NOTES AND CORRESPONDENCE

Comments on “Numerical Study on Impacts of Multi-Component Aerosols on Marine Cloud Microphysical Properties” by Zhao et al. (J. Meteor. Soc. Japan, Vol. 83, 977–986)

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1. Introduction

Many studies have used parcel models to identify the effects of aerosols on cloud microstructure (Mordy 1959; Fitzgerald 1974; Takeda and Kuba 1982; Kuba and Takeda 1983; Chen and Lamb 1999; Feingold et al. 1999; Kuba et al. 2003; Kuba and Fujiyoshi 2006). A recent paper (Zhao et al. 2005, henceforth Z05), also used a parcel model. However, the following essential information for readers is missing in Z05; cloud thickness, information of soluble organic components, information of bin method (one-moment or two-moment, one-dimension or two-dimension or more, technique of calculation), and starting point of calculation (near the cloud base or far from the cloud base). Additionally, results from their study contradict those from the previous papers as follows:

- (1) According to Z05 (Table 2), an increase in the number of sea-salt particles causes a decrease in both cloud drop concentration and effective radius, when nss-sulfate is 1000 cm^{-3} . This contradicts the general rule that adiabatic parcel models support a negative correlation between cloud droplet number and effective radius.

- (2) Figure 3 in Z05 shows that updraft velocities exceeding 0.2 ms^{-1} , do not affect the cloud droplet concentration.
- (3) Z05 suggested that indirect forcing by nss-sulfate may be overestimated in cases when sea salt is present.

Because of these differences from earlier studies, the present study sought to reproduce the results found by Z05.

2. Model description

The parcel model used has been described in detail by Kuba et al. (2003), and Takeda and Kuba (1982). Only the particle method (not the bin method) in the parcel model was used in this study, to focus attention on activation of cloud condensation nuclei (CCN), and achieve accurate estimates of the number of cloud drops. For simplicity, only condensation processes were considered in this study. Time change of water vapor, caused by condensation/evaporation of droplets, is calculated explicitly.

Based on previous studies (Mordy 1959; Fitzgerald 1974; Feingold et al. 1999), we assumed that CCN smaller than $0.1 \mu\text{m}$ in radius, and CCN larger than $1 \mu\text{m}$ in radius, are initially assumed to be in equilibrium at 99% and 90% RH respectively, at cloud base (100% RH). Intermediate CCN are initially assumed to be in equilibrium between 99 and 90% RH, as a function of radius. In spite of selected arbitrariness, the initial sizes of the nuclei are probably

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not unrealistic (Fitzgerald 1974). It does not seem appropriate to add complexity that is not well resolved by the model (Feingold et al. 1999), and that does not affect significantly such numerical results as cloud droplet concentration and effective radius. Z05 did not describe the assumption about initial radii of aerosol particles. Brownian coagulation was taken into account in their model, but the effect of Brownian coagulation on their results, was not mentioned in Z05. Z05 did not describe the starting point of calculation. If calculation starts near the cloud base, Brownian coagulation may not have a large influence on the results.

CCN activation and initial cloud droplet growth by condensation were computed in a Lagrangian framework (particle method) that incorporates the solute effect of CCN. This method therefore avoids numerical diffusion of the droplet size distribution. The CCN size distribution was approximated using discrete radius classes, and initial class values were chosen prudently. The number of nuclei included in one class, was less than 1% of the number of activated cloud droplets. This limited errors in the concentration of activated cloud droplets to 1%. CCN (up to 10 μm in radius) in this study were partitioned into 185 separate classes. More than 100 of the classes could activate and evolve into cloud droplets. The initial minimum radius for CCN must be smaller than the radius of the smallest CCN that can be activated, because simulated results would be invalid if all CCN are activated. The time evolution of the representative droplet radius in each class is calculated as detailed in Takeda and Kuba (1982), and Kuba et al. (2003). The time step in this model is 0.05 seconds. Because droplets approach their equilibrium radius and do not exceed it before they reach their critical radius, a short time step of time integration is needed. In case 0.05 seconds is not short enough, the growth by condensation of each droplet during each step is limited, so that the radius of each droplet does not exceed its equilibrium radius. Z05 did not describe the length of the time step clearly.

3. Numerical experiments

Two nss-sulfate CCN supersaturation spectra (1000 cm^{-3} and 300 cm^{-3}), and one sea-salt

Table 1. Microphysical properties at 300 m above the cloud base. (Updraft velocity is 0.2 ms^{-1})

Initialization		Max.		
Nss-sulfate (cm^{-3})	Wind speed (ms^{-1})	Supersaturation (%)	Effective radius (μm)	Cloud drops concn. (cm^{-3})
300	0	0.254	7.95	278
300	15	0.201	7.85	313
1000	0	0.151	6.30	636
1000	15	0.122	6.72	556

CCN (for wind speeds of 15 ms^{-1}), were prepared following Fig. 4 and Table 1 by Z05. Sea-salt CCN supersaturation spectrum was made following Fig. 3 in O'Dowd (1997), but composition of sea-salt was assumed to be the same as nss-sulfate particles, $(\text{NH}_4)_2\text{SO}_4$, for simplicity. Since supersaturation spectra is more important than size distribution to describe the ability of CCN, this simplification does not cause a serious error in the results. Air temperature and pressure were initially 280 K and 880 hPa, respectively, as in the study by Z05. Temperature in the air parcel changes according to a vertical pressure gradient and condensation rate of the vapor. Z05, however, did not mention a vertical pressure gradient, so a value of -0.1 hPa m^{-1} was used, according to international standard atmosphere.

4. Results

Table 2 in Z05 shows cloud top microphysical properties, but Z05 did not describe cloud depth, or the altitude above the cloud base where the results were obtained. Z05 should have described them, because cloud microphys-

Table 2. Ratios of cloud droplet numbers between cases of nss-sulfate 1000 and 300

Updraft velocity (ms^{-1})	No sea salt	With sea salt ($U = 15 \text{ ms}^{-1}$)
0.1	1.4	1.2
0.2	2.3	1.8
0.3	2.7	2.3
0.5	3.2	2.8
1.0	3.2	2.9

Fig. 1. Vertical profiles of supersaturation and of the radii of cloud droplets condensed on different nss-sulfate particles (dry radii are 0.0746, 0.0768, 0.0774, 0.2062, and 2.0435 μm , respectively), for nss-sulfate of 1000 cm^{-3} and an updraft velocity of 0.2 ms^{-1} .

ical properties change with altitude above the cloud base. Figure 1 shows vertical profiles of supersaturation and of the radii of droplets condensed on different nss-sulfate particles (dry radii were 0.0746, 0.0768, 0.0774, 0.2062, and 2.0435 μm , respectively), for a case in which nss-sulfate was 1000 cm^{-3} , and the updraft velocity was 0.2 ms^{-1} . Supersaturation was calculated from temperature and water vapor every 0.05 seconds in this study, so that the maximum value can be estimated accurately. Small droplets grew very slowly under the low supersaturation conditions caused by very low updraft velocities and high CCN concentrations. In fact, even after growing beyond their critical radii (it means that their critical supersaturations are smaller than the maximum supersaturation), some of them evaporate and remain as wet CCN, because of a decrease in supersaturation. It was therefore difficult in this case to distinguish between activated and inactivated particles. Z05 suggested that droplets were activated if critical supersaturations were

smaller than the maximum supersaturation. However, it is difficult to estimate the number of cloud droplets in this condition. Table 1 is the same as Table 2 in Z05, but only for cases of nss-sulfate 300 cm^{-3} and 1000 cm^{-3} . Droplets with radii exceeding $1 \mu\text{m}$ at 300 m above the cloud base, were considered cloud droplets in Table 1.

Figure 2 is the same as Fig. 3 by Z05, but only for cases of nss-sulfate concentrations of 300 and 1000 cm^{-3} , with and without sea salt particles at $U = 15 \text{ ms}^{-1}$. Results of the present study differ from those by Z05 as follows:

- (1) Cloud droplet concentrations and maximum supersaturations, were smaller than those found by Z05. The difference in the cloud droplet concentration for cases of nss-sulfate 1000 cm^{-3} is more than 30%, and that in the maximum supersaturation is more than 0.1% RH, between the present study and Z05.
- (2) Correlation between cloud drop concentra-

Fig. 2. Cloud droplet number versus updraft velocity when nss-sulfate were 300 or 1000 cm^{-3} . Solid lines show the case with no sea-salt particles; broken lines show the case with sea-salt particles at $U = 15 \text{ ms}^{-1}$.

tion and effective radius was negative in the present study. In contrast, Z05 showed that the correlation was negative for the case of nss-sulfate 300 cm^{-3} , and positive for the case of nss-sulfate 1000 cm^{-3} . In addition it was negative for the case of nss-sulfate 5000 cm^{-3} in their study.

- (3) The effect of updraft velocity on the number of cloud drops was larger in this study than in the study by Z05, especially for updraft velocities that exceeded 0.2 ms^{-1} .
- (4) When the updraft velocity was small, the effect of the CCN number on the number of cloud drops, was small in the present study, but large in the study by Z05.

Table 2 shows the ratios of the number of cloud droplets between cases with nss-sulfate of 1000 and 300 cm^{-3} . The effect of sulfate particle concentration on cloud droplet concentration, was somewhat larger without sea-salt particles than with sea-salt particles. However, the difference was not as large as that found by Z05.

5. Conclusions

Several results of Z05 disagreed with previous studies, and were not reproduced in this study. Our study showed that nss-sulfate indirect forcing is not significantly reduced by the

presence of sea salt particles. Nss-sulfate particles have a small effect on a cloud microstructure when the updraft velocity is small, and a large effect when the updraft velocity is large. The influence of sea salt particles on nss-sulfate indirect forcing is not large. Therefore, the results obtained by Z05, do not seem to be general (or universal). More discussion and comparison with other models will be needed. Additionally, they should show the following essential information that is missing for readers to understand the content of the paper; cloud thickness, information of soluble organic components, information of bin method (one-moment or two-moment, one-dimension or two-dimension, technique of calculation), and the starting point of calculation (near the cloud base or far from the cloud base).

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