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The effect of the impaction scavenging efficiency on the wet deposition by a convective warm cloud

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ABSTRACT

To study the influence of the impaction scavenging efficiency in a cloud, we have reevaluated our 2-D dynamical cloud model. This model, which includes spectral microphysics and scavenging was previously applied to a warm precipitating convective cloud at Day 261 (18 September 1974) of the GATE campaign. The original computation used a table for the impaction scavenging efficiency depending on drop radius, particle radius and relative humidity. The re-evaluation used an impaction scavenging efficiency of unity. For both cases the results show general deposition properties that lie within the observed range for marine clouds. Thus, for the simulations of average clouds at a given location the detailed knowledge of the impaction scavenging efficiency is not required. If, however, the calculations are used to simulate the distributions of the pollutant concentration across the rain drop spectrum of a specific cloud, then the detailed knowledge of the impaction scavenging efficiency in addition to the detailed knowledge of the vertical distribution of the pollutants are essential since they determine the deviation from the well mixed spectral concentration distribution produced inside the cloud through collision and coalescence processes.

1. Introduction

Wet removal of aerosol particles by precipitation plays an important role in the distribution and concentration of pollutants in the atmosphere. In warm clouds, aerosol particles enter the drops via two different mechanisms. On the one hand, aerosol particles are taken up by cloud drops while serving as cloud condensation nuclei (CCN). This process, called nucleation scavenging, is confined to supersaturated regions of the cloud and is determined by the size and chemical composition of the particles (Pruppacher and Klett, 1978). On the other hand, aerosol particles are scavenged by colliding with already existing cloud drops through the mechanisms of Brownian diffusion, inertial impaction, hydrodynamic forces and through phoretic and electric effects. The efficiency with which impaction scavenging operates has been discussed on the basis of theoretical studies and laboratory tests by Barlow and Latham (1983), Leong et al. (1982), Carstens and Martin (1982), Wang et al. (1978), Lai et al. (1978), Grover et al. (1977), Wang and Pruppacher (1977), Kerker and Hampl (1974) and Hampl et al. (1971), among others. It is a function of the sizes of the aerosol particle and the drop, the relative humidity and electrical charges.

To include these dependencies in a numerical simulation, a model needs to treat explicitly the spectra of aerosol particles and drops. Most models, however, include the cloud constituents and pollutants only in a highly parameterized form which results in a variety of different scavenging rates. For example, Ahr et al. (1989) investigated in a warm cloud deviations from the nucleation scavenging by assuming the relation $N_{\mathrm{CCN}} = cS^k$ (with S being the supersaturation in the cloud and c, k constant) for the number of CCN, instead of explicitly following the aerosol particle spectrum and the resulting CCN.

In the present paper we investigate how a parameterized impaction scavenging efficiency influences the scavenging behavior of a warm cloud.

In current scavenging models, numerous different parameterizations for the impaction scavenging efficiency are used. For example, Chaumerliac et al. (1986, 1987) used the formula for the impaction scavenging efficiency according to Dana and Hales (1976), Rutledge et al. (1986) used a constant efficiency between 0.042 and 0.052 according to Radke et al. (1980) while some models, e.g., Walcek and Taylor (1986) completely neglect impaction of aerosol particles by drops.

These models, among others, are used to estimate the overall scavenging of aerosol particles and gases, and subsequent wet deposition, to determine the acidification of precipitation and the general processing of pollutants by storms. In addition to these more general investigations, scavenging models nowadays are also challenged to explain isolated phenomena, such as observed concentrations of pollutants in individual raindrops (e.g., Bächmann et al., 1991, 1992).

2. The present study

In an attempt to shed light on the influence of different impaction efficiencies on the scavenging behavior of a warm cloud we have used our detailed scavenging simulation as described in Flossmann (1991) and redone the calculations with an impaction efficiency of unity. In Flossmann (1991) our 2-D dynamic model (Clark, 1977, 1979; Clark and Gall, 1982; Clark and Farley, 1984) including spectral microphysics and scavenging (Flossmann, 1987; Flossmann et al., 1985) was evaluated for a warm precipitating convective cloud at Day 261 (18 September 1974) of the GATE campaign. Two different chemical species ($(NH_4)_2SO_4$ and NaCl) of aerosol particles were followed in the air, inside the drops in the cloud, and inside the drops reaching the ground. Each drop was allowed to pick up aerosol particles by

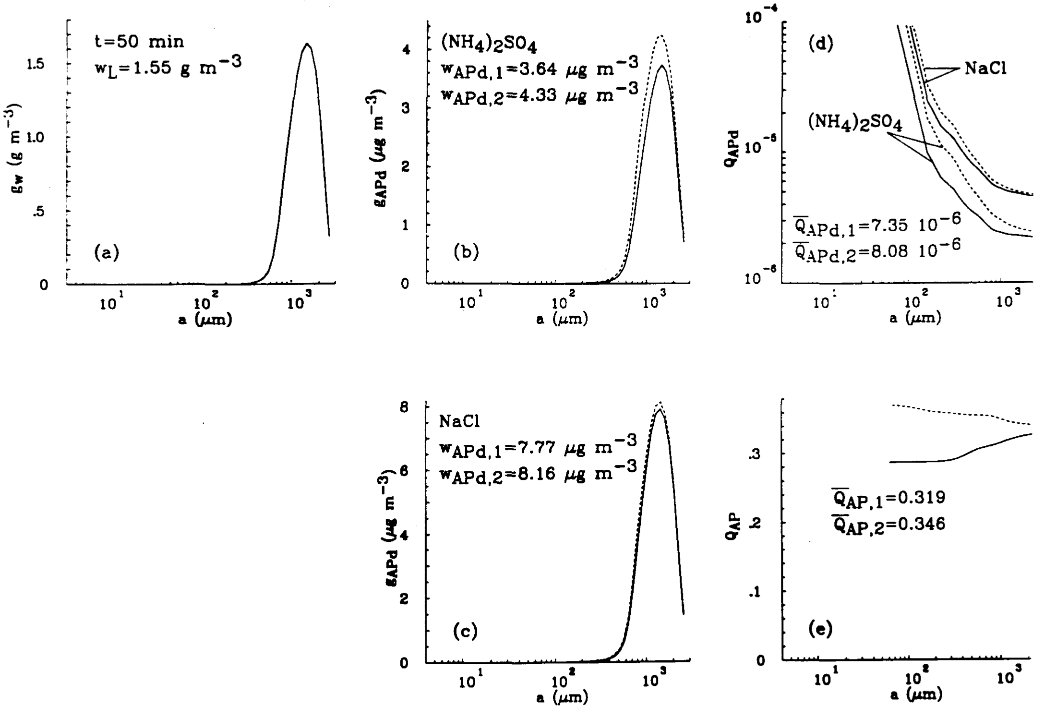

Fig. 1. Water mass density distribution function $g_w(m)$ (a), mass density distribution function for $(NH_4)_2SO_4$ (b) and NaCl (c) in the drops $g_{APd}(m)$, mixing ratio of $(NH_4)_2SO_4$ and NaCl in the drops Q_{APd} (d), and the fraction of $(NH_4)_2SO_4$ compared to the total scavenged aerosol material Q_{AP} (e) at model time $t = 50$ min at $x = 13.4$ km at the earth's surface. Solid curve: results with impaction efficiency table (Case 1), dashed curve: results with $E = 1$ (Case 2). (The density distribution functions are displayed per logarithmic radius interval.)

impaction scavenging with an efficiency as given by Grover et al. (1977) and Wang et al. (1978) in a compilation of values given by Flossmann (1987). Electrical charges, however, were disregared. To estimate the maximum amount of impaction scavenging possible for this specific simulation we reran the calculations with an impaction efficiency of unity.

3. Results

The dynamical and microphysical development of the cloud is the same for the case of the impaction scavenging efficiency being unity (Case 2) as for the results with the complete impaction scavenging efficiency table (Case 1). Case 1 results are given in Flossmann (1991) and thus shall not be repeated here. However, the scavenging properties of the cloud have changed as can be seen in Fig. 1.

Fig. 1 displays the cloud drop and pollution spectra at the earth's surface at 50 min of model time, when the maximum amount of precipitation passed over this specific location (see Fig. 2). In Fig. 1a we see that the liquid water spectrum $g_w(m)$ has its maximum in the range of the millimeter sized drops. The mass density distribution function of aerosol particles in the drops g_{APd} for $(NH_4)_2SO_4$ (b) and NaCl (c) display a quite similar behavior. It was already noted by Flossmann et al. (1985) that the main scavenged pollutant mass follows the main water mass. The dashed curves in these figures correspond to

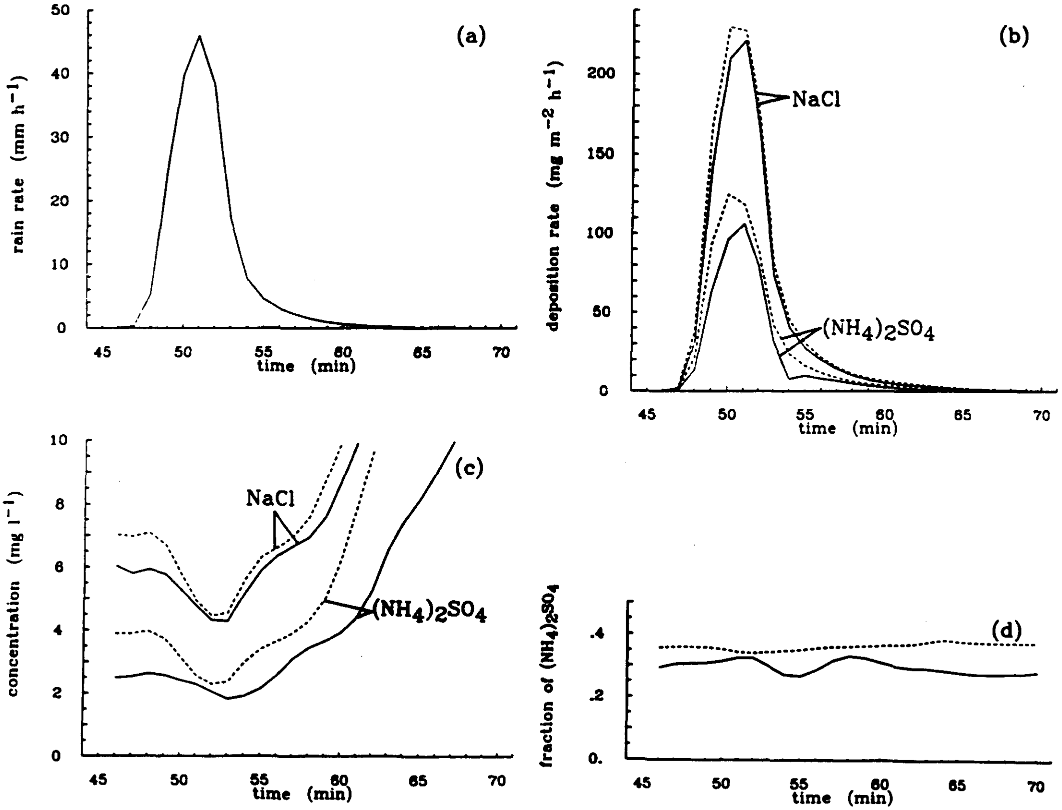

Fig. 2. Evolution of the rainfall rate (a), the amount of material deposited by the rain (b), the concentration of scavenged material in the rain (c), and the fraction of $(NH_4)_2SO_4$ in the total deposited material (d) at 13.4 km averaged over 1-min intervals. Solid curve: results with impaction efficiency table (Case 1); dashed curve: results with $E = 1$ (Case 2).

Case 2 with an impaction scavenging efficiency $E = 1$. Here it becomes obvious that the amounts of $(NH_4)_2SO_4$ and NaCl deposited at the surface are larger for Case 2 than for Case 1. The amount of deposited $(NH_4)_2SO_4$ has increased even more than the amount of scavenged NaCl.

Figs. (d) and (e) display some information on the mixing ratio of pollutant mass in the drops. Fig. 1d shows the mixing ratio Q_{APd} of $(NH_4)_2SO_4$ and NaCl (mass of pollutant/total drop mass) for the two cases considered. We note that the pollutant mixing ratio in the precipitation sized drops no longer displays the well mixed character which we found inside the cloud (see Fig. 8; Flossmann, 1991). Instead, the mixing ratio in the rain increases with decreasing drop radius. This is caused by an increase in concentration through evaporation, as well as the shorter exposure time to below-cloud scavenging for the larger drops due to their higher terminal velocity. This general behavior of the pollutant mixing ratio in rain drops has already been observed e.g., by Georgii and Wötzel (1970) and Bächmann et al. (1991, 1992).

We also note a dependency of the mixing ratio on the chemical species considered. With $E = 1$, the amount of $(NH_4)_2SO_4$ increased more than the amount of NaCl as was already illustrated in Fig. 1b and c by the mass density distribution functions g_{APd}. This increase becomes smaller the larger the drops.

This is also reflected in Fig. 1e which displays the fraction of $(NH_4)_2SO_4$ in the total scavenged material. For drops larger than 100 μm the fraction of $(NH_4)_2SO_4$ for Case 1 no longer has the constant value of around 34% as was found inside the cloud by Flossmann (1991) (compare their Fig. 8), but ranges between 28% and 33%. This reduction in the fraction of $(NH_4)_2SO_4$ between cloud base and the ground is caused by the dominant impaction of NaCl, which is present in the atmosphere below cloud base in a larger mass concentration than $(NH_4)_2SO_4$ (see Fig. 2 and Table 1; Flossmann, 1991) and in a particle size range captured very efficiently through inertial impaction (Grover et al., 1977). For drops larger than 500 μm, however, the impaction scavenging efficiency was found to decrease (Wang and Pruppacher, 1977). Consequently, less NaCl was captured, which resulted in the displayed increase in the fraction of $(NH_4)_2SO_4$ across the drop spectrum. This is no longer valid for Case 2 with the constant impaction scavenging efficiency of unity. There, the overall level of $(NH_4)_2SO_4$ is higher than for Case 1 and the fraction of $(NH_4)_2SO_4$ is almost constant with a slight decrease with increasing drop radius from 37% to 34%. This results from the fact that $(NH_4)_2SO_4$, whose main mass is located in the particle sizes in the so-called greenfield gap (Fig. 2; Flossmann, 1991), is now captured very efficiently. The contribution of $(NH_4)_2SO_4$ and NaCl scavenged below cloud base, however, decreases as the large drops experience a shorter exposure time due to their terminal velocity. Consequently, both curves approach the in-cloud scavenging fraction of around 34%.

To gain a better impression of the overall scavenging behavior of the cloud we have displayed in Fig. 2 the evolution of the rainfall rate (a), the amount of material deposited by the rain (b), the concentration of scavenged material in the rain (c) and the fraction of $(NH_4)_2SO_4$ in the total deposited material (d) at 13.4 km averaged over 1 min intervals for Case 1 and Case 2. As already mentioned, the amount of deposited material is larger for Case 2, with a stronger increase for $(NH_4)_2SO_4$ than NaCl. The same information is reflected in the scavenging efficiencies which are defined, analogously to Flossmann (1991), as:

$$
\begin{aligned}
E_{\mathrm{p}} &= \frac{\text{cumulative rain mass on the ground}}{\begin{array}{c}\text{cumulative water mass converted}\\ \text{from the vapor}\end{array}},\\
E_{\mathrm{n}} &= \frac{\text{cumulative nucleation scavenging}}{\begin{array}{c}\text{cumulative aerosol mass scavenged}\\ \text{by nucleation and impaction}\end{array}},\\
E_{\mathrm{t}} &= \frac{\begin{array}{c}\text{cumulative aerosol particle}\\ \text{mass in rain}\end{array}}{\begin{array}{c}\text{cumulative aerosol mass scavenged}\\ \text{by nucleation and impaction}\end{array}},\\
E_{\mathrm{i}} &= \frac{\text{cumulative impaction scavenging}}{\begin{array}{c}\text{cumulative aerosol particle}\\ \text{mass in rain}\end{array}}.
\end{aligned}
\tag{1}
$$

The values for the precipitation and scavenging efficiencies for Case 1 and Case 2 are displayed in Table 1. For Case 2 the enhanced influence of

Table 1. *Precipitation and scavenging efficiencies for the different species of aerosol particles considered as defined by eq. (1). First column: results with impaction efficiency table (Case* 1); *second column: results with* $E = 1$ *(Case 2)*

		Case 1	Case 2
E_p		46%	46%
E_n	$(NH_4)_2SO_4$	93%	77%
	NaCl	85%	79%
	total	87%	78%
E_t	$(NH_4)_2SO_4$	34%	46%
	NaCl	43%	48%
	total	40%	47%
E_i	$(NH_4)_2SO_4$	22%	51%
	NaCl	36%	45%
	total	32%	47%

impaction scavenging contributes 47% to the total aerosol material deposited on the groud, as compared to 32% for Case 1 (E_i). The overall deposited material on the ground for Case 2 by this specific cloud has increased for $(NH_4)_2SO_4$ by 40% and for NaCl by 11% as compared to the values reported in Table 4 of Flossmann (1991).

4. Summary and conclusions

By changing the impaction scavenging efficiencies from best available estimates (Flossmann, 1991) to $E = 1$, the wet deposition increased by up to 50%. Nevertheless, the resulting concentrations of sulfate and chloride in the rainwater (Fig. 2c) still lie within the range of values reported by Church et al. (1982), Galloway et al. (1982, 1983), Varhelyi and Gravenhorst (1983), and Parungo et al. (1986) for marine precipitation. This is due to the fact that the marine atmosphere and the evolving clouds experience quite a wide range of variations and no deposition values have thus far been reported from just one thoroughly investigated cloud. Thus, we conclude that for general scavenging simulations of an average cloud, detailed consideration of the impaction scavenging efficiency is not of vital importance.

Also the general conclusion of Flossmann (1991) that the scavenging efficiency of a convective warm cloud is closely related to its precipitation efficiency still holds for both cases (see Table 1).

If, however, scavenging calculations are used to simulate observed distributions of pollutant concentrations in surface-level drop spectra (e.g., by Bächmann et al., 1991, 1992), then detailed knowledge of the impaction scavenging efficiency is important. Below-cloud scavenging leads to a distribution of the pollutant concentration across the drops spectrum which differs from the well mixed state the large drops experienced inside the cloud through the collision and coalescence process (see Fig. 1). Below-cloud scavenging is significantly determined by the impaction scavenging efficiency and the vertical distribution of the pollutant mass. Furthermore, it is influenced by processes not considered here as, e.g., electrical effects and scavenging of gases which have to be considered if one attempts to realistically reproduce observed pollutant concentrations in rain drops.

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