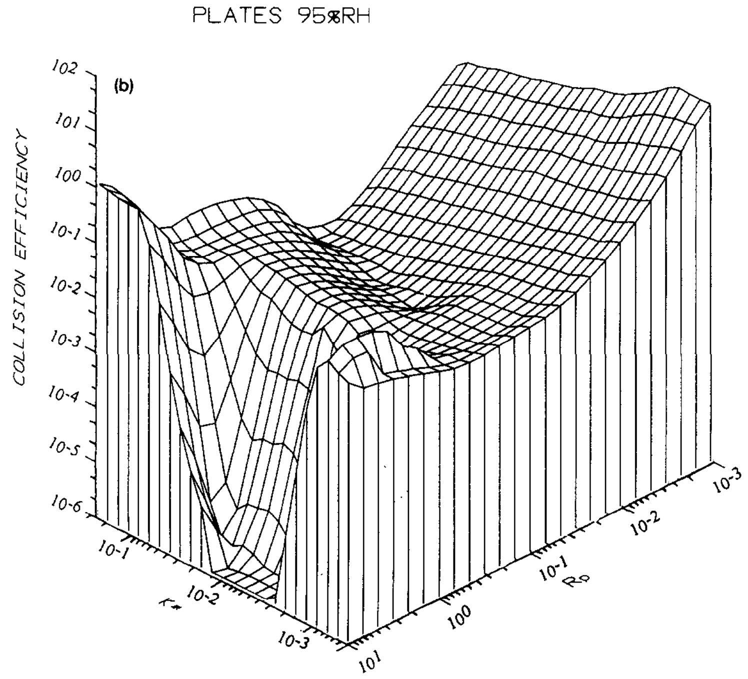


Fig 2.

aerosol particles. Thus it should be expected that the removal of these small particles should be fairly independent of the details of cloud processes since it is mainly a function of the size of aerosol particles. Although calculations of the efficiencies of larger hydrometeors (large raindrops, graupel, hailstones) have not yet been done, it is to be expected that their efficiencies will not be too much different from these three collectors when the removal of very small particles are concerned. Thus, to these small particles, clouds and precipitation behave like filters whose filtration efficiency depends mainly on the mechanical arrangement of the filter elements (number of collectors, pressures, etc.) but not much on the physical and chemical properties of these elements. (2) To aerosol particles in the size range between 0.01 and 1 μm (the Greenfield–Gap particles), ice plates have the highest efficiencies in removing them. The efficiencies of cloud droplets and columns are generally one to two orders of magnitude lower than the plates. At present, no observations have been made of the habit distributions of ice crystals in clouds. Hence it is impossible to deduce what this fact may imply. However, if we take the results of crystal habit obtained in laboratory experiments (see, e.g., Magono and Lee, 1966; also fig. 2-26 in Pruppacher and Klett, 1978), then it may be said that plate ice crystals exist mainly in the middle level of a deep convective clouds where the temperatures are between − 10 and − 20°C whereas in the higher part of the cloud where temperatures are below − 20°C columns are dominant. Thus the Greenfield–Gap particles may be most efficiently removed in the middle level of the cloud and inadequately removed in the high level. Thus these Greenfield–Gap particles may "leak" into the upper troposphere/lower stratosphere due to the inefficient filtration of

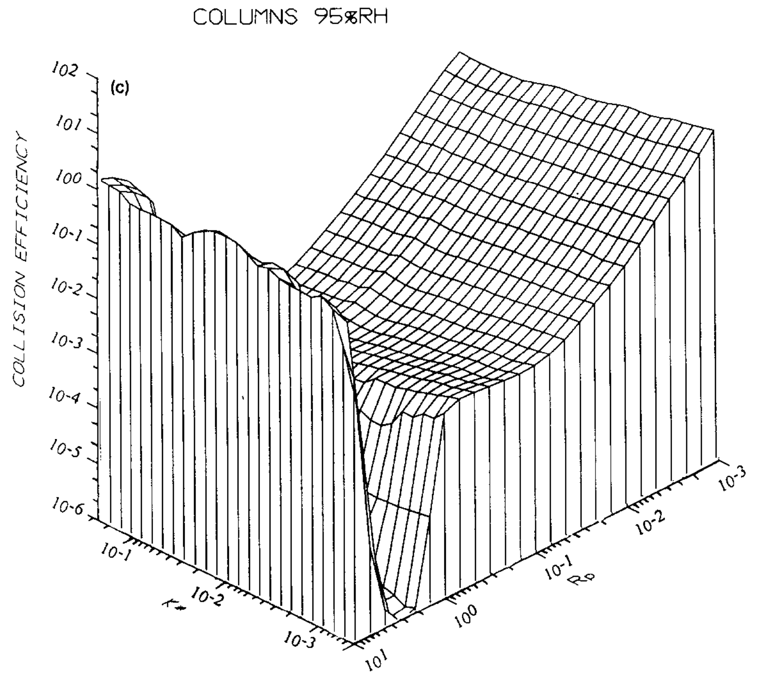


Fig. 2. Same as Fig. 1 except viewing from a different angle.

columnar crystals whereas the middle troposphere may be most "clean" in terms of the number concentration of these particles right after such a cloud process. This also seems to be consistent with the observations of Changnon and Junge (1961) where there is a local maximum of "large" particles at 15–20 km level and a local minimum at 5–10 km level. Of course, due to the insufficient observations of both the crystal habit distributions in clouds and the actual aerosol size distributions in different levels, this remains a conjecture. (3) For aerosol particles of radius greater than 1 μm, both droplets and columnar ice crystals are fairly efficient in removing them, with columns somewhat more efficient especially when the collectors themselves are small. This may be relevant to situations near the top of a deep convective cloud and in the initial stage of cloud glaciation. In these cases the ice crystals are expected to be small. On the other hand, ice plates are relatively inefficient in particle removal due to reasons mentioned before. Thus depletion of "giant" particles may be faster near the cloud top than in the middle levels. In a deep convective storm, there are of course other processes that must be considered before definite conclusions about the aerosol particle removal can be drawn. These processes include the nucleation of drops and ice crystals by aerosol particles, removal of particles by other types of hydrometeors, interactions between cloud and precipitation particles, convection and turbulent diffusion of particle plumes, electrical processes, etc. Nevertheless, it is expected that these processes can also be modeled in a similar fashion as in the present study.

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