NOTES AND CORRESPONDENCE

CCN Ability of Asian Mineral Dust Particles and Their Effects on Cloud Droplet Formation

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Abstract

To understand the cloud condensation nuclei (CCN) ability of Asian mineral dust particles, we investigated the activation spectrum (fraction) and the hygroscopicity parameter $\kappa$ of the certified reference materials for Asian mineral dust (CAD) particles. The CAD particles were dispersed in dry conditions using a rotating brush generator and then measured with a cloud condensation nuclei counter and a condensation particle counter. The concentration ratios of CCN to condensation nuclei (CN) of the CAD particles were 0.7 at supersaturation (SS) = 0.2% and 0.8 at SS = 0.4%. This suggests that Asian mineral dust particles, by themselves, may effectively act as CCN. The mean hygroscopic parameter $\kappa$ of the CAD particles estimated from the relationship between the measured critical supersaturation and dry diameter was 0.014. Using this $\kappa$-value, we simulated the change in the size distribution from CCN to droplets during adiabatic ascent by a parcel model. The simulation suggested that the CAD (low hygroscopicity) particles do not grow large enough to promote collision-coalescence in the early stage of cloud formation. The hygroscopic parameter $\kappa$ of internally mixed particles of CAD and sea salt was calculated by using the $\kappa$-Köhler theory, and their activation and subsequent condensation growth processes during adiabatic ascent were also simulated with the parcel model. The simulation results suggest that the hygroscopicity of Asian mineral dust particles was considerably enhanced, and droplets activated on them grew much larger than those activated on pure mineral dust particles.

1. Introduction

A large amount of mineral dust particles are released in East Asia. Mineral dust particles have important roles that impact the global climate via their direct effect on the radiation budget of the atmosphere (Tegen et al. 1997; Sokolik et al. 2001). Acting as cloud condensation nuclei (CCN) and/or ice nuclei (IN), they also impact global climate by modulating the radiative properties, energy and water cycle of cloud systems through changes in cloud microphysical structures, coverage, and the lifetime of clouds (indirect effect) (Levin et al. 1996; Wurzler et al. 2000). Recent reports indicated that Saharan dust acts as not only IN (DeMott et al. 2003; Sassen et al. 2003; Cziczo et al. 2004) but also CCN (Koehler et al. 2007, 2009; Twohy et al. 2009). The CCN ability of Asian mineral dust particles has not yet been studied, although it is well known that they act as IN (Isono et al. 1959; Sakai et al. 2004; Mohler et al. 2006; Field et al. 2006). Due to the fact that seas (the Yellow Sea, East China Sea, and Sea of Japan) and polluted regions (industrial and/or populated cities) are located leeward of the source regions of Asian mineral dust; mineral dust particles form internally mixed particles through coagulation with a variety of aerosol particles and heterogeneous reactions of gas species. As a result of internal mixing,
particles undergo changes in size, shape, surface conditions, and chemical properties; these modifications are important because they change both the radiative properties of mineral dust particles and their ability to act as CCN (Liou and Ou 1989; Dentener et al. 1996; Quijano et al. 2000; Sokolik et al. 2001; Kelly et al. 2007). However, details of the CCN abilities and modification processes are currently poorly understood.

If Asian mineral dust particles, a large amount of which floats in the atmosphere, effectively act as CCN, there is a significant possibility that they impact climate change and the hydroosphere. Therefore, it is important to investigate the CCN ability of Asian mineral dust particle. Furthermore, if CCN ability is changed by physico-chemical modification during their transport, it is necessary to understand their CCN ability before and after the modification.

To understand the CCN ability of Asian mineral dust particles and to parameterize the composition dependent water activity of a solution droplet with the volume of the dry solute and the droplet, we investigate the activation spectrum and the hygroscopic parameter \( k \), the dry solute and the droplet, we activate the solution droplet with the volume of particles and to parameterize the composition dependent activation spectrum, the hygroscopic parameter \( k \), and the droplet size distribution (DSD) activated on CAD in a cloud condensation nuclei counter (CCNC). Values of \( k \) for specific compounds, or arbitrary mixtures can be determined experimentally by fitting CCN activity or hygroscopic growth factor data.

Nishikawa et al. (2000) produced the certified reference material from the loess plateau (Gansu Province, China) to reference Asian mineral dust. Detailed information (e.g., elemental composition) was provided by Nishikawa et al. (2000). They demonstrated that the mineral composition of the CAD particles and Asian mineral dust aerosols collected in Japan (Osaka Pref., Kagoshima Pref., and Yamaguchi Pref.) were very similar in elemental composition and claimed that CAD is an appropriate reference material for Asian mineral dust aerosol particles.

A rotating brush disperser (Palas, Model RBG-1000) was used for dry particle generation. To simulate dust particles in the atmosphere, the dispersed particles were stored in an aerosol buffer tank (600 L volume) via two cyclones (50% cutoff diameter 2.5 \( \mu m \) and 1.0 \( \mu m \)), and then supplied to aerosol instruments.

The activated fraction of the CAD particles was determined by measuring CCN concentrations with the CCNC (DMT Inc., Model CCN-2) and CN concentrations with CPC (TSI Inc., Model 3772). The CCNC is a continuous-flow thermal-gradient diffusion chamber for measuring aerosols that can act as cloud condensation nuclei. The aerosol sample is introduced into the supersaturation column along its centerline and is surrounded by a particle-free sheath flow.

The wall temperature along the column gradually increases to create a well-controlled and quasi-uniform centerline supersaturation by taking advantage of the difference in diffusion rates between water vapor and heat. By increasing the temperature gradient, the supersaturation can be varied between 0.07–2.0%. Particles (\( D_p > 0.75 \mu m \)) large enough to be detected by the built-in optical particle counter (OPC) of the CCNC are counted as CCN. The CCNC measured CCN number concentrations and size distributions of droplets from 0.75 \( \mu m \) to 10 \( \mu m \) in optical equivalent diameter with a refractive index of 1.33 (water). The temperature and supersaturation with respect to water are well controlled in the supersaturation column of the CCNC, and the critical supersaturation and condensation growth rates of the particles vary with their physico-chemical properties. The measured DSD was therefore used to validate a parcel model implemented with the \( \kappa \)-Köhler theory for these low-hygroscopicity particles.

A differential mobility analyzer (DMA, TSI Inc., Model 3080), CCNC, and CPC were used for measuring hygroscopicity parameter \( k \). The particles classified by DMA were simultaneously measured with CPC and CCNC. The activated fraction of the particles (\( N_{cn}/N_{cn} \)) was calculated from the concentrations of CN (\( N_{cn} \)) and CCN (\( N_{cn} \)), and the supersaturation was increased in a stepwise manner, the critical supersaturation was then determined to be the supersaturation at which a minimum of 50% of the particles activated as cloud droplets. The critical supersaturation as a function of dry particle diameter was obtained from each classification for particle diameter. Details of this procedure have been described by Koehler et al. (2007) and Rose et al. (2008).

3. Results

The size distribution of the dry CAD particles is shown in Fig. 1. The circles denote the size distribu-
Fig. 1. Size distributions of the dry CAD particles (squares and circles) and droplets activated and grown in the supersaturation column of CCNC, where SS is set to 0.4% (triangles and black line). The triangles were measured with the CCNC, and the black line was calculated from the size distribution of the dry CAD particles (gray line) by using a parcel model implemented with the $k$-Köhler theory.

In the $k$-Köhler theory (Petters and Kreidenweis 2007), the $k$-value of the particle indicates its hygroscopicity. Once the $k$-value was known, the activation and subsequent condensation growth of the particle could be calculated. Figure 3 indicates the relationship between critical supersaturation and the dry diameter of CAD, Arizona Test Dust (ATD), and sodium chloride (NaCl) particles. The $k$-value of the sodium chloride particle determined in the present study was 1.28, which is consistent with the $k$-value obtained by Petters and Kreidenweis (2007). The measured $k$-values of CAD and ATD particles in Fig. 3 are distributed between the theoretical lines for $k = 0.011$ and $k = 0.020$. The mean $k$-value of CAD particles was 0.014, and that of ATD was 0.017; both are within the range of the hygroscopicity parameter, $0.01 \leq k \leq 0.08$, for mineral dust particles suggested by Koehler et al. (2009).

4. Validation of the parcel model

The experimental results presented in the previous section suggest that CAD particles by themselves may effectively act as CCN. In the next section, we discuss the effect of mineral dust particles on cloud droplet formation over East Asia. To investigate this effect, the activation and subsequent condensation growth of CCN particles during adiabatic ascents are simulated by using a parcel model. To simulate such processes, the parcel model of Chen and Lamb (1994) was modified to include the $k$-Köhler theory of Petters and Kreidenweis (2007) instead of the classical Köhler theory.

First, to check the performance of the improved model for CCN particles with high hygroscopicity, the simulation results of an adiabatic ascent are compared with those simulated with the original model. In both
Fig. 3. Theoretical (solid lines) and measured (marks) critical supersaturation as a function of dry diameter for NaCl, ATD, and CAD particles.

Simulations, CCN particles were assumed to be ammonium sulfate \((\kappa = 0.61)\) and to have the same size distribution as that of the dry CAD particles (circles and squares in Fig. 1). The initial air temperature, pressure and ascent rate were set to 15°C, 1000 hPa, and 3 m s\(^{-1}\), respectively. The simulation results of the improved model were in good agreement with those of the original (Figs. 5, 6). Thus, it can be safely concluded that the improved model accurately simulates cloud droplet formation from CCN particles with high hygroscopicity.

Next, to determine how accurately the activation and subsequent condensation growth processes were reproduced in the supersaturation column of CCNC, we compared the measured DSD with the simulated DSD grown from sodium chloride particles \((\kappa = 1.28)\) in the supersaturation column of CCNC (Fig. 4). In the simulation, the air pressure, the prescribed maximum water supersaturation and the residence time of the particles in the supersaturation column were set to 1000 hPa, 0.4%, and 12 sec, respectively. The air temperature and supersaturation profiles along the center line of the supersaturation column were referenced to Fig. 1(b) in Roberts and Nenes (2005). This comparison was also in good agreement, indicating that the measured DSD reflected the activation and subsequent condensation growth in the supersaturation column with a given thermodynamic profile.

To check the performance of the improved model for CCN particles with low hygroscopicity, we simulated the activation and subsequent condensation of the dry CAD particles with the size distribution indicated in Fig. 1 in the supersaturation column. The profiles of thermodynamic parameters along the center line of the CCN supersaturation column are the same as those for the simulation of sodium chloride particles mentioned above. The black line in Fig. 1 indicates the simulation result when the \(\kappa\)-value is assumed to be 0.014 (corresponding to the CAD particle). The simulated DSD is generally consistent with the DSD measured with the CCNC at SS = 0.4%.

It was confirmed that the improved model can simulate cloud droplet formation from CCN particles, both high and low hygroscopicity, with reasonable accuracy.

5. Discussion

This section discusses the effect of mineral dust particles on cloud droplet formation over East Asia based on the results of the parcel model simulations. The broken lines in Figs. 5b to 5d indicate the simulation results when the air parcel containing the CAD particles is lifted adiabatically. The CCN size distribution, initial temperature, initial pressure, and ascent speed were same as those mentioned in Section 4. The time evolution of mean volume diameter \((D_{\text{vol}})\) before activation (240 sec) for ammonium sulfate was compared with that for CAD. The \(D_{\text{vol}}\) of the CAD particles did not change until immediately before droplet activation because the hygroscopicity was low. Since the water vapor was not consumed by the CAD particles until droplet activation, the maximum supersaturation for CAD exceeded that for ammonium sulfate (Fig. 5b). Although the critical supersaturation was higher, the concentration of droplets \((5 < D < 100 \mu m)\) for CAD was lower than that for ammonium sulfate (see Fig. 5c) because the critical diameter at maximum super-saturation for CAD
Fig. 5. Time series of (a) pressure and temperature, (b) relative humidity, (c) number concentration of droplets between 5.0 \( \mu m \) and 100 \( \mu m \), and (d) mean volume diameter during adiabatic ascent (updraft velocity of 3 m s\(^{-1}\)) simulated by the parcel model. The solid line denotes ammonium sulfate (using Köhler theory), the dotted line denotes \( k = 0.61 \) (ammonium sulfate; using the \( k \)-Köhler theory), and the broken line denotes \( k = 0.014 \) (CAD; using the \( k \)-Köhler theory).

Figures 6a, b, and c depict DSDs 50-sec before activation, 50-sec after activation, and 300-sec after activation, respectively. The DSDs for CAD were always narrower than those for ammonium sulfate. The simulations of an adiabatic ascent with the updraft velocity of 0.5 m s\(^{-1}\) (stratiform cloud) had similar results (not shown here). These results suggest that the mineral dust (low hygroscopicity) particles do not grow into large droplets that promote collision-coalescence in a short time after activation. Figure 3 clearly demonstrated that the critical sizes at which pure dust particles activate at a given supersaturation are several times larger than those for high-hygroscopicity particles, like sea salt (NaCl) and ammonium sulfate (\( k = 0.61 \)). Furthermore, various types of aerosol particles exist in the atmosphere and the number concentrations of Asian mineral dust particles are generally lower than those of CCN (hygroscopic) particles generated from natural and anthropogenic sources. Therefore, it is not likely that pure Asian mineral dust particles significantly affect cloud
droplet formation. Mineral dust particles over East Asia may be modified and become internally mixed particles through the adsorption of gases or coagulation with other kinds of aerosol particles during their transport. The internally mixed particles of dust and sea salt were often found in the boundary layer over Japan in the spring and its fraction often exceeded 80% of total dust particles (e.g., Zhang et al. 2003, 2006; Yamashita et al. 2005). Although it is possible that the CCN ability of the mineral dust particles changes with such modifications, few studies have addressed this issue. In the Köhler theory, the $k$-values of the internally mixed particle can be calculated if the $k$-values and the volume ratio of each of the particles that compose the mixed particle are known. We calculated the $k$-value of the internally mixed particles of mineral dust and sea salt by the $k$-Köhler theory, assuming the $k$-value of dust to be 0.014 (mean value of the CAD) and that of sea salt to be 1.28. According to Zhang and Iwasaka (2004), the weight ratio of dust and sea salt in mixed particles varied from dust-dominated to sea salt-dominated. If the volume ratio of dust to sea salt is 3:1, the calculated $k$-values of the mixed particles is 0.33; if the ratio is 2:2, the $k$-values is 0.65; and if the ratio is 1:3, the $k$-values is 0.96. The calculated $k$-values of internally mixed particles are much higher than those of pure dust particles. The broken lines in Fig. 6 denote the DSDs obtained from adiabatic ascent simulation when hygroscopicity was 0.33 (volume ratio of 3:1). It is clear that the DSDs of the internally mixed particles of dust and sea salt were always broader than those of pure dust particles and narrower than those of sea salt particles (Fig. 6). These results suggest that the CCN ability of dust particles changes considerably toward high hygroscopicity, and droplets grow much larger than those activated on pure mineral dust due to the mixing with sea salt. The coagulation between dust particles and sea salt particles decreases fine CCN number concentrations in the atmosphere, and may drastically enhance the hygroscopicity of dust particles and produce giant CCN particles. It is likely that the internal mixing of dust particles with hygroscopic particles produces preferable conditions for large droplet formation through the enhanced collision-coalescence growth of cloud droplets and/or direct formation of raindrop embryos. In order to fully understand the effect of Asian mineral dust particles on warm rain cloud formation, it is necessary to investigate the CCN ability of mineral dust internally mixed with other types of particles in the future.

6. Summary

Investigation of the CCN ability of CAD particles indicates that Asian mineral dust particles may effectively act as CCN. Using the mean hygroscopic parameter $k$ of the CAD particles ($k = 0.014$) determined experimentally in this study, their activation and subsequent condensation growth during adiabatic ascent were simulated using the validated parcel model. The results suggested that pure mineral dust (low hygroscopicity) particles do not grow into large droplets that promote collision-coalescence in a short time after CCN activation. Calculation of the hygroscopicity of mineral dust particles internally mixed with sea salt suggests the possibility that the hygroscopicity of mineral dust particles is considerably enhanced by internal mixing with sea-salt particles and the droplets activated on them grow much larger than those on pure mineral dust particles.

Clarification of the effects of Asian dust particles on cloud formation requires more intensive field observations and laboratory experiments on the CCN abilities of Asian mineral dust particles internally mixed with natural and anthropogenic hygroscopic particles.

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References


