SCAVENGING OF CONDENSATION NUCLEI IN CLOUDS: DEPENDENCE OF SIGN OF ELECTROSCAVENGING EFFECT ON DROPLET AND CCN SIZES

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1. INTRODUCTION

The effect of electric charge on the collection efficiencies for scavenging of aerosol particles by droplets was extensively modeled in the 1970s and 1980s by Pruppacher and his group. The interpretation of this work was that only in well developed thunderstorms would the amount of charge on the droplets and the particles they were scavenging be great enough to affect cloud microphysical processes. In recent years it has been realized that even in weakly electrified clouds such as marine stratocumulus the amount of electrical charge is sufficient to affect contact ice nucleation and CCN concentrations (Tinsley et al., 2001). Here we consider the dependency of the sign of the electroscavenging effect on the sizes of the droplet and CCN, and the implications for cloud processing of aerosol particles.

2. CLOUD CHARGING

Measurement of droplet charge in non-thunderstorm clouds have shown charges of ~ 100e ($e = 1.6\text{x}10^{-19}$C) (Pruppacher and Klett, 1997, sec. 18.4; MacGorman and Rust, 1998, ch.2, Bead et al. 2004). The source of this charge, especially for ice-free clouds of relatively large horizontal extent, appears to be the ionosphere-earth current density (J_z) which is the return current in the global electric circuit (Bering et al 1998). The conductivity (σ) is about an order of magnitude lower in clouds than in clear air at the same altitude (Pruppacher and Klett, 1997), and the flux of J_z through the conductivity gradients at cloud boundaries generates positive space charge at cloud tops and negative space charge at cloud bases. The electric field $E = Jz/\sigma$ and the vertical gradient in conductivity is $dE/dz = J_z d/dz(1/\sigma)$. The space charge ($\rho$) is given by Poisson's equation $\nabla\cdot E = \rho/\varepsilon_o$ so that $\rho = \varepsilon_o J_z d/dz(1/\sigma)$.

Aerosol particles and cloud droplets collect this space charge. A process that can produce the more highly charged aerosol particles is the evaporation of some of the charged droplets, e.g., with mixing at cloud boundaries. The evaporation residues retain the droplet charge, which decays on a time-scale of tens of minutes down to a lower charge, determined by the amount of space charge present and the particle radius (Tinsley et al. 2000). Aerosol particles are independently charged to this level by the attachment of air ions directly from the space charge, as will be discussed later.

3. ELECTROSCAVENGING

The effect on collision efficiencies of the charges on the aerosol particles and on the droplets has most recently been modeled by Tinsley et al. (2000, 2001). The scavenging for same-sign charges (q) on particles as charges (Q) on droplets will be the normal situation when space charge is present. For particle radii small compared to droplet radii the electrical force F is given by $F = C[(Kr + 1)/r^3 - r/(r^2 - 1)^2]$ where $K = Q/q$ and $C = q^2/(4\pi\varepsilon_o A^2)$ where A is the droplet radius and a is the particle radius and $r = a/A$ (Tinsley et al., 2000). For Q and q of the same sign the first term on the RHS is a long range repulsion while the second term is the short range attraction due to the image force that is independent of the sign of Q/q and increases rapidly as the particle approaches the droplet surface and r tends to 1.

The numerical integration of trajectories shows that for droplets of radii about 10 μm and greater the long range repulsion is unable to prevents the particles with radii greater than about 0.1 μm being carried, by the flow around the falling droplet, close enough to the droplet surface so that the short range attraction ensures collection. For the smaller particles with higher mobility, with the smaller droplets with smaller fall speeds, the long range repulsion keeps the particle out of range of the image force and can reduce the collection efficiency well below that would otherwise be due to collection by phoretic processes and Brownian diffusion.

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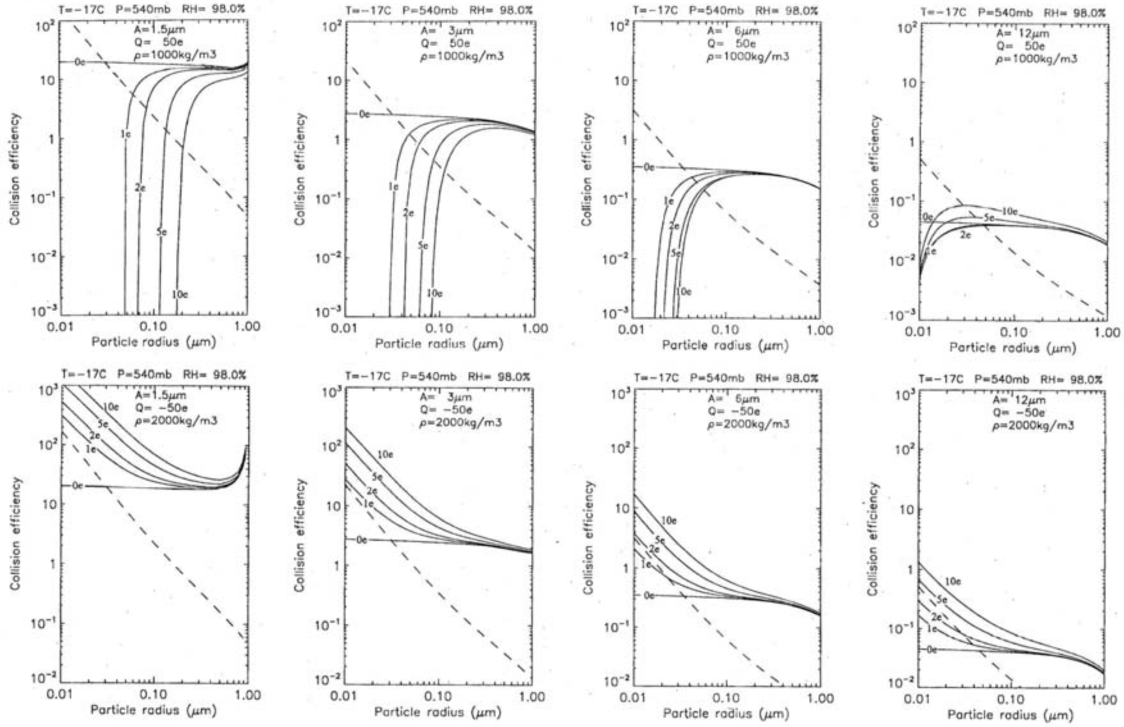

Figure 1. Variation of collision efficiency as a function of particle radius for four different droplet radii A. The top four panels are for droplet charge Q and for particle charge q of like sign; the bottom four panels are for Q and q of opposite sign. The curves are labeled by particle charge q. The atmospheric pressure, temperature, and relative humidity are 540 hPa, -17° C and 98% respectively. The curves for 0e show the effect of phoretic forces alone. The collection efficiencies for Brownian diffusion are calculated separately and are shown as dashed curves.

Figure 1 is a plot of collection efficiencies for droplets of 1.5, 3, 6 and 12 μm radii respectively, with charge Q of 50e, interacting with charged aerosol particles with charges q of 0, 1, 2, 5, and 10e, with the curves labeled by the particle charge. The upper four panels are for Q and q of the same sign, and the lower panels are for them of opposite sign. There is little effect of particle density for radii below 0.5 μm; the upper panels are for 1000 kg/m^3 and the lower panels are for 2000 kg/m^3 as can be seen in the curves for 0e.

The calculated collision efficiencies are a combination of phoretic scavenging and electroscavenging. The collection efficiency due to Brownian diffusion is calculated separately (Pruppacher and Klett, 1997). The droplet charge of 50e is consistent with the measurements of Beard et al. (2004).

4. APPLICATIONS TO CLOUDS

In order to determine the magnitude of the electrical enhancement or reduction of the scavenging of CCN, to produce the indirect aerosol effect, or of scavenging of aerosol particles in general, relevant to cloud processing of aerosol and gases, it is necessary to determine the distribution of charge q on the aerosol particles. There is little observational or theoretical information available. For monodispersive aerosol of radius a the equilibrium charge distribution is given by Harrison and Carslaw (2003) as:

$N_j/N_o = (x)^j \sinh(\lambda j)(\exp(-\lambda j^2))/(\lambda j)$, where N_j is the concentration of particles with j elementary charges, and; N_o the concentration of uncharged particles; $\lambda = e^2/(8\pi\varepsilon_o kTa)$ where T is the absolute temperature. The charge distribution is related to the space charge ρ by the parameter x. For a concentration n^+ of positive ions and n^- of negative ions the space charge $\rho = (n^+ - n^-)e$, and it corresponds to a positive conductivity $n^+\mu^+$ and a negative conductivity $n^-\mu^-$, where μ^+ and μ^- are the mobilities of the positive and negative ions. Then $x = (n^+\mu^+)/(n^-\mu^-)$.

This equation implies that for a plausible negative space charge accumulation at cloud base, say giving x=0.3, and temperatures corresponding to the middle troposphere (256K) about 70% of the particles of radius 0.05 μm are negatively charged with a mean charge of about –1e. For a = 0.1μm the percentage is ~80% with mean charge about -2e; for a = 0.2 μm it is ~97% with mean charge about –4e, and for a = 0.5μm it is ~99.9% with mean charge about –9e.

The actual atmospheric aerosol size distributions are far from monodispersive. The results from Pueschel et al. (1994) for maritime air far from land show concentrations of particles falling steeply with increasing radius, e.g. with more than three orders of magnitude concentration decrease for radii increasing from 0.05μm to 0.5μm. The effect of this on the charge distributions has not been evaluated, but if they remain similar to monodispersive distributions then a high percentage of the particles would remain negatively charged.

The effect of aerosol charge, and therefore space charge, on the scavenging of aerosol particles also depends on the droplet size distribution in the cloud. For the base of the marine stratus cloud in Figure 2-13 of Pruppacher and Klett (1997) the mean droplet radius in the distribution is near 4 μm, increasing to about 10μm at the top of the cloud. With reference to Figure 1here, and in view of the preponderence of aerosol particles with radii less than 0.1μm, one might expect that at cloud base the effect of space charge, for x = 0.3, would be a reduction in the phoretic and Brownian scavenging rates by a non-negligible amount.

4. COMPLEXITIES TO BE CONSIDERED

The electrically induced processes in clouds are likely to be even more complex than the above description would imply, as there are strong time dependencies present. The charging time to produce the space charge is of order 10 minutes, with the production beginning with the initial stages of cloud formation as updrafrs produce cooling and increased relative humidity. The formation of droplets from activated CCN lowers the conductivity due to attachment of air ions, and the increasing conductivity gradient begins to generate space charge. The reduction of conductivity in fog and pre-fog conditions has been reviewed by Dolezalek (1963).

The formation of charged droplets quickly scavenges the charged aerosols (and ions) of opposite sign (as illustrated in the lower row of panels of Figure 1) so that there is a positive feedback on the developing conductivity asymmetry. Whether the rate at which this occurs is fast enough to prevent equilibrium charge distributions from forming is not yet determined.

Overall, it seems possible that there are significant effects on aerosol and CCN concentrations from electroscavenging of aerosol particles at cloud bases, whether charged directly by space charge or as charged residues from evaporated droplets when mixing is present. For the aerosol particles of CCN size it seems that the net effect would be an increase in CCN concentration with increasing space charge, as particles are preserved from being lost by other scavenging processes. This could lead to macroscopic cloud and precipitation changes by the indirect aerosol effect. However, much cloud modeling work is needed to test this possibility.

5. SCAVENGING OF ICE FORMING NUCLEI

With larger aerosol particles, with radii in the range 0.1 μm to 1 μm, the relatively large charge on them and their lower mobility can result in an increase rather than a decrease in scavenging rates, relative to phoretic and Brownian rates, especially where the mean droplet radius is ~ 10 μm and greater. For cloud tops where the temperature is below freezing the scavenging of ice-forming nuclei can occur. Larger aerosol particles are more likely to be effective IFN than smaller ones, on account of the greater surface area for ice nucleation sites, and larger droplets are more likely to be present near cloud tops. For these conditions it seems likely that the rate of electroscavenging leading to contact ice nucleation will increase with increasing space charge (see Tinsley et al 2001).

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