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1. Introduction

East Asia is one of the place where releases a large amount of the mineral dust particles. Mineral dust particles have important role that impact upon the global climate via their direct effects on the radiation budget of the atmosphere [Tegen et al., 1997, Sokolik et al., 2001]. They also impact upon the global climate through modifying the radiative properties through the microphysics, amount and lifetime of clouds as contributing CCN and IN (indirect effect) [Levin et al., 1996; Wurzler et al., 2000]. From recently report, Sahara dust acts as not only IN (DeMott et al., 2003, Sassen et al., 2003, Cziczo et al., 2004) but also CCN (Twohy et al., 2009; Koehler et al., 2007, 2009). Although it is well known that the mineral dust particles generated from Asia act as IN (Isono et. al., 1959, Mohler et al., 2006, Field et al., 2006, Sakai et al., 2004), it is not known to act as CCN.

If Asian mineral dust particles that float a large amount in the atmosphere effectively act as CCN, there is possibility to impact significantly upon the climate change and the hydrosphere. So, it is important to investigate the CCN property of the Asian mineral dust particles. And if CCN property of Asian mineral dust changes by modification during transport, it is necessary to grasp the CCN property before modification.

To understand CCN property of Asian mineral dust particles, we investigated activation spectra and hygroscopicity parameter κ defined by Petters and Kreidenweis (2007) of the certified reference materials for Asian mineral dust particles produced by Nishikawa et al. (2000). And we observed CCN concentration and droplet size distribution (hereafter DSD) activated from aerosol particles in the supersaturation column of the CCNC during the dust event (17th - 19th March 2009) at

Tsukuba (36.06°N, 140.13°E), Japan. In this paper, we present the results of the CCN property of the certified reference materials for Asian mineral dust particles, and CCN concentration and DSD activated in the CCNC during the dust event observed at Tsukuba.

2. Method

2.1. Investigation of CCN property

The investigations of CCN property of certified Asian mineral dust particles (hereafter CAD) were conducted by measuring the activation spectrum, hygroscopicity parameter κ , and DSD activated from CAD in the CCNC.

Nishikawa et al. (2000) produced two certified reference materials from the loess plateau (Gansu Province, China) and from desert surface soil of the Tengger Desert to reference Asian mineral dust. The first of these is designated as China Loess (CJ-1) and the other as simulated Asian mineral dust (CJ-2). Although CJ-2 is desirable as reference for Asian mineral dust, CJ-1 was used in this study because we do not have much amount of CJ-2. The detailed information such as elemental composition is described by Nishikawa et al. (2000). They showed that the mineral composition of CJ-1, CJ-2, and Asian mineral dust aerosols collected in Japan (Osaka, Yakushima, and Yamaguchi) are very similar and claim that CJ-1 and CJ-2 are appropriate reference material for Asian mineral dust aerosol.

The rotating brush disperser (Palas, Model RBG-1000) was used as dry CAD generation. The dispersed particles were once stored in the aerosol buffer tank (600 liter volume), then supplied to instruments.

The measurement of activation spectrum was conducted by measuring CCN and CN concentration. CCN and CN concentration were measured with the DMT-CCNC (DMT, Model CCN-2) and the CPC (TSI, Model 3772).

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In the measurement of the hygroscopicity parameter κ , DMA (TSI, Model 3080), DMT-CCNC, and CPC were used. The particles classified by the DMA were simultaneously measured with CPC and DMT-CCNC. The activated particle fraction (N_{ccn}/N_{cn}) was calculated from the concentrations of CN (N_{cn}) and CCN (N_{ccn}), and a N_{ccn}/N_{cn} over dry diameter was obtained from every classification of particle diameters at constant supersaturation. Detailed information of this procedure is described by Koehler et al. (2007) and Rose et al. (2008).

2.2. Observation of dust event

DMT-CCNC and OPC (YGK, Model TD-100) were used for observation during dust event observed on 17th - 19th March 2009. Since the DMT-CCNC has two measurement systems, two different measurements were conducted. One system measured CCN concentration at constant supersaturation ($SS=0.4\%$) at 1 Hz. Another one measured CCN concentration at various supersaturation ($SS=0.07, 0.2, 0.4, 0.8, 1.0\%$). It took 27 minute for 1 scan. CCN particles were drawn from outside of our cold environmental simulation building to aerosol buffer tank (600 liter volume) located in the building at 70 lpm, then drawn into the DMT-CCNC at 1.0 lpm through 3 m long conductive tube. OPC measurement was conducted at our main building. Aerosol particles were directly drawn by OPC at 1 lpm through 1 m long conductive tube.

3. Results

3.1. Investigation of CCN property

Fig. 1 shows the activation spectrum of CAD. The ratio N_{ccn}/N_{cn} at 0.2% and 0.4% supersaturation was 0.6 and 0.8, respectively. More than half of CAD was activated at 0.2% supersaturation. Fig. 2 shows DSDs activated in the DMT-CCNC at 0.4%, 1.0% supersaturation and dry CAD size distribution. Since CAD included particles larger than 0.75 μm (see Fig. 2), particles which did not activate to droplet might be included in the DSDs shown in Fig. 2. However, we ignored it because its fraction was a little judging from the dry CAD size distribution and DSDs in Fig. 2. The DSDs was clearly different from dry CAD size distribution. Therefore, this result revealed

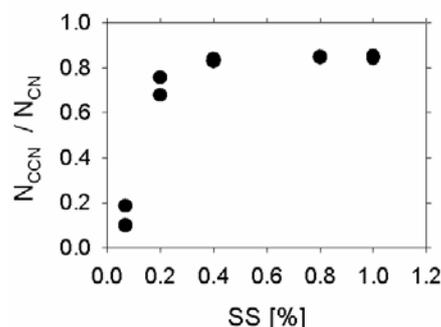


Fig. 1. Activation spectrum of the Certified Asian mineral dust (CAD) particle.

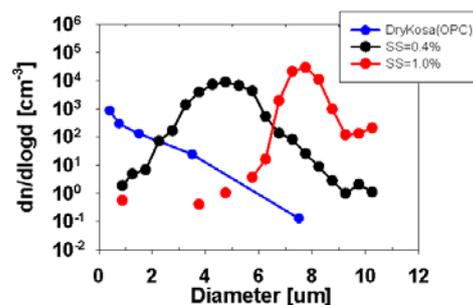


Fig. 2. Droplet size distributions grown up in the DMT-CCNC and dry size distribution of CAD measured with OPC.

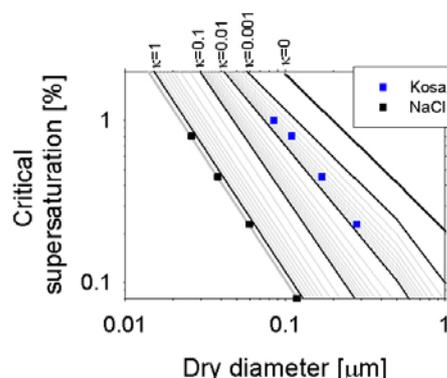


Fig. 3. Measured critical supersaturations as a function of dry diameter for NaCl and CAD.

that CAD activated and then grow up by condensation in the DMT-CCNC. Therefore, it found that CAD effectively acts as CCN in the case of its only.

Fig. 3 shows supersaturation vs dry diameter plot of CAD and NaCl. According to Petters and Kreidenweis (2007), κ of NaCl particle is 1.28. The value of NaCl particle in this paper also indicated 1.28. We confirmed that our measurement didn't have any problem. The plots of supersaturation and diameter plane of CAD were distributed between 0.004 and 0.008. The mean κ of CAD was 0.005. The black dots and line in Fig. 4 shows the DSDs measured with the DMT-CCNC and calculated the condensation growth in the DMT-CCNC by κ -Köhler theory using dry CAD size distribution. To calculate the condensation growth of CAD in the DMT-CCNC, pressure, supersaturation and exposure time in supersaturator were assumed to be 1000 hPa, 0.4%, and 10 second, respectively. The DSD simulated by κ -Köhler theory reproduced DSD measured with the DMT-CCNC above 5 μm size range. This result suggests that κ value measured in this study was plausible value.

3.2. Observation during the dust event observed on 17th – 19th March 2009.

Fig. 5 shows that points reported dust event by local meteorological observatory of Japan Meteorological Agency from 16th to 19th March 2009. It is clear that dust event was distributed wide area of Japan during the dust event observed at Tsukuba, Japan. HYSPLIT Back trajectories (not shown here) from Tateno aerological observatory (36.06°N, 140.13°E) showed that air mass come from the continent. Time series of the vertical distribution of backscatter coefficient and depolarization ratio observed in Tsukuba by National Institute of Environmental Science (not shown here) showed that dust layer reached surface around 7:00 JST on 17th March. It is clear that air mass observed at Tsukuba on 17th March were included Asian mineral dust particles.

Fig. 6 shows time series of concentrations of aerosol particles (a), CCN (b), SO₂ (c), NO (d), and NO₂ (e). Aerosol number concentration larger than 1 μm suddenly increased around 7:00 JST on 17th March. Timing of this increase was consistent with that which dust layer reached surface. So, we consider that dust event began at 7:00 JST on 17th March.

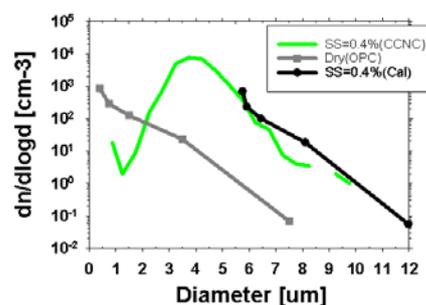


Fig. 4. DSD measured with DMT-CCNC and calculated condensation growth in the DMT-CCNC by κ -Köhler theory using dry CAD size distribution.

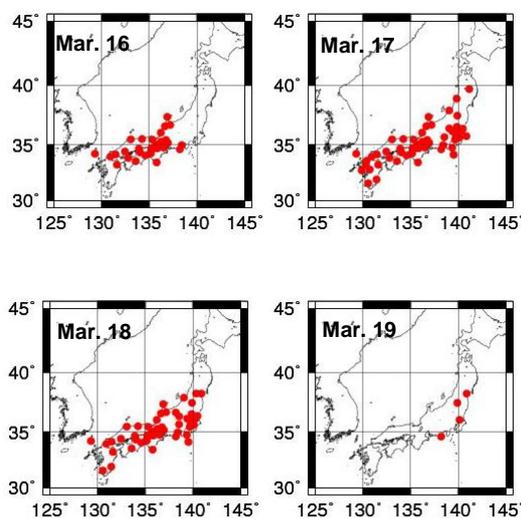


Fig. 5. Points reported dust event by local meteorological observatory of Japan Meteorological Agency from 16th to 19th March 2009.

According to the report from Tateno aerological observatory, it was shower from 11:10 to 11:40 JST. At that time, number concentration of aerosol particle and CCN were decreased. After then aerosol concentration larger than 1 μm returned to high concentration. However, CCN concentration was low immediately after the shower. Then CCN concentration gradually increased.

The peaks of SO₂, NO, and NO₂ gases synchronized that of CCN and fine (0.3-1.0 μm) aerosol particle concentration. This result suggests that mineral dust particles were hardly included in fine aerosol particle and that the mineral dust particle

hardly influence the variation of CCN concentration during the dust event.

Fig. 7 shows time series of size distribution of aerosol particles (a) and droplet activated in the DMT-CCNC (b). Aerosol size distribution during the dust event indicated broader tail than that before the dust event. The peak size of DSD activated in the DMT-CCNC between around 11:00 JST (near showering time) on 17th and 6:00 JST on 18th March shifted bigger than that before the shower. On the other hand, that after 6:00 on 18th March was almost same as that before the shower. These results mean that DSD changed during the dust event. We do not analyze this cause yet.

4. Discussion

Results of the investigation of CCN property of CAD and the observation during the dust event were shown in Section 3. In this section, type of CCN included DSD activated in the DMT-CCNC during dust event was discussed by using hygroscopicity parameter κ and observation results.

Fig. 8 shows DSDs activated in the DMT-CCNC at 17:00-18:00 JST on 17th March and simulated by κ -Köhler theory to assume ammonium sulfate ($\kappa=0.61$) and CAD ($\kappa=0.005$) from aerosol size distribution measured with OPC (black dot and line). When ammonium sulfate was assumed, the calculated concentration was higher than that measured with the DMT-CCNC. The concentration larger than $7.5 \mu\text{m}$ gradually separated from the one measured with the DMT-CCNC. On the other hand, when CAD was assumed,

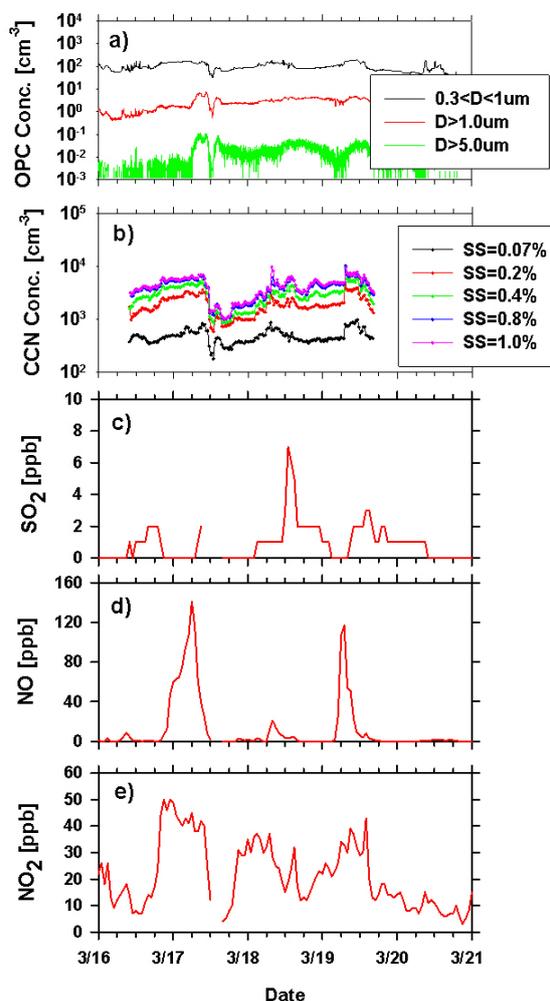


Fig. 6. Time series of number concentrations of aerosol particles (a), CCN (b), SO₂ (c), NO (d), and NO₂ (e).

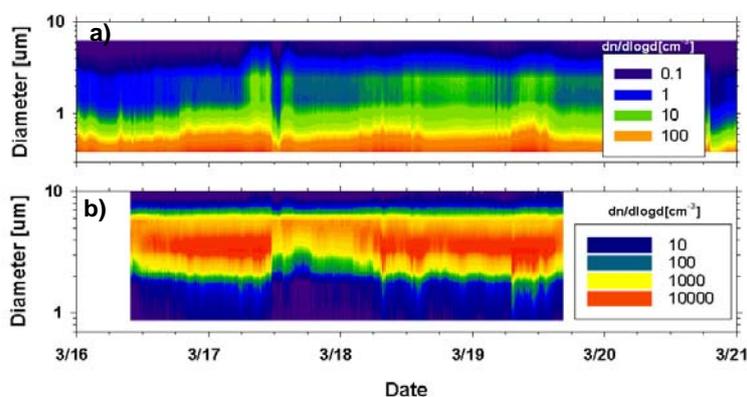


Fig. 7. Time series of size distribution of aerosol particles (a) and droplet grown up in the DMT-CCNC (b).

the calculated concentration was lower than that measured with the DMT-CCNC. The concentration smaller than 7.0 μm gradually closed to the one measured with the DMT-CCNC. These results suppose that droplets in the size range between 7 and 9 μm grown up in the DMT-CCNC during the dust event externally mixed with low (Asian mineral dust?) and high (ammonium sulfate?) hygroscopic particle.

Fig. 9 indicates coarse (left in Fig. 9) and fine (right in Fig. 9) particles taken through the transmission electron microscope. Fine particles were mainly quasi-spherical shape (liquid?). Coarse particles were mainly irregular shape (mineral dust?) these results suggest that particle composition of fine particle is different from that of coarse particle. This result supports our supposal. We have been analyzing elemental composition of sampled particles using EDX. So, we will understand the particle composition near future.

5. Conclusion

We investigated the activation spectrum and the hygroscopicity parameter κ of the CAD. And we observed the CCN concentration and DSD grow up in the DMT-CCNC during the dust event (17-20 March 2009) at Tsukuba (36.06°N, 140.13°E), Japan. From results of the investigation and observation, we conclude following points:

1. Asian mineral dust may effectively act as CCN.
2. Hygroscopicity parameter κ of Asian mineral dust may be between 0.004 and 0.008.
3. It is suggested that the mineral dust particle hardly influence the variation of CCN concentration during the dust event observed from 17th to 19th in March 2009.
4. There is possibility that droplets in the size range between 7 and 9 μm grown up in the DMT-CCNC during the dust event externally mixed with particles with low and high hygroscopicity parameter κ .

6. Acknowledgement

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7. References

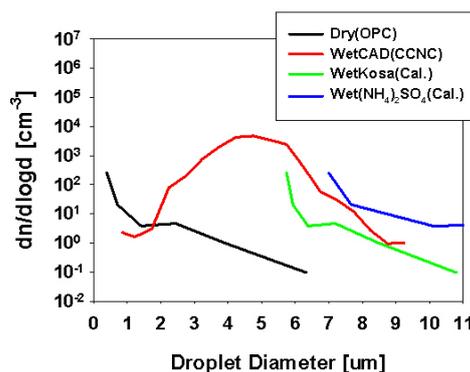


Fig. 8. DSD measured with DMT-CCNC at 17:00-18:00 in 17th 2009 (red line) and calculated by assuming ammonium sulfate ($\kappa=0.61$: blue line) and CAD ($\kappa=0.005$: green line) from size distribution measured with OPC (black line).

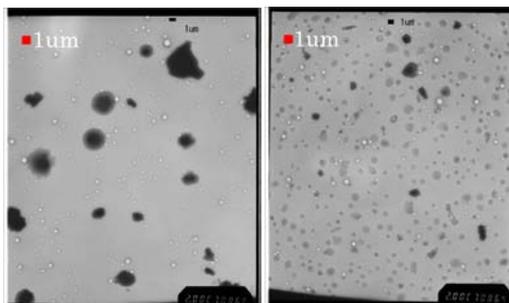


Fig. 9. Photo of aerosol particles taken through the transmission electron microscope. Aerosol particles were collected on mesh around 11:00 JST on 17th March 2009 by using two stages impactor. Left and right was collected on the 1st and 2nd stage mesh, respectively.

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