

COLLECTION OF AEROSOL PARTICLES BY SNOW CRYSTALS

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ABSTRACT

Each year a substantial amount of aerosol particles are washed out from the atmosphere by precipitation. This process, commonly called precipitation scavenging, is probably the most significant natural process of aerosol removal. It is therefore very important to determine quantitatively the rates with which particles are collected by precipitation. The quantitative determination of the scavenging rates of aerosol particles by snow was not reported previously. In this paper we will report experimental measurements of the scavenging efficiencies of aerosol by snow crystals. A convective diffusion model will be presented also to provide theoretical explanation of the observed efficiencies. The convective diffusion model will consider the simultaneous effects of Brownian diffusion, thermo- and diffusiophoretic forces, electric forces, and ventilation effect into account. Theoretical results will be compared to that of the experiments.

1. INTRODUCTION

Each year a significant amount of aerosol particles (AP) in the atmosphere are removed by precipitation. Unquestionably, many are removed by snow. In order to assess the effect of snow events on the global AP equilibrium it is necessary to know quantitatively what are the removal rates of AP by snow crystals. This is the problem that the present paper intends to address. In the following we treat first the theoretical problem first and then experimental measurements.

2. THEORETICAL CONSIDERATION

The main difficulty in treating the theoretical problem of the scavenging of AP by snow crystals is in the complicated shapes of snow crystals, as a glance at the Magono-Lee diagram (P. 34 of Ref. 1) of snow crystals would reveal. In contrast, the AP scavenging by cloud and small raindrops is simpler because of the spherical shape of the collectors. This can be done for submicron aerosol particles by considering their convective diffusion toward the collector.

The following development is similar to that in Refs. 2-5 but here the shape factor will be considered explicitly. The concentration n of AP around a collector of an arbitrary shape under the simultaneous influence of conservative and non-divergent forces and Brownian motions can be described by the convective diffusion equation (see Ref. 3) where D is the diffusion coefficient of AP in air, B their mobilities,

$$\bar{f}_p D \nabla^2 n - B \vec{F} \cdot \nabla n = 0 \quad (1)$$

\vec{F} the vector sum of all external forces, and \bar{f}_p the ventilation factor due to the relative motion of the collector in air. The ventilation factor is included because Eq. (1) is

strictly satisfied when there is no relative motion between the collector and air. However, as long as the particles are small compared to the collector, such as the submicron AP collected by snow crystals, the inertial impaction is not important and the enhancement of the particle flux toward the collector due to the relative motion can be taken into account by multiplying the stationary flux by the ventilation factors (see Ref. 2 for detail). This approach has been experimentally verified for the case of drop-AP collision (Refs. 6-8).

The boundary conditions for the present problem are

$$\begin{aligned} n &= 0 & \text{at the surface of the snow crystal} \\ n &= n_\infty & \text{at infinity.} \end{aligned} \quad (2)$$

This set of boundary conditions can be replaced by

$$\begin{aligned} n &= 0 & \text{at } \phi = \phi_0 \\ n &= n_\infty & \text{at } R \rightarrow \infty \end{aligned} \quad (3)$$

where ϕ_0 is the surface force potential. The solution of (1) subject to this set of B.C.'s is

$$n = n_\infty \left[e^{-\frac{B}{D\bar{F}_p} (\phi_0 - \phi)} - 1 \right] / \left(e^{-\frac{B}{D\bar{F}_p} \phi_0} - 1 \right) \quad (4)$$

and the resulting collection kernel is

$$\begin{aligned} K &= -\frac{1}{n_\infty} \frac{\partial N}{\partial t} = -\frac{1}{n_\infty} \oint (n B \vec{F} - D\bar{F}_p \nabla n) \cdot d\vec{S} \\ &= \frac{B}{D\bar{F}_p} \oint \vec{F} \cdot d\vec{S} \\ &= \frac{B}{D\bar{F}_p} \phi_0 \quad (5) \\ &= e^{-1} \end{aligned}$$

Now for an arbitrary shaped snow crystal the integral in (5) can be determined thru the use of capacitance c , as has been done in the so-called electrostatic analog theory of crystal growth (e.g., P. 448 of Ref. 1). Since the present case

$$\vec{F} = \vec{F}_e + \vec{F}_{th} + \vec{F}_{df} \quad (6)$$

we immediately have

$$\oint \vec{F}_e \cdot d\vec{S} = 4\pi C q (V_s - V_\infty) = 4\pi Qq \quad (7)$$

since the total collector charge $Q = 4\pi C(V_s - V_\infty)$ where V 's are the electric potentials, and

$$\begin{aligned} \oint \vec{F}_{th} \cdot d\vec{S} &= 4\pi C f_h \frac{12\pi\eta}{5(1 + 3\frac{N_p}{kn})} \frac{r(k + 2.5k\frac{N_p}{kn})k}{(k_p + 2k_a + 5k\frac{N_p}{kn})p} (T_s - T_\infty) \\ &= 4\pi C \bar{F}_n Z_{th} (T_s - T_\infty) \end{aligned} \quad (8)$$

$$\int_{\phi} \vec{H} \cdot d\vec{S} = 4\pi C \bar{f}_h \frac{6\pi \eta_a r(0.74 D M_a)}{(1 + \alpha N_{kn}) M_w \rho_a} (\rho_{v,s} - \rho_{v,\infty})$$

$$= 4 C \bar{f}_h Z_{df} (\rho_{v,s} - \rho_{v,\infty}) \quad (9)$$

The collection kernel is therefore

$$K = - \frac{B}{\frac{B}{D \bar{f}_p} \phi_0 - 1} 4\pi [Qq + C f_h Z_{th} (T_s - T_\infty) + C f_h Z_{df} (\rho_{v,s} - \rho_{v,\infty})]$$

$$= - \frac{4\pi B \phi_0 C}{\frac{B}{D \bar{f}_p} \phi_0 - 1} \quad (10)$$

since the term in the square brackets is exactly $(\phi_0 C)$. Therefore K can be determined if the capacitance C is known.

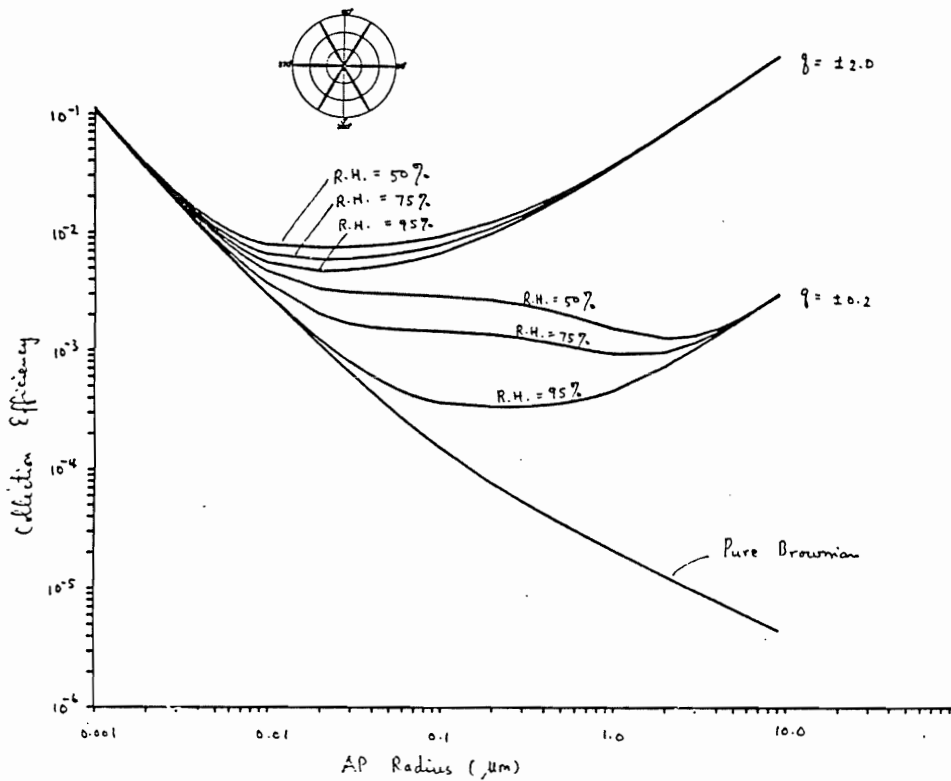


Fig. 1.

Fig. 1 shows an example of the theoretical results. This is the collision efficiency of aerosol particles of various size collected by stellar snow crystal of 0.2 mm diameter. It is seen here that for very small particles the efficiencies converge to one value. This is because the only important collection mechanism here is Brownian diffusion which is only a function of particle size under a fixed environmental condition. Particles of radii 0.01 μm to 1 μm will be subject to strong phoretic and electric force, as seen in the figure. The charges (in e.s.u.) on snow and AP are assumed to be qa^2 and

gr^2 with a and r in cm, respectively. These forces can sometimes increase the collection efficiency to two orders of magnitudes. For particles larger than $1 \mu\text{m}$ the present model may not be adequate because inertial impaction is not taking into account here.

3. EXPERIMENTAL

Thus far only a handful of experimental studies have been carried out to investigate the snow-AP scavenging (Sood and Jackson, 1969, 1970; Magono et al., 1974; Knutson and Stockham, 1974; Knutson et al., 1976; Graedel and Franey, 1976; Vittori and Prodi, 1967; Prodi, 1976; Murakami and Magono, 1983). Their results reveal that aerosol particles are readily captured by ice crystals. While these earlier experiments contributed significantly to our knowledge of snow-AP scavenging, many important factors such as relative humidities and electric charges were not carefully measured. As said before, these factors have significant influence to the magnitude of collection efficiencies. Some of these were field experiments where various effects could not be separated. All these were field experiments where various effects could not be separated. All these would greatly reduce the reliability of the results. The recent experiment by Murakami and Magono (1983) is probably the most careful one in that humidities and electric charges were measured. However they only obtained results for one particular shape. It is known that snow crystal shapes have strong influence on their hydrodynamical and physical properties and hence also their scavenging abilities. It is therefore desirable to have carefully controlled experiments which also take the shape factor into account.

The schematic of the experimental set-up is shown in Fig. 2. The main facilities include the snow charge detector, aerosol chamber, and sample collector. The snow charge

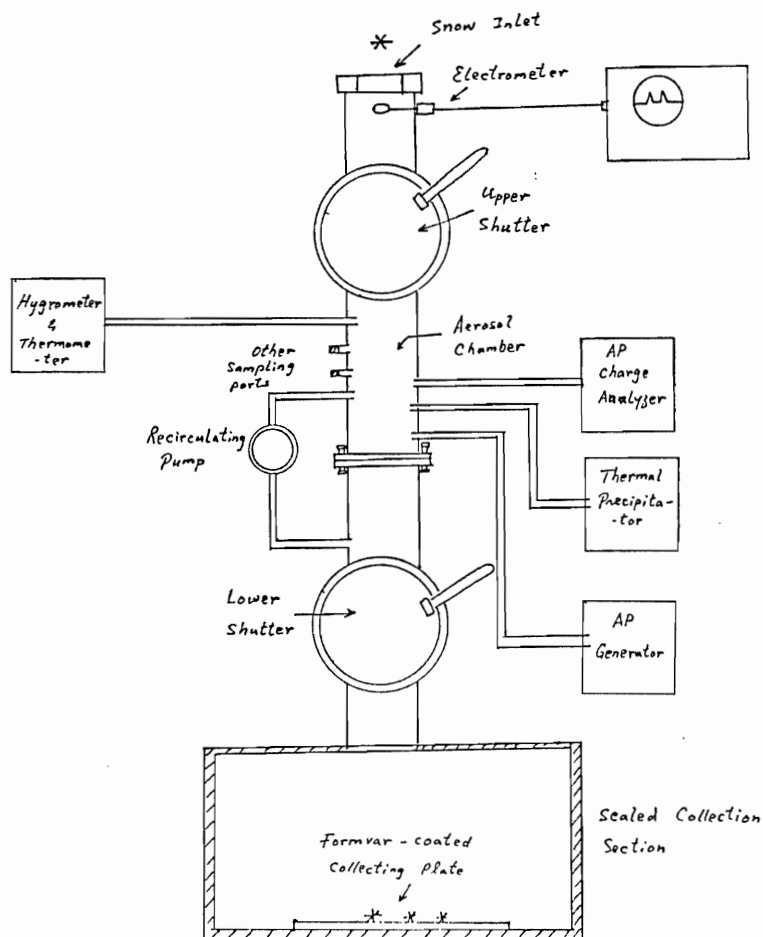


Fig. 2.

detector consists of a metal cylinder serving as a shield of electro-magnetic noises and a ring electrometer. The electrometer is connected to an oscilloscope so that when a snow falls through the ring its charge is measured. The aerosol chamber consists of two shutters at the top and bottom and aerosol inlet and outlets to the aerosol generator and various monitoring instruments, respectively. The shutters can be opened to allow snow crystals to fall through. The monitoring instruments include an aerosol mass monitor, an aerosol charge analyzer, and a thermal precipitator. The collector consists of a plastic closure. Snow samples are collected on a plastic sheet coated with formvar such that the shapes and dimensions of the snow crystals are preserved. The aerosol generator is a LaMer type generator which produces uniform size ($\sim 0.5 \mu\text{m}$ in radius) spherical indium acetate particles. Sizing and counting of particles are done using scanning electron microscope.

Fig. 3 shows an example of the experimental results. A major feature here is that, in general, collection efficiency tends to decrease with snow flake size. This trend seems to exist also in all cases of snow shapes. Fig. 4 shows a comparison of the theoretical results with that of Murakami and Magono (Ref. 9). The agreement between the two is fairly good. We are currently summarizing our experimental results and will compare to the theoretical results in the near future.

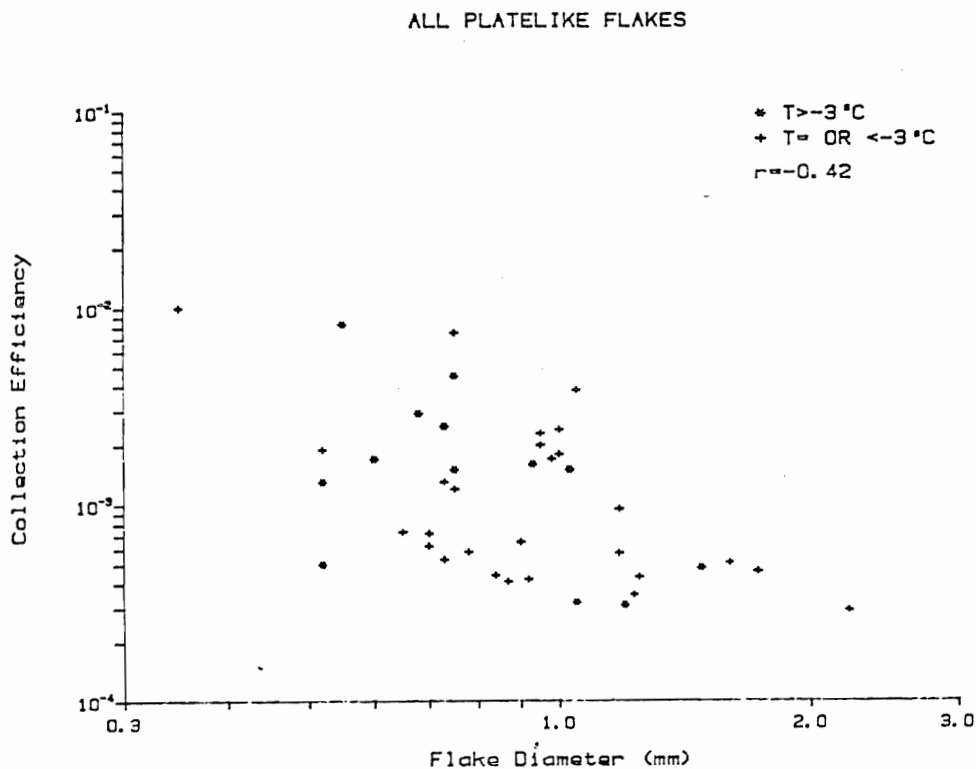


Fig. 3.

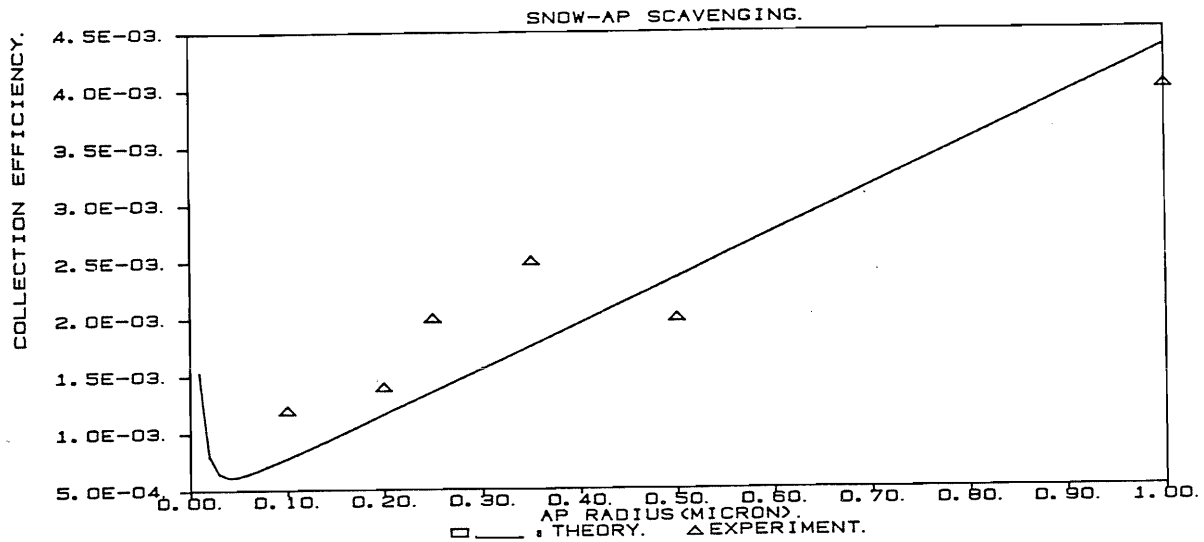


Fig. 4.

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