Numerical Study on Impacts of Multi-Component Aerosols on Marine Cloud Microphysical Properties

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Abstract

In this paper impacts of nss-sulfate, sea-salt and organic particles on microphysical properties of marine cloud are investigated, using a multi-component size-resolving aerosol model. Numerical results show that the number and type of cloud condensation nuclei (CCN) depend on particle physical and chemical properties (size distribution, chemical composition) of particles, and on environmental conditions (updrafts or supersaturation). Sea-salt particles play a critical role in cloud microphysical processes. Due to its large radius, sea-salt particles are activated into cloud drops in the initial cloud development. Sea-salt activation decreases supersaturation by consuming water vapor and suppresses nss-sulfate activation. Nss-sulfate indirect forcing may be overestimated in some conditions (such as updraft is low), because of the presence of sea-salt particles. Soluble organic components decrease maximum supersaturation, and lead to a decrease of cloud drops activated at the case of a high nss-sulfate and a high updraft velocity. Nss-sulfate CCN account for most variations of the cloud optical depth (COD). Sea-salt increases COD in the case of low nss-sulfate, but decreases COD when nss-sulfate concentration is high. The organic component enhances this influence of sea-salt on COD.

1. Introduction

The indirect climatic effect of aerosol refers to the influence of aerosols on cloud optical depth and albedo, and cloud lifetime, as a result of anthropogenic changes in the number

and composition of cloud condensation nuclei (CCN). One of the shortcomings in attempting to quantify indirect radiative forcing, particularly by sulfate aerosols, has been the neglect of other sources of CCN, particularly sea salt nuclei and organic aerosols over the oceans (IPCC 2001).

Sea salt aerosols may be the dominant contributor to both light scattering and cloud nuclei in those regions of the marine atmosphere where wind speeds are high, and/or other aero-

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sol sources are weak (O'Dowd et al. 1997; Murphy et al. 1998; Quinn et al. 1998). They are very efficient CCN, and therefore characterization of their surface production is of major importance for aerosol indirect effects. Ghan (1998) studied the competition between sea-salt and sulfate particles as CCN. Measurements and modeling studies (O'Dowd et al. 1997) illustrated that the effect on cloud droplet concentration is not only to reduce the relative contribution of sulfate aerosols to activated CCN, but also to reduce the droplet concentration. Recently the role of organic aerosols has been watched with much attention. Novakov and Penner (1993) indicated that 37% of CCN number concentration, measured at a marine site, could be accounted for by total sulfates and the remaining 63% by organic aerosols. Measurements by Hegg et al. (1995) also showed that most CCN over the northeastern Atlantic coast of the United States were not sulfates. Saxena et al. (1995) found that organics could enhance water absorption by inorganic, and pointed out the potential importance of organic compounds in influencing the CCN properties (Saxena and Hildemann 1996). Matsumoto et al. (1997) studied the contribution of particulate sulfates and organic carbon to CCN in the marine atmosphere. They found that not only sulfates, but also water-soluble organic salts were dominant in fine aerosols. Rosenfeld et al. (2002) also investigated the role of sea spray in precipitation processes. They concluded that precipitation from similar polluted clouds over oceans is much less affected because large size of sea salt nuclei overrides the precipitation suppression effect of the large number of small pollution nuclei.

So large-scale models must be able to represent several factors related to CCN in order to assess the indirect effect more properly: the mass size distribution of water-soluble species, the degree of solubility of the represented species, and the amount of mixing of individual species within a given size fraction (IPCC 2001). It is clear that the simple representation of sulfate aerosols in global and regional models has become insufficient to account for important chemical and climate effects. The most critical species are sulfates, organics, sea salt, nitrates and probably other types of particles.

The abilities of these species to serve as CCN are relatively well known, whereas our understanding of the ability of organic species to act as CCN is relatively poor. This is a critical area of uncertainty for the global-scale modeling of cloud droplet nucleation (IPCC 2001). The water-soluble fraction of organic species in aerosols can be high relative to sulfate concentration in aerosols (Zappoli et al. 1999), and organics may be important sources of CCN in at least some circumstances (Novakov and Penner 1993; Rivera-Carpio et al. 1996).

In this paper we use a size-resolving model to study the microphysical interactions between nss-sulfate, sea-salt, organic components, and cloud drops. The influence of sea-salt and organic components on aerosol activation processes, resultant number concentration of cloud droplet, and cloud optical depth are also investigated.

2. Model description

A spatially homogeneous aerosol undergoing nucleation, condensation and coagulation is simulated by numerical solution of the general dynamic equation (GDE). A particularly powerful technique for solving GDE is the so-called sectional method or bin model. In this discrete approach to modeling aerosol dynamics, the aerosol size range is divided into a number of sections. Approximating the mass, number or surface area distribution function as a constant within each section results in a set of differential equations for the properties of each section. Simulation of particle dynamics thus takes the form of adding new particles to the appropriate bins, removing them, and transferring particles from bin to bin. For multicomponent particles, each of these bins will also be characterized by the total mass of each species within the bin. The size-resolved aerosol model used in this paper was originally developed by Zhao (1996, 1998) to study aerosol size formation and evolution processes in the remote marine boundary layer. Here, only a general description of the overall characteristics of the model is given. Aerosol and cloud microphysical processes included in this model are depicted in Fig. 1. These processes are simply described in the following paragraphs. The particle size distributions for aerosols and droplets are each divided into 100 bins with geometrically increas-

Fig. 1. Modeled aerosol and cloud processes.

ing size starting from $0.005 \mu\text{m}$ of radius, such that the particle volume doubles between successive bins. Four aerosol types are calculated as nss-sulfate, sea-salt aerosol, mixed aerosol (sulfate, sea-salt, and organic) and water droplets. As depicted in Fig. 1, in this model there are four microphysical processes considered: activation, evaporation, condensation and coagulation. In this model two bin structures are dealt with: one is for dry particles which do not experience condensational growth and one for water droplets which experience not only coagulation but also condensation/evaporation. After activation, dry particles will become water droplets and then experience condensational growth process. Within each droplet size bin, the model solves a continuity equation for the total volume concentration of dissolved cloud condensation nuclei to allow calculation of equilibrium reduction in droplet vapor pressure (the solute effect).

In this model supersaturation is explicitly calculated by water vapor concentration and temperature at each time step. This explicit treatment of supersaturation requires a small time step to avoid numerical instabilities. Here a variable time step method is used. The aerosol particles in a size bin are activated to CCN as soon as supersaturation exceeds their critical supersaturation. The maximum value of supersaturation for which a droplet can maintain stable equilibrium, with respect to con-

densation, is determined by a simplified form of the Kohler equilibrium relations (Pruppacher and Klett 1997):

$$\frac{e_a}{e_{sat,w}} = \exp \left[\frac{2M_w \sigma_{s/a}}{RT \rho_w a} - \frac{\nu \Phi_s \epsilon_v M_w \rho_s r_N^3}{M_s \rho_w (a^3 - r_N^3)} \right] \quad (1)$$

where e_a is water vapor pressure in environment at droplet surface, $e_{sat,w}$ saturation vapor pressure, R gas constant, T temperature, M_w molecular weight of water, M_s molecular weight of salt, $\sigma_{s/a}$ surface tension of solution against air, ρ_w density of water, a radius of droplet, ν number of ions into which a salt molecule dissociates in water, ϵ_v is the volume fraction of salt mass in a particle of radius r_N . Φ_s osmotic coefficient, and ρ_s is the density of aqueous salt solution.

The numerical approach for condensation/evaporation is the same as in the work of Zhao (1996, 1998) and only brief description of this process is made here. Condensational growth is one of the most important processes for cloud droplets in stratus clouds after CCN activation. The rate of growth of cloud droplets is determined by considering the simultaneous quasi-steady diffusion transport of mass, and latent heat from a falling water droplet. Once cloud nuclei are activated their further growth is governed by the droplet condensational growth equation, which is taken in the form similar to that employed by (Pruppacher and Klett 1997):

$$\frac{dm}{dt} = \frac{A_1 * e_s * (S + 1 - A_3)}{1 + A_1 * A_2 * e_s} \quad (2)$$

$$A_1 = \frac{4\pi r * D * f_v * M_w}{B * A_v * T} \quad (3)$$

$$A_2 = \frac{M_w L^2}{4\pi r f_t T_c R T^2} \quad (4)$$

$$A_3 = \exp\left(\frac{2M_w \sigma_s}{r R T \rho} - \frac{\nu M_s M_w}{S_w M_c}\right) \quad (5)$$

where S is supersaturation which is calculated by water vapor concentration and temperature at each time step, m is the particle mass concentration, R is the gas constant, ρ is water density, r is the particle radius, f_v and f_t are ventilation factors, D and T_c is diffusion coefficient and thermal conductivity modified for free molecular limit, M_w is the molecular weight of water, e_s is saturation vapor pressure, σ_s is the surface tension of water-air interface, A_1 and A_2 are growth kernels, A_3 represents Kelvin effects and solution effects, ν is the number of ions solute dissociates into a particle, M_s is molecular weight of solute, M_c is mass concentration of condensed water in a particle.

Coagulation and coalescence may be driven by any process that is able to force two or more particles together, whether Brownian diffusion, gravitational settling, shear flow or any of a number of other mechanisms. In the atmosphere Brownian coagulation tends to dominate under most circumstances, and sedimentation driven forms is generally significant only for particles greater than a few microns in radius. In this model both Brownian coagulation and gravitational collection are taken into account by the coagulation kernels, the same as Zhao (1996, 1998). The numerical treatment of coagulation is based on the method of Zhao (1998).

In order to avoid numerical instabilities a variable time step scheme is used in this model. The relative change of particle concentration and supersaturation is calculated and is used as criterions to modify the time step. The time step will be halved if particle concentration or supersaturation changed larger than that model allowed, and will be doubled if they changed less than that model allowed.

Marine stratus cases under different initialization of nss-sulfate, sea-salt, and organic com-

Table 1 Sea-salt concentration under different wind speeds

Wind speed (ms ⁻¹)	Film drop mode (cm ⁻³)	Jet drop mode (cm ⁻³)	Spume drop mode (cm ⁻³)
2	3.0	0.63	2.1×10^{-6}
5	5.7	0.84	3.4×10^{-6}
10	17.1	1.36	7.6×10^{-6}
15	51.1	2.21	1.7×10^{-6}

ponents are simulated using this model. All calculations showed in this paper are made at temperature 280 K, atmospheric pressure 880 hPa and relative humidity 100% representing the marine stratiform cloud base. The updraft used in each case is assumed constant. According to the measurements of O'Dowd et al. (1993), nss-sulfate concentration can be represented by a lognormal distribution, with mean radius 0.08 μm and standard deviation 1.4. Sea-salt can be described using three lognormal distributions (O'Dowd et al. 1997) as fine mode, jet mode and spume mode. Mean radius and standard deviation is 0.1 μm and 1.9 for film drop mode, 1 μm and 2 for jet drop mode, and 6 μm and 3 for spume drop mode, respectively. Sea-salt concentrations under different wind speed are listed in Table 1.

3. Influence of sea salt particles on marine cloud microproperties

Recently the role of sea-salt in marine cloud microphysics is studied (O'Dowd et al. 1997a,b; D. Rosenfeld et al. 2002). The impact of supplementing the nss-sulfate CCN population with sea salt CCN illustrates that the effect on cloud droplet concentration is not only to reduce the relative contribution of sulfate aerosols to activated CCN, but also to reduce the droplet concentration (O'Dowd et al. 1997a,b). Rosenfeld et al. (2002) also investigated the role of sea spray in precipitation processes. Here a number of simulations examined the influence of sea salt particles on marine cloud properties.

Figure 2 shows the influence of sea-salt on maximum supersaturation under different nss-sulfate and updraft velocities. Panel a, b, and c of Fig. 2 are the model results under three

Fig. 2. Influence of sea-salt on maximum supersaturation under different updraft velocities.
(a) sulfate 100 cm^{-3} , (b) sulfate 300 cm^{-3} , and (c) sulfate 1000 cm^{-3} .

Table 2 Cloud top microphysical properties of different initializations. (updraft velocity is 0.2 ms^{-1})

Initialization		Max. supersaturation (%)	Effective radius (μm)	Cloud drops concen. (cm^{-3})	Standard deviation
nss-sulfate (cm^{-3})	Wind speed (ms^{-1})				
100	0	0.482	16.913	93	1.116
	2	0.478	16.729	93	1.121
	5	0.468	15.886	101	1.120
	10	0.442	15.117	114	1.123
	15	0.388	14.751	147	1.129
300	0	0.358	11.800	287	1.116
	2	0.357	11.722	293	1.212
	5	0.352	11.630	296	1.120
	10	0.339	11.531	304	1.154
	15	0.314	11.273	334	1.237
1000	0	0.253	7.533	969	2.080
	2	0.252	7.515	957	2.047
	5	0.250	7.479	954	2.098
	10	0.242	7.440	940	2.093
	15	0.226	7.180	938	2.402
5000	0	0.122	3.525	3630	3.016
	2	0.120	3.540	3626	2.980
	5	0.120	3.613	3526	2.984
	10	0.117	3.713	3341	2.835
	15	0.110	3.773	3251	2.935

different nss-sulfate concentration: 100 cm^{-3} for clean case, 300 cm^{-3} for typical case and 1000 cm^{-3} for polluted case of marine atmosphere. The maximum supersaturation increases as updraft velocity increases and decreases when nss-sulfate or sea-salt particles increase. The presence of sea-salt tends to enhance the water vapor consumption for nss-sulfate particles activated as CCN. Because of its larger size of sea-salt particle than that of nss-sulfate particle, sea-salt particles are more likely to be preferentially activated than nss-sulfate particles. Thus increasing the sea-salt particles enhances the supersaturation decreasing.

Table 2 shows the calculated cloud top microphysical properties of total 20 runs with different initializations. Initialization of nss-sulfate is set to 100, 300, 1000 and 5000 cm^{-3} representing clean, typical, polluted and extremely polluted case respectively. From Table 2 we can see that increasing nss-sulfate increases cloud droplet number concentration and reduces the cloud droplet radius. The influence of sea-salt is a little complicated. For clean and typical conditions, increasing sea-salt particle increases cloud droplet number concentration. But for other conditions there is a decrease of cloud drops number concentration, due to increasing sea-salt.

Fig. 3. Relationship between activated particles and updraft velocity. Nss-sulfate concentration is 100, 300 and 1000 cm^{-3} respectively. Sea-salt concentration is that under horizontal wind speed of 2, 5, 10 and 15 ms^{-1} .

The relationship between various updraft velocities and particle number concentration activated at different horizontal wind speed is depicted in Fig. 3. It can be seen from this figure that an increasing of sea-salt leads to an increase of particle number concentration activated when updrafts are strong (Region A in Fig. 3). Interestingly, in region B of Fig. 3 where updraft velocity is weak there is an opposite trend, that an increasing of seasalt leads to a decrease of activated particles. The maximum supersaturation is low when vertical velocity is low as shown in Fig. 2. Sea-salt particles would be activated firstly due to their relative larger size. Activated sea-salt particles consume the water vapor and reduce the supersaturation. Thus total activated particle number concentration would decrease with increasing of sea-salt particles for high nss-sulfate level and weak updraft velocity. The results suggest that the presence of sea-salt may offset the effects of nss-sulfate as CCN in some circumstances. The indirect forcing of nss-sulfate is somewhat overestimated because of the presence of sea-salt for some certain conditions.

4. Role of soluble organic components

Cadle and Groblicki (1982) found about 40% organic in their samples are water soluble. Zappoli et al. (1999) showed 50% of water solu-

Fig. 4. CCN spectrum for different nss-sulfate and soluble organic components.

ble mass in fine particles are organic and over 80% of organic matters are water soluble for background concentrations. Mircea et al. (2002) show that the increase of CCN number concentration in the presence of water soluble organic components varies with aerosol type, with an increase up to 13% in the marine case, up to 97% in the rural case, and up to 110% in the urban case, for the supersaturation range of typical atmospheric conditions.

The water-soluble organic compounds influence the critical supersaturation in two ways: directly by increasing the soluble mass in Raoult term and indirectly by decreasing the cloud drop surface tension in Kelvin term. This leads to a decrease of the critical supersaturation. According to observations of Fachini et al. (1999) and Zappoli et al. (1999), the organic components used in our calculations were a dicarboxylic acid and a fulvic acid. The mass of aerosol organic components is 20% and 45% of that of nss-sulfate. Figure 4 shows the CCN spectrum for different nss-sulfate and soluble organic components. We can see that for the case of nss-sulfate 5000 cm^{-3} increasing organic components may increase CCN number concentration when supersaturation is relative low. The soluble organic components have little effects on CCN number concentration when supersaturation is high or nss-sulfate is low. The measurement study of Matsumoto et al. (1997) also showed the soluble organic salts were relatively more predominant than nss-sulfate and made a significant contribution to CCN number

concentration under the lower sulfate conditions.

In order to further investigate the role of organic components in presence of sea-salt particles, we assume updraft is 0.2 ms^{-1} which represents the ordinary stratiform cloud conditions over the marine areas. Wind speed is 15 ms^{-1} and water soluble organic mass is 45% of the total condensation nuclei mass. Figure 5 shows that cloud droplets activated at different nss-sulfate for various initializations. Solid line is case with nss-sulfate only, dash line the case with sea-salt, and dotted line the case with sea-salt and organic component. The presence of sea-salt particles decrease the activated particle number and the organic components enhance this influence at high nss-sulfate. This suggests that it is necessary to consider the effects of various aerosol types in some polluted air mass with a high nss-sulfate. Otherwise the effects of nss-sulfate will be overestimated.

Water soluble organic enhances the competition between sea-salt and nss-sulfate as CCN. When nss-sulfate concentration is high and updraft is low, the density of cloud droplets activated is lower than nss-sulfate concentrations. In presence of sea-salt and organic components, the number of cloud droplets activated is lower further. The impacts of nss-sulfate on cloud microphysical properties will be overestimated if sea-salt and organic components are not taken into account.

Fig. 5. Cloud drops activated at different nss-sulfate for various initialization. Solid line is case with nss-sulfate only, dash line the case with sea-salt, and dotted line the case with sea-salt and organic component.

5. Influence of multicomponent aerosols on cloud optical depth

Cloud optical depth (COD) is often expressed in the following equation:

$$COD = \frac{3}{2} \frac{W}{r_e} D_c \quad (6)$$

where r_e is the effective radius, W liquid water path, and D_c cloud thickness.

Fig. 6. Cloud optical depth at different nss-sulfate. a) sea-salt, b) organic components, and c) sea-salt and organic components.

Impacts of sea-salt concentrations on cloud optical depth are depicted in Fig. 6a. COD increases obviously as nss-sulfate increases. The role of sea-salt is different if the nss-sulfate concentration is changed. In case of high nss-sulfate concentration an increase of sea-salt decreases the COD whereas it increases COD when nss-sulfate is low. From Fig. 6b, it can be seen that organic components have a little influence on COD. But it is interesting that the role of organic components is different under different nss-sulfate concentrations with presence of sea-salt particles. Organic components enhance the influence of sea-salt on COD (Fig. 6c). For case of low nss-sulfate (means clean case) sea-salt and organic components increase COD and for case of high nss-sulfate (means polluted case) an increase of sea-salt and organic decrease COD.

On the challenge of quantifying the relationship between changes in aerosol number and composition and cloud properties, there are few studies demonstrating clearly this relationship. Recently Higurashi and Nakajima (2002) showed the distribution of four major aerosols types (soil dust and carbonaceous, sulfate, and sea-salt aerosols) in the East China Sea region using SeaWiFS data. Satellite data, field measurement data and numerical modeling can be used for revealing and representing indirect climatic effects of aerosols.

6. Summary and conclusions

In this study impacts of nss-sulfate, sea-salt and organic particles on microphysical properties of marine clouds are investigated using a multi-component size-resolving aerosol model. The multi-component size-resolved aerosol-cloud model is a powerful tool to study chemistry-aerosol-cloud interactions because it allows multiple aerosol types to compete for existing water vapor. Numerical results show that the number and type of CCN depend on physical and chemical properties of particle (size distribution, chemical composition) and on environmental conditions (updrafts or supersaturation).

Sea-salt plays a critical role in cloud microphysical processes. Due to its large radius sea-salt particles are activated into cloud droplets in the initial stage of cloud development. Sea-salt activation decreases supersaturation by

consuming water vapor and suppresses nss-sulfate activation. Nss-sulfate indirect forcing may be overestimated in some conditions (such as updraft is low) because of the presence of sea-salt particles. Soluble organic components decrease maximum supersaturation and lead to a decrease of cloud droplets activated at the case of a high nss-sulfate and a high updraft velocity. Nss-sulfate can account for most cases of COD. Sea-salt increases COD in case of low nss-sulfate but decreases COD when nss-sulfate concentration is high. Organic component enhances this influence of sea-salt on COD.

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