Vertical distribution of cloud condensation nuclei concentrations and their effect on microphysical properties of clouds over the sea near the southwest islands of Japan

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[1] Aircraft observations were performed over the sea near the southwest islands of Japan under Asian Atmospheric Particulate Environmental Change Experiment 2/Asian Pacific Regional Aerosol Characterization Experiment (APEX-E2/ACE-Asia) project during the period of 16–28 April 2001. The polluted air mass from east Asia was associated with very high concentrations of SO_2 (1–10 ppb) and aerosol particles (3000–5000 cm^{-3}) in the marine boundary layer. The cloud condensation nuclei (CCN) concentration at 0.3% supersaturation was as high as 800–2000 cm^{-3} during the penetrations of air pollutants from east Asia. The correlation coefficient between SO_2 and aerosol particles was significant in such polluted atmosphere. Concentration of CCN (N_{CCN}) was linearly related to concentration of aerosol particles (N_{AP}) according to $N_{CCN} \sim 0.75 N_{AP}$. The ratio of CCN to aerosol condensation nuclei particle concentrations was lower than 0.3 in the relatively clean maritime atmosphere, but it was as high as ~0.5 in the continentally influenced atmosphere in the boundary layer. These results indicated that the influence of anthropogenic pollutants from east Asia increased the contribution percentage of aerosol particles to CCN in the polluted atmosphere over the observation area. The observational results also indicated that a mean cloud droplet concentration (N_C) in the continentally influenced clouds was ~2 times as much as N_C in the relatively clean maritime clouds. The slope of log-log relationship between N_C and N_{CCN} was ~0.39. This study strongly suggests that high CCN concentration formed many cloud droplets and decreased their effective radius at similar liquid water content under the outflow of air pollutants from east Asia.

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1. Introduction

[2] Gases and aerosol particles are increasing at an alarming rate in the east Asia region because of rapid industrialization [*van Aardenne et al.*, 1999], increasing automobiles, and biomass and coal burning [*Elliott et al.*, 1997] in Asia. It is also noted that aerosol concentrations have increased over the northwestern Pacific Ocean because of anthropogenic pollutants in east Asia [*Nagao et al.*, 1999]. Similarly, high concentrations of SO_2 were observed over the China Sea, Yellow Sea, and Sea of Japan under the outflow of anthropogenic pollutants from the Asian continent [*Akimoto and Narita*, 1994; *Hatakeyama et al.*, 1995; *Thornton et al.*, 1999]. Recently, it has also been suggested that sulfate concentrations in the free troposphere were higher over Japan than over the northwest Pacific region [*Osada et al.*, 2002]. Anthropogenic particles, which contain soluble sulfate and nitrate compounds, are important contributors to the atmospheric budget of cloud condensation nuclei (CCN) [e.g., *O'Dowd et al.*, 1999].

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[3] In the atmospheric aerosol, CCN have become one of the central issues in climate change. Particles influence the radiation budget in two ways, directly through the scattering and absorption of solar radiation (direct effect) and indirectly through modifying the microphysical properties of clouds (indirect effect). CCN are usually $\sim$100 cm^{-3} in the relatively clean maritime atmosphere, while they are 1000 cm^{-3} or greater in the polluted atmosphere [*Twomey and Wojciechowski*, 1969; *Radke and Hobbs*, 1976; *Hudson and Xie*, 1999; *Ishizaka and Adhikari*, 2003]. *Ishizaka et al.* [1995] observed CCN concentrations as high as 1500 cm^{-3} in the boundary layer over the southwest islands of Japan. It was also noticed that the CCN concentrations were highly influenced by anthropogenic inorganic pollutants [*Ishizaka and Adhikari*, 2003]. On the other hand, CCN concentrations were often enhanced by clouds [*Radke and Hobbs*, 1969; *Saxena et al.*, 1970; *Dinger et al.*, 1970; *Hobbs*, 1993]. However, the relative importance of natural and anthropogenic sources of CCN is still unknown. It has been determined that a few percent to more than 50% of aerosol particles (larger than 0.01 μm in diameter) in the marine atmosphere act as CCN at a supersaturation (SS) of 1% or less [*Hudson and Frisbie*, 1991; *Hegg and Hobbs*, 1992; *Delene et al.*, 1998]. Although CCN are only a fraction of the total aerosol particles, they play an important role in cloud stability [e.g., *Hobbs et al.*, 1974]. High CCN concentrations increase cloud droplets [e.g., *Fitzgerald and Spyers-Duran*, 1973] and decrease the size of cloud droplets [e.g., *Garrett and Hobbs*, 1995]. The first indirect effect of aerosol particles derives from the fact that cloud albedo depends on the cloud liquid water path and droplet size [*Twomey*, 1977; *Hobbs*, 1993; *Vong and Covert*, 1998]. It has widely been realized that CCN are not only the precursors of cloud droplets but also strongly modulate cloud microstructure and hence the radiative properties of clouds [*Charlson et al.*, 1987; *Yu*, 1996]. The decrease in droplet size reduces gravitational collection efficiency of cloud droplets and precipitation efficiency [*Young*, 1993]. The modification of cloud microphysics thus leads to increased cloud lifetime and/or increased spatial extent and hence increased planetary albedo [*Hudson and Yum*, 2001; *Yum and Hudson*, 2002]. This microphysical effect on clouds is referred to as the second indirect effect [*Albrecht*, 1989; *Liou and Ou*, 1989; *Radke et al.*, 1989].

[4] The effect of pollutants on microphysical and optical properties of clouds comes up as an important problem from the above historical reviews. However, the role of CCN in east Asia and their impact on climate change are still unknown. We analyzed SO_2, aerosol particles, CCN, and cloud droplet concentrations from aircraft observational data over the sea near the southwest islands of Japan as a part of Asian Atmospheric Particulate Environmental Change Experiment 2/Asian Pacific Regional Aerosol Characterization Experiment (APEX-E2/ACE-Asia) project. The effects of pollutants on microphysical properties of clouds are discussed on the basis of the observational data.

2. Methodology

[5] Aircraft observations were performed over the sea near the southwest islands of Japan between 16 and 28 April 2001. A total of 11 flights was made using a King Air aircraft, Airborne Research Australia. Figure 1 shows the location of the flight observation area. Vertical profiles of aerosol and CCN concentrations were made in clear sky well away from any clouds on 16, 18, 23, 26, and 28 April 2001 and in the vicinity of clouds on 21, 22, 24, and 27 April 2001. In addition to these flights, horizontal measurements of microphysical properties of clouds were performed at some levels on 21, 22, 24, and 27 April 2001. Stratiform clouds were observed on 21 and 24 April, and convective clouds were observed on 22 and 27 April in the observation area. On 27 April, there were flights in morning and in afternoon at different locations.

[6] CCN concentrations and their spectra at low supersaturations (0.3% > SS) were measured with a CCN counter developed by Y. Ishizaka (model ACN-1000, Sigmatec). CCN were detected with a vertical-type thermal diffusion cloud chamber first described by *Laktionov* [1972] and later applied by *Hudson* [1980]. The counter consists of three vertical cloud chambers with optical detectors and one separate optical aerosol analyzer. The length of each vertical cloud chamber was 100 cm. The cloud chamber was built as a continuous flow diffusion chamber. The sample air introduced in the cloud chamber was surrounded by sheath air, which was circulated in the laminar condition. Flow rate of sampling air was 100 cm^3 min^{-1} in each chamber. The required supersaturation was created by setting temperature inside the vertical plates. The temperatures were accurately controlled with temperature controllers, and inside plates were wetted with distilled and deionized water. After the measurements the supersaturation of the cloud chamber was calculated from recorded temperatures of vertical plates in each chamber. The sample aerosols were subjected for 50 s to attain the critical equilibrium size. In most cases this is enough time to allow the drops to attain their steady sizes at which they are counted and sized by an optical detector. The droplets larger than 0.3 μm in diameter within the chamber were divided into six channels (0.3, 0.5, 1.0, 3.0, 5.0, and 10.0 μm in diameter). A set of data was recorded in a sequence of four systems (S1, S2, S3, and S4) for every 1 min. Each system measured the data every 15 s. The systems S1, S2, and S3 measured the droplet concentrations, and system S4 measured the ambient aerosol particles larger than 0.1 μm in diameter. The detection limit of this instrument was 0.04 cm^{-3}. Droplet concentration lower than 1200 cm^{-3} can be measured with $\pm$5% accuracy of counting values and concentrations between 1200 and 3000 cm^{-3} with lower accuracy by an optical detector. Droplet-counting data measured with the S1 system are analyzed in this study.

[7] To verify the performance of the instrument, the calibration was done using standard NaCl particles. NaCl particles were generated by atomizing dissolved salt (NaCl) solution and passed through a differential mobility analyzer (DMA) (model 3081). Only particles that were within a certain range of electrical mobility were passed through the DMA. Standard NaCl particles of known chemical composition were measured with a scanning mobility particle sizer (model 3936L10, Thermo Systems Inc. (TSI)). NaCl particles with the modal diameter of 100 nm were introduced in the cloud chamber (0.3% supersaturation). As an example, the cumulative size distributions of standard NaCl particles and droplets formed inside the cloud chamber at 0.3%

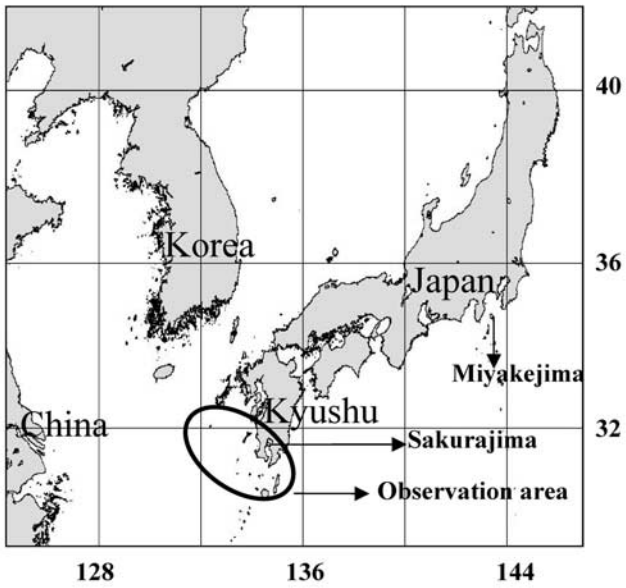


Figure 1. Map showing the observation area.

supersaturation are shown in Figure 2. Figure 2 shows that the concentration of NaCl particles ($d > 0.053$ μm) was 595 cm^{-3} and the concentration of droplets ($d > 0.5$ μm) was 578 cm^{-3}. According to the Köhler equation the critical droplet diameter corresponding to 0.3% supersaturation for NaCl particles at 20°C is ∼0.49 μm [*Mason*, 1971]. Hence the droplets larger than ∼0.49 μm in diameter were considered to be CCN activated at 0.3% SS in this study. The number concentration of droplets larger than critical droplet size of 0.49 μm in diameter agreed with that of total NaCl particles larger than critical size (dry) of ∼0.053 μm in diameter within ±5% accuracy of counting values.

[8] Ambient aerosol particles larger than 0.003 μm in diameter were measured with an ultrafine condensation particle counter (model 3025, TSI) and aerosol particles larger than 0.01 μm in diameter were measured with a condensation particle counter (model 3010, TSI). The concentration of ultrafine particles (UP) in the size range 0.003–0.01 μm in diameter was estimated from the difference between the total concentrations measured by the two condensation particle counters. Further, the size distribution of particles larger than 0.12 μm in diameter was measured with an active scattering aerosol spectrometer probe (ASASP-100X, Particle Measuring System, Inc. (PMS)). With regard to particle terminology, particles between 0.003 and 0.01 μm in diameter were named UP, particles between 0.01 and 0.12 μm in diameter were named nuclei mode particles (NP), particles between 0.12 and 1.05 μm in diameter were named fine particles (FP), and particles between 1.05 and 3.11 μm in diameter were named coarse-mode particles (CP) in this study. Separately, aerosol particles larger than 0.01 μm in diameter were roughly called condensation nuclei particles (CN), and particles 0.12–3.11 μm in diameter were called atmospheric particles (AP).

[9] SO_2 concentration was measured with a pulsed fluorescent ambient SO_2 analyzer (model 43S, Thermo Electron Corp.). The detection limit of this instrument was 0.1 ppb, and a maximum limit was set at 10 ppb during the observation period. SO_2 and aerosol particles were measured every second. Mean values of aerosol and/or SO_2 data for 15 s were compared with the CCN data. A Forward Scattering Spectrometer Probe (model FSSP-100, PMS) was mounted under the fuselage of the aircraft to measure number concentration and size distribution of cloud droplets (diameter of 2–47 μm) classified into 15 channels. The sampling rate of FSSP-100 was 1 s. Number concentrations and effective radius (r_e) of cloud droplets and liquid water content (LWC) were calculated from size distribution of cloud droplets in clouds with LWC $\geq$ 0.05 g m^{-3}.

[10] The isentropic back trajectories were used to investigate the origin and history of the air for 3 days (72 hours) prior to their arrival at the observation area. The back trajectories were computed for the final levels of 500, 1000, and 3000 m in altitude over observation areas using the Hybrid Single Particle Lagrangian Integrated Trajectory (HY-SPLIT) code [*Draxler*, 1992] from NOAA.

3. Results

[11] The vertical profiles of air temperature and relative humidity on clear days (18, 23, 26, and 28 April) are shown in Figures 3a and 3b, respectively. An inversion layer was often found at the altitude of ∼1 km in the temperature profiles (Figure 3a). The inversion was weak on 18 April. The relative humidity in the boundary layer was often found

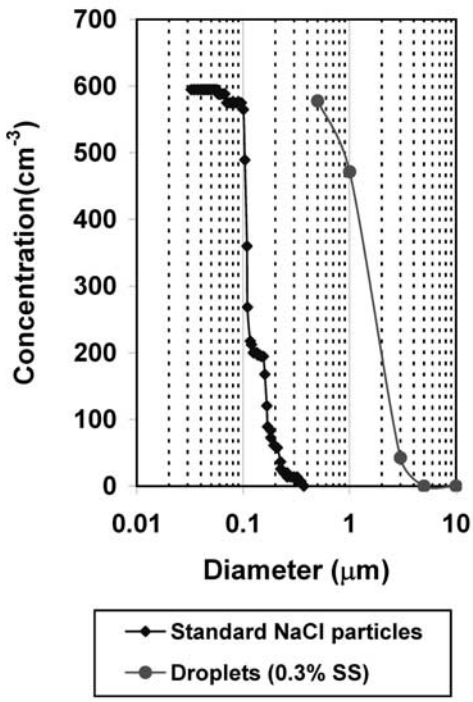


Figure 2. Cumulative size distributions of standard NaCl particles introduced in the cloud condensation nuclei (CCN) counter and droplets formed in the cloud chamber for its calibration. Diamonds represent aerosol particles measured with a scanning mobility particle sizer. Circles represent the concentration of droplets formed in the cloud chamber at 0.3% supersaturation (SS).

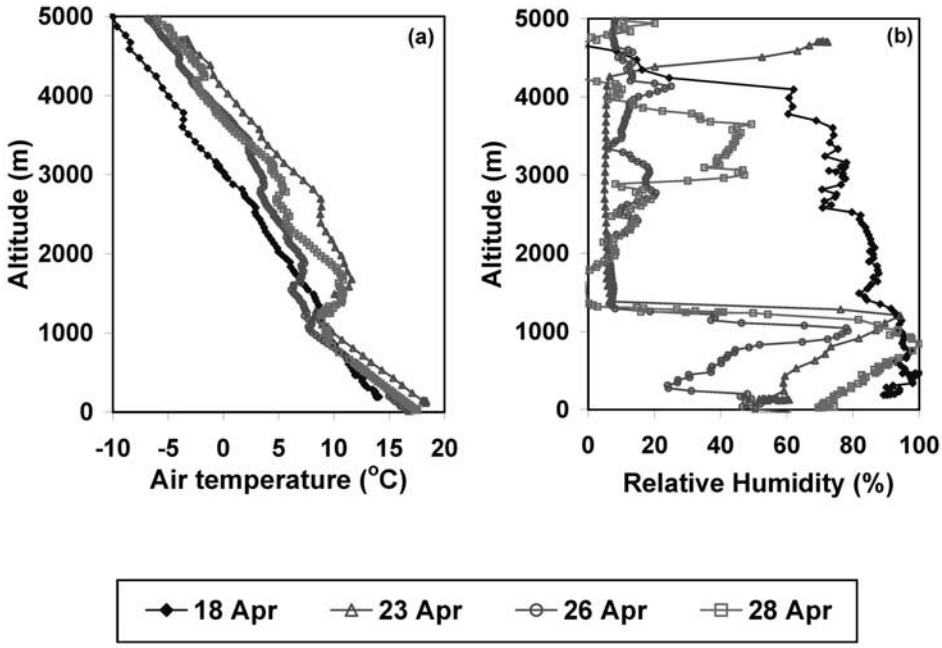


Figure 3. Vertical profiles of (a) air temperature and (b) relative humidity on clear days (18, 23, 26, and 28 April 2001).

to be significantly different from that in the free troposphere in the maritime atmosphere [*Chuang et al.*, 2000]. As expected, the relative humidity was higher than ~80% in the boundary layer and was reduced to 20% above the boundary layer on 23, 26, and 28 April (Figure 3b). There was much less relative humidity difference between the boundary layer and the free troposphere on 18 April, as the relative humidity was high (~60%) above the boundary layer.

3.1. Vertical Measurements of SO_2, Aerosol Particles, and CCN Concentrations on the Clear Days

[12] It is well known that pollutants from continents can be transported several thousand kilometers over the oceans. During this long-range transport they can affect the radiative balance directly and indirectly. *Akimoto and Narita* [1994] and *Hatakeyama et al.* [1995] noticed high concentrations of SO_2 under the outflow of anthropogenic pollutants from the Asian continent. Likewise, *Osada et al.* [2002] suggested high concentrations of sulfate particles in the free troposphere over Japan. Hence to characterize the distributions of aerosol particles and CCN in the east Asia region, vertical observations were performed on clear days over the ocean near the southwest islands of Japan. The observational data showed that the total aerosol concentrations ($d >$ 0.003 μm, not shown) were higher than 3000 cm^{-3} on 23 and 28 April. The concentrations were ~6000 cm^{-3} near the sea surface, and they increased drastically to ~30,000 cm^{-3} near the top of the boundary layer on 26 April. In order to distinguish the clean atmosphere from the polluted one, vertical distributions of concentrations of NP ($0.12 > d > 0.01$ μm), FP ($1.05 > d > 0.12$ μm), and CP ($3.11 > d > 1.05$ μm) are plotted in Figures 4a, 4b, and 4c, respectively. It was generally seen that the NP and FP concentrations were high in the boundary layer and low above the boundary layer. The NP concentrations varied from 1200 to 3600 cm^{-3} near the surface on 23, 26, and 28 April. The concentrations were as high as ~17,000 cm^{-3} near the top of the boundary layer on 26 April. The TSI data were not available on 18 April. The vertical profiles of FP concentrations on 18 April showed a slight decrease in concentrations with height in agreement with those presented by *Hudson and Frisbie* [1991] for the clean marine environments off the coast of California. On the other hand, FP concentrations were lower near the sea surface and increased to a maximum at 200 m altitude on 26 and 28 April. The concentrations were almost constant in the boundary layer on 23 April. The peak concentrations of FP were <500 cm^{-3} on 18 April, but peak concentrations were 2–4 times higher on the other days (23, 26, and 28 April). In contrast, CP peak concentrations were ~30 cm^{-3} on 18 April and <2 cm^{-3} on 23, 26, and 28 April. Strong surface winds may actively produce sea-salt particles by the bubble-bursting process [*Blanchard and Syzdek*, 1972]. As per *Moore* [1952] and *Woodcock* [1953] the number and size of sea-salt particles in the air near the sea surface increases with the increase in wind speed. The wind speed near the sea surface was ~6 m s^{-1} on 26 and 28 April, but it was ~17 m s^{-1} on 18 April. Therefore it is reasonable to suppose that the high concentrations of CP on 18 April were due to the sea-salt particles introduced from the sea surface. The results described above indicate that the observation area was covered by relatively clean maritime atmosphere on 18 April and by more polluted atmosphere on 23, 26, and 28 April.

[13] Similarly, from the observation of SO_2 concentration it was also inferred that the marine atmosphere was often polluted by east Asia. The SO_2 concentration was ~0.3 ppb

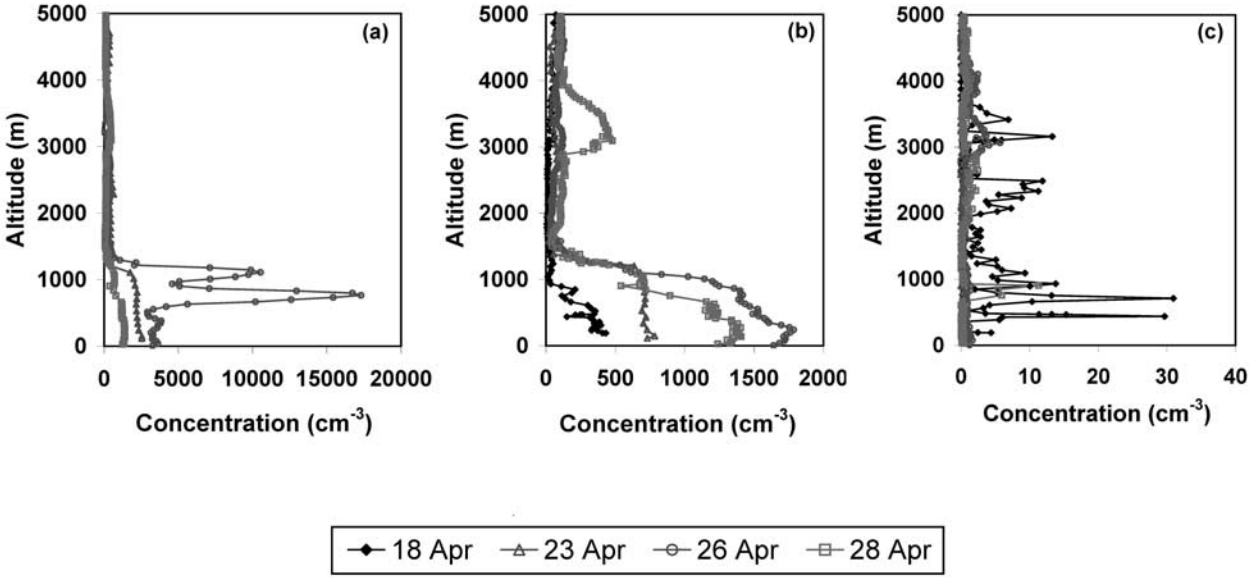


Figure 4. Vertical distributions of (a) nuclei mode ($0.12 > d > 0.01$ μm), (b) fine ($1.05 > d > 0.12$ μm), and (c) coarse ($3.11 > d > 1.05$ μm) particle concentrations observed during descending flights over the sea on clear days.

in the boundary layer on 18 April, whereas the concentration was ~1–5 ppb near the surface on 23 and 28 April (Figure 5). On 26 April the concentration was 2 ppb near the surface and increased to more than 10 ppb (beyond the scale limit) near the top of the boundary layer. These extremely high peak concentrations of SO_2 and aerosol particles (UP and NP) were observed near the top of the boundary layer on 26 April. The concentrations of SO_2 and aerosol particles were associated with the changes in air trajectories. In order to infer the sources of the polluted air on 26 April, back trajectories were computed for the final levels of 500, 1000, and 3000 m altitude over the observation areas using the HY-SPLIT code [*Draxler*, 1992] from NOAA. As expected, the back trajectories showed that the air resided over the Pacific Ocean for the previous 3 days prior to arrival at the observation area on 18 April. On the other hand, the air passed through the eastern part of Japan on 28 April and through Mongolia, China, Korea, and Japan on 23 and 26 April for the 3 days prior to arrival at the observation area. Sources of anthropogenic sulfate and its precursor (SO_2) are located near the coastal area of the Yellow Sea and near the rim of the west Pacific Ocean, including Japan. *Hatakeyama et al.* [1995] observed high concentrations of SO_2 over the China Sea, Yellow Sea, and Sea of Japan under the outflow of anthropogenic pollutants from the Asian continent. Further, *Akimoto and Narita* [1994] concluded that the coastal area of the Yellow Sea is the dominant source region of anthropogenic SO_2. Active volcanoes, Sakurajima (31.58°N, 130.63°E) and Miyakejima (34.07°N, 139.55°E) of Japan, also emit huge amounts of SO_2. Further, *Satsumabayashi et al.* [1999] and *Katsuno et al.* [2002] observed high concentrations of SO_2 and particulate SO_4^{2-} at Mount Happo caused by volcanic gas from Sakurajima and Miyakejima volcanoes. It was seen from back trajectory analysis that the air passed through the coastal area of China (anthropogenic sources) and the area of Sakurajima in Kyushu on 26 April. On the other hand, the air passed through the area of anthropogenic sources in Japan and Miyakejima in the west Pacific Ocean during their transport on 28 April and through anthropogenic sources in China on 23 April. On 18 April the air passed through neither the area near anthropogenic sources nor the active volcanoes. It is considered from the analytical results described above that the high concentrations of SO_2 and aerosol particles were

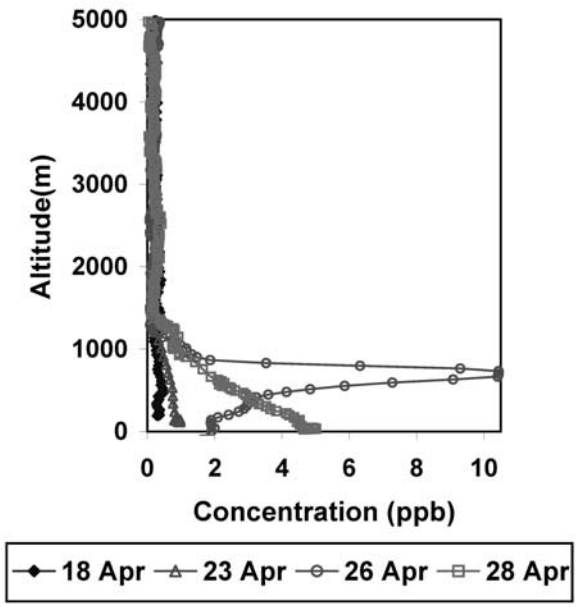


Figure 5. Vertical distributions of SO_2 concentration on clear days.

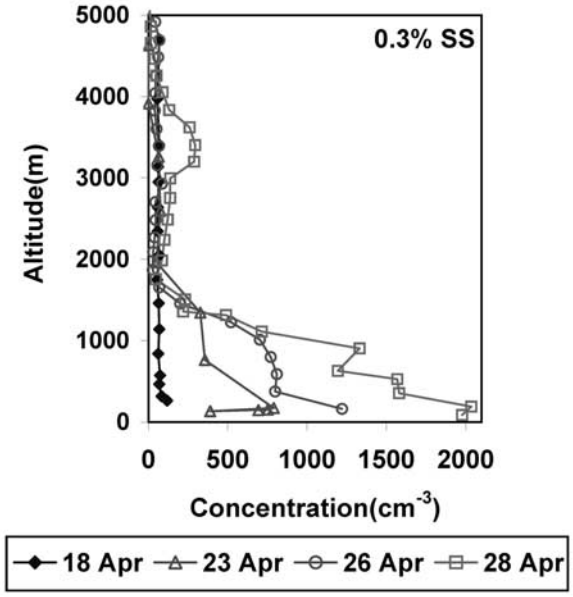


Figure 6. Vertical distributions of CCN activated at 0.3% SS on clear days.

due to transport of anthropogenic and/or volcanic pollutants on 26 and 28 April and due to anthropogenic pollutants on 23 April. Concentrations of SO_2 and aerosol particles were comparatively low on 18 April, because the observation area was covered with the relatively clean maritime atmosphere.

[14] Vertical profiles of CCN activated at 0.3% supersaturation over the sea near the southwest islands area are plotted in Figure 6. The vertical profile shows that the concentrations of CCN in the boundary layer were very high (about 750–2200 cm^{-3}) on 23, 26, and 28 April and comparatively low (120 cm^{-3}) on 18 April. Although CCN concentration on 18 April was relatively low, it was somewhat high compared with previous measurements in the background maritime atmosphere. Significant variations of CCN concentrations had been observed in the maritime atmosphere. *Twomey* [1959] and *Twomey and Wojciechowski* [1969] reported average CCN concentrations of 40–200 cm^{-3} at 0.3% SS over the main oceans of the world. The airborne measurements of CCN observed subcloud CCN concentrations of 88 cm^{-3} at 0.4% SS in clean air and 661 cm^{-3} in polluted air in the vicinity of the Azores Islands in the northeast Atlantic Ocean [*Hudson and Li*, 1995]. Similarly, *Matsumoto et al.* [1998] suggested that the CCN activated at 1.0% SS were 30–150 cm^{-3} and those at 0.5% SS were 10–100 cm^{-3} for marine background conditions, and they also suggested that the activated CCN were 150–1000 cm^{-3} at 1.0% and 100–500 cm^{-3} at 0.5% SS under the continentally and/or anthropologically affected conditions at Hahajima, the Ogasawara Islands, in the northwest Pacific Ocean. *Pawlowska and Brenguier* [2000] observed ~62–383 cm^{-3} at 0.5% SS over the sea between Portugal and the Canary Islands. *Hudson and Yum* [2001] presented the CCN concentration of ~150 cm^{-3} at 0.1% SS and 359 ± 142 cm^{-3} at 1% SS for modified maritime air in the eastern Florida. It is inferred from the above results that CCN concentrations in the background marine atmosphere are about 10–100 cm^{-3} at 0.5% SS and 30–150 cm^{-3} at 1.0% SS, although it may depend on wind speed, location, and meteorological conditions [*Moore*, 1952; *Woodcock*, 1953]. However, the observation data suggested that the CCN concentrations in the marine atmosphere were increased under the inflow of continental pollutants. Many researchers sometimes classified CCN concentrations measured in the marine atmosphere into groups: the clean case, the modified case, and the polluted case. On the basis of the CCN concentrations in this study the atmosphere on 18 April was considered to be the maritime atmosphere modified slightly by continental pollutants (hereinafter the atmosphere on 18 April is termed the relatively clean maritime rather than the clean maritime). The atmosphere on the other days was considered to be maritime strongly influenced by continental pollutants originating from east Asia. Rather high concentrations of CCN in the relatively clean maritime atmosphere in addition to polluted atmosphere suggested that the air pollutants significantly contributed to the CCN over the sea near the southwest islands of Japan.

3.2. SO_2, Aerosol Particles, and CCN Concentrations Under the Cloud Bases

[15] The observed results described in section 3.1 demonstrated that the atmosphere was mostly covered by polluted air with high concentrations of aerosol particles, CCN, and SO_2 over the sea near the southwest islands of Japan. It is also known that the microphysical properties of clouds depend strongly upon the characteristics of air parcels under the cloud base [e.g., *Brenguier et al.*, 2000; *Hudson and Yum*, 2001]. To examine the air parcels, horizontal flights were made under cloud bases where concentrations of SO_2, aerosol particles, and CCN were measured on 21, 22, 24, and 27 April. The distances between cloud bases and horizontal flights under cloud bases were ~110, 270, 190, 130, and 190 m on 21, 22, 24, 27A, and 27B April, respectively. The CN concentrations were ~1000 cm^{-3} on 24 April and twofold higher on the other days (21, 22, and 27B April; CN data were not available on 27A) (Figure 7). SO_2 concentration in the boundary layer was ~0.3 ppb on 21, 22, and 24 April, but it was as high as 2 ppb on 27 April (Figure 7). From the CN concentrations it should also be noted that the observation area was covered with polluted air on 21, 22, and 27 April and by relatively clean maritime air on 24 April. Moreover, CCN concentrations were ~170 cm^{-3} on 24 April and ~430–700 cm^{-3} on the other days. Hence, as described in section 3.2, the atmosphere on 24 April was considered relatively clean maritime, and the atmosphere on the other days (21, 22, and 27) was polluted. Relatively low concentrations of CCN in the relatively clean maritime atmosphere and high concentrations of CCN in the polluted atmosphere suggested that pollutants greatly contributed to CCN under the cloud bases, which might affect microphysical properties of the clouds.

3.3. Microphysical Properties of Clouds

[16] As observed in sections 3.1 and 3.2, CCN concentrations were low in the relatively clean maritime and high in the polluted atmosphere. On the basis of the subcloud CCN concentrations it was considered that clouds on

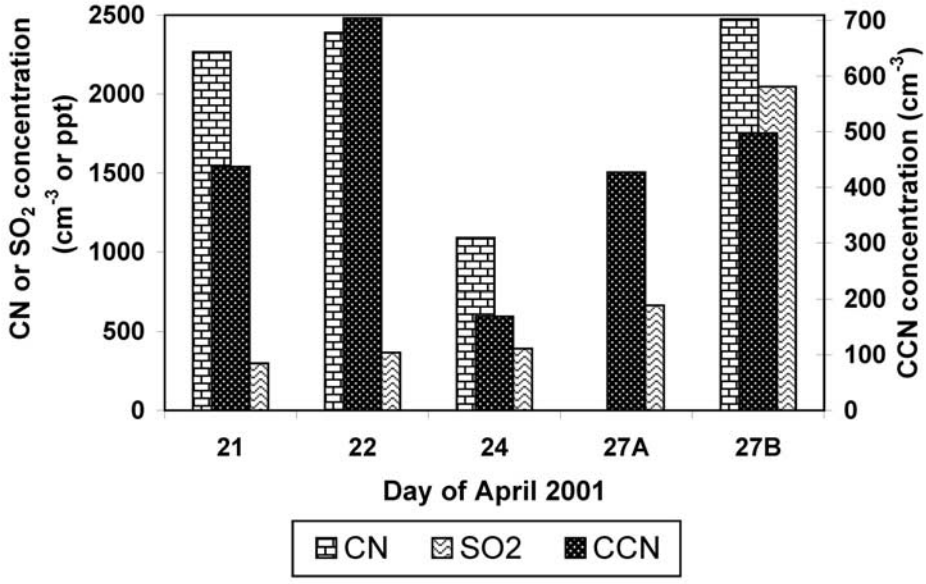


Figure 7. Average concentrations of SO_2, condensation nuclei particles (CN) ($d > 0.01$ μm), and CCN (0.3% SS) under the cloud bases on 21, 22, 24, and 27 April 2001. CN data are not available on 27A April.

24 April were relatively clean maritime and clouds on 21, 22, and 27 April were polluted. *Yum et al.* [1998] and *Hudson and Yum* [2001] suggested that the concentrations of cloud droplets depend on the concentrations of CCN entering the clouds through the bases and the updraft velocity of air parcels. In order to distinguish the microphysical properties of the relatively clean maritime and polluted clouds the droplet concentrations were measured at various levels. The effective radius and liquid water content were derived from the size distributions of cloud droplets. Vertical profiles of mean cloud droplet concentrations, effective radius of cloud droplets (r_e), and LWC

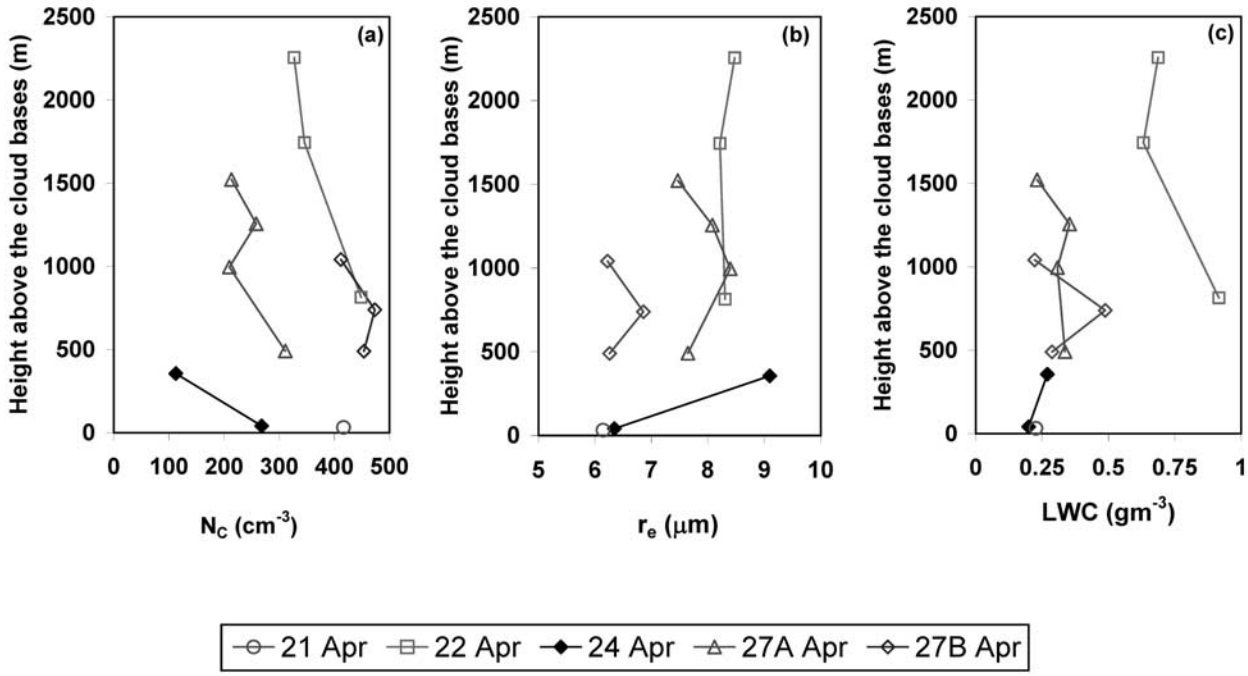


Figure 8. Vertical distributions of (a) cloud droplet concentration (N_C), (b) effective radius (r_e), and (c) liquid water content (LWC) on 21, 22, 24, and 27 April 2001. The *y* axis represents height above the cloud bases. Each symbol represents the average value of a level flight in the clouds.

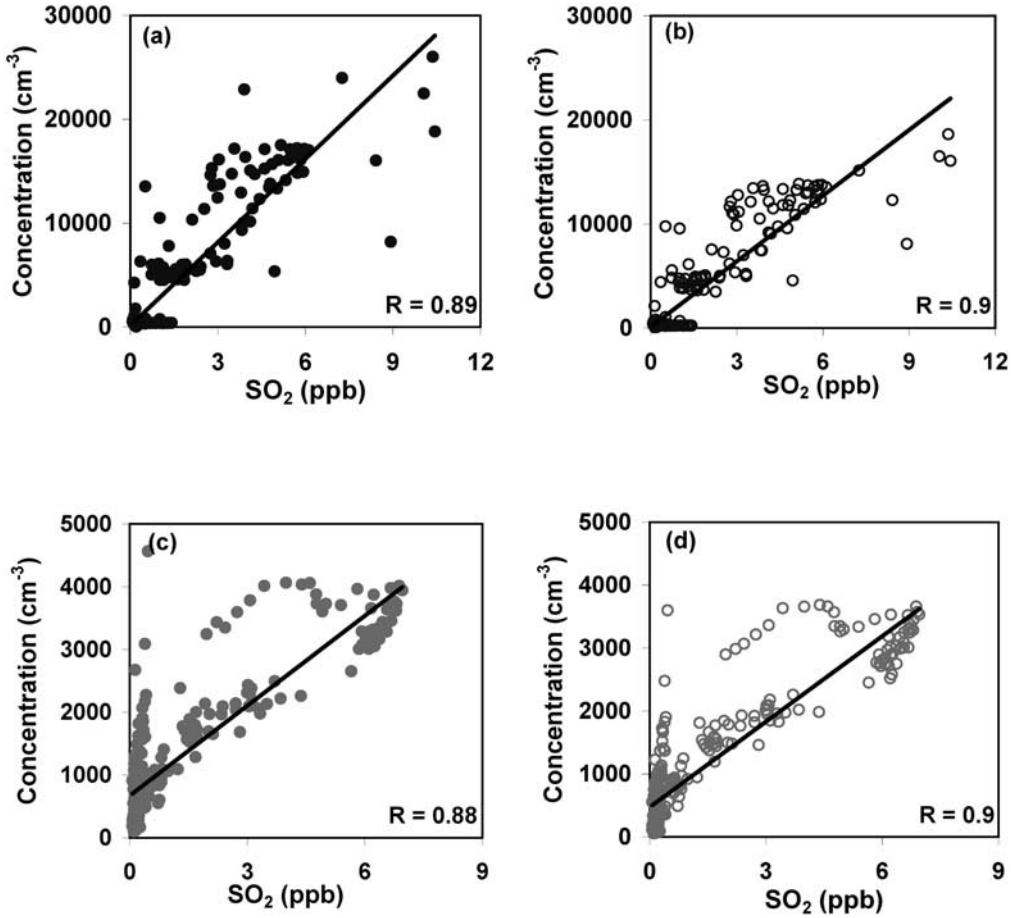


Figure 9. Relationship between SO_2 and aerosol particles (a) $d > 0.003$ μm and (b) $d > 0.01$ μm on 26 April and (c) $d > 0.003$ μm and (d) $d > 0.01$ μm on 28 April 2001.

obtained from horizontal measurements are plotted in Figure 8. The y axis in Figure 8 represents the height above cloud bases. Each symbol represents average values at the cloud penetration level. The lowest cloud penetration levels on 21, 22, 24, 27A, and 27B were ~30, 800, 40, 500, and 500 m from the cloud bases, respectively. Consequently, it was observed that the mean cloud droplet concentrations (N_C) were relatively low (270 cm^{-3}) in the relatively clean maritime clouds on 24 April and high (~420 cm^{-3}) in the polluted clouds on 21 April near the cloud bases and comparatively high (~310–450 cm^{-3}) at the height of ~500–800 m from the cloud bases in the polluted clouds on 22, 27A, and 27B April (Figure 8a). It was observed that the average cloud droplet concentrations were 150 ± 49 cm^{-3} in maritime clouds and 312 ± 68 cm^{-3} in continental clouds during summertime flights in eastern Florida [*Hudson and Yum*, 2001]. Cloud droplet concentrations higher than ~400 cm^{-3} were also observed by *Brenguier et al.* [2000] and *Costa et al.* [2000] in polluted clouds. The cloud droplet concentration in the relatively clean maritime clouds in this study was somewhat high compared to that (55 cm^{-3}) in the clean atmosphere and almost comparable to that (244 cm^{-3}) in the polluted atmosphere in ACE-2 [*Pawlowska and Brenguier*, 2000]. The results suggest that even the relatively clean maritime atmosphere over the sea near the southwest islands of Japan contains some anthropogenic/continental components.

[17] It is often observed in typical lower clouds that N_C was almost constant with increasing altitude above the cloud bases, whereas LWC and r_e increased with increasing altitude [e.g., *Slingo et al.*, 1982; *Ishizaka et al.*, 1995]. Vertical distributions of N_C, r_e, and LWC in clouds shown in Figure 8 were a little different from those observed in typical lower clouds. Mean LWC at ~500–800 m above the cloud bases on 22, 27A, and 27B April was around 0.5 of adiabatic LWC value, although LWC of the central parts in the clouds was higher than ~0.8 of adiabatic LWC (Y. Ishizaka et al., The effect of CCN around the clouds on their microphysical properties—Aircraft observation over the sea near the southwest islands area in Japan, submitted to *Atmospheric Research*, 2004, hereinafter referred to as Ishizaka et al., submitted manuscript, 2004). These results suggested that microphysical properties of clouds at levels higher than ~500 m above the cloud bases were considerably influenced by microphysical processes such as inhomogeneous mixing, entrainment, in-cloud scavenging, and/or drizzle formation [*Hudson and Frisbie*, 1991; *Pawlowska and Brenguier*, 2000; Ishizaka et al., submitted manuscript, 2004]. At ~50 m altitude (30 m on

Table 1. Relationship Between SO_2 and Aerosol Particle Concentrations on 18, 23, 26, and 28 April 2001

	Correlation Coefficient (r)[a]			
Size Range of Aerosol Particles, μm	18 April	23 April	26 April	28 April
Ultrafine particles ($0.01 > d > 0.003$)	...	0.36	0.71	0.33
Nuclei mode particles ($0.12 > d > 0.01$)	...	0.76	0.89	0.92
Fine particles ($1.05 > d > 0.12$)	0.1	0.76	0.71	0.81
$d > 0.003$	...	0.69	0.89	0.87
$d > 0.01$	...	0.78	0.90	0.90
$d > 0.12$	0.1	0.76	0.71	0.81

[a]SO_2 concentration. Data points, $n > 250$.

21 April and 40 m on 24 April) the LWC was almost similar in the relatively clean maritime cloud on 24 April and in the polluted clouds on 21 April. Similarly, at ~500 m altitude, LWC was almost similar in the polluted clouds on 27A and 27B April. The cloud droplet concentration was higher, but r_e was significantly smaller in the polluted clouds than in the relatively clean maritime clouds at ~50 m altitude (Figure 8b). Further, it is worth noting that the cloud droplet concentrations were higher but r_e was smaller in the polluted clouds on 27A and 27B April than in the relatively clean maritime clouds on 24 April at ~500 m altitude.

4. Discussion

4.1. Relation Among SO_2, Aerosol Particles, and CCN

[18] The relationship between SO_2 and aerosol particle ($d > 0.003$ and $d > 0.01$ μm) concentrations on 26 and 28 April is plotted in Figure 9. Figure 9 shows excellent correlation coefficients for both 26 and 28 April. The linear correlation coefficients obtained for different size ranges are summarized in Table 1. The correlation coefficient is excellent for NP and FP concentrations but poor for UP concentrations on 28 April. On the other hand, the correlation is excellent for NP concentrations and good for FP concentrations as well as UP concentrations on 26 April. Backward trajectories showed that the air took ~54 hours to move from Japan (anthropogenic sources) to the observation area on 28 April. It took ~48 hours to move from the Yellow Sea coast of China (anthropogenic sources) and 12 hours to move from the area of Sakurajima volcano to the observation area on 26 April. It was observed that sulfates such as ammonium sulfate contributed considerably to the CCN at different locations in Japan [e.g., *Ishizaka and Adhikari*, 2003]. Further, it was suggested that stagnant airflow around the coastal area of the Yellow Sea and around Japan provides suitable conditions for conversion from SO_2 to fine sulfate during its transport [*Osada et al.*, 2003]. Similarly, *Hayasaka et al.* [1990] had observed that the submicron aerosol particles consist mainly of the secondary particles such as sulfate and nitrate particles produced through gas-to-particle conversion and coagulation processes in the boundary layer around Japan. Further, chemical composition of atmospheric aerosols measured by *Ohta et al.* [2001] at Amami-Ohshima Islands in APEX-E2 found that the sulfate content was about 30–50% by weight in fine and coarse particles. It is considered from their observational data that sulfate was a major component of the aerosol particles and contributed considerably to the CCN concentrations in the spring season in 2001 over the sea near the southwest islands area. From high correlation coefficients between SO_2 and NP and/or FP concentrations and back trajectory it might be reasonable to suppose that most of the sulfate in submicron particles originated from anthropogenic SO_2 over the sea near the southwest islands area.

[19] The observational data suggested that CCN concentrations in the boundary layer were strongly related to aerosol particle concentrations. It was also observed that high concentrations of CCN were associated with the pollutants over the sea near the southwest islands of Japan. To examine the relationship between concentrations of aerosol particles and CCN in the relatively clean maritime and polluted atmospheres, vertical distributions of the ratios of CCN (N_{CCN}) to CN (N_{CN}, $d > 0.01$ μm) concentrations and CCN (N_{CCN}) to AP (N_{AP}, $3.11 > d > 0.12$ μm) concentrations are plotted in Figures 10a and 10b, respectively. Figures 10a and 10b show that the ratio (N_{CCN}/N_{CN} and/or N_{CCN}/N_{AP}) was mostly higher in the boundary layer than above the boundary layer in the polluted atmosphere. In contrast, the ratio of N_{CCN}/N_{AP} was higher above the boundary layer than in the boundary layer in the relatively clean maritime [*Hudson and Xie*, 1999] (Figure 10b). The relationship between CCN (N_{CCN}) and AP (N_{AP}, $3.11 > d > 0.12$ μm) is also shown in Figure 11. The slopes of N_{CCN}/N_{AP} are almost similar in the relatively clean maritime and polluted atmospheres, but the slope on 28 April is significantly higher than the others ($\sim 1.0 > N_{CCN}/N_{AP} > 0.2$). In order to presume hygroscopic characteristic of aerosol particles in the polluted atmosphere on 28 April the ratios of N_{CCN}/N_{CN} and N_{CCN}/N_{AP} on 28 April were compared with those in the relatively clean atmosphere on 18 April. N_{CCN}/N_{CN} and N_{CCN}/N_{AP} were <0.3 and ~0.3 on 18 April and ~0.5 and ~1.5 on 28 April, respectively. The ratios strongly suggest that aerosol particles ($d > 0.05$ μm) activated as CCN were more soluble in the polluted atmosphere on 28 April than in the relatively clean maritime atmosphere on 18 April. It is inferred from the results that a possibly heavy loading of air pollutants increases number concentration of hygroscopic fine particles and CCN concentrations.

4.2. Contribution of AP to CCN

[20] CCN concentration measured at 0.5% supersaturation at Cape Grim, Tasmania, during the First Aerosol Characterization Experiment suggested that the aerosol particles of $0.2 > d > 0.08$ μm in diameter contributed 71% of the CCN. Larger sea-salt ($d > 0.2$ μm) and Aitken ($0.07 > d > 0.02$ μm) mode particles contributed 16 and 13%, respectively, to CCN in the baseline marine air mass [*Covert et al.*, 1998]. While the aerosol had been influenced by anthropogenic sources or local biomass

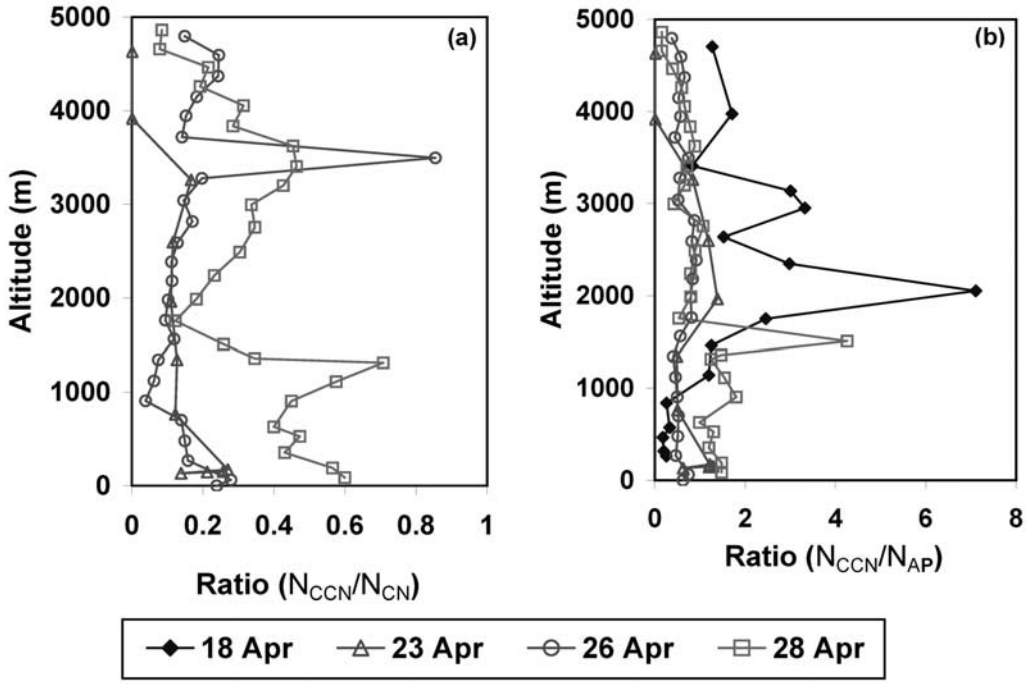
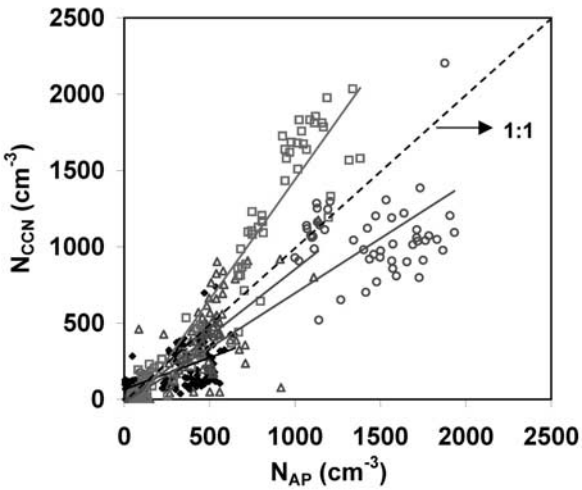


Figure 10. Vertical profiles of ratio of (a) N_{CCN} to N_{CN} ($d > 0.01$ μm) and (b) N_{CCN} to N_{AP} ($3.11 > d > 0.12$ μm) on clear days.

burning, the contributions were 80, 6, and 14%. Similarly, *Intergovernmental Panel on Climate Change* [2001] reported that accumulation mode ($1 > d > 0.1$ μm) aerosol particles form the majority of CCN. It is also known that the accumulation mode aerosol particles have the longest atmospheric lifetime. Further, *Schwartz and Slingo* [1996] concluded that aerosol particles in the accumulation mode modify microphysical and optical properties of clouds and cloud lifetime. In addition, to get useful information for remote sensing data analysis, it is important to know the contribution of AP to CCN in the east Asia region. The relationship between AP and CCN concentrations showed that CCN concentrations increased with increasing AP concentrations in the polluted atmosphere. For heavy aerosol loading ($N_{AP} > 500$ cm^{-3}), CCN concentrations were substantially high. Moreover, the slope of N_{CCN}/N_{AP} is 1.57 on 28 April, 0.88 on 23 April, and 0.72 on 26 April in the polluted atmosphere but as low as 0.42 in the relatively clean maritime atmosphere (Figure 11).

[21] In order to compare the ratio of N_{CCN}/N_{AP} in the east Asia region with those of previous studies the data from APEX-E1 (clear-sky measurements during 15–24 December 2000 over the sea near the southwest islands area in Japan [*Adhikari et al.*, 2001]) and APEX-E2/ACE-Asia (present observations) were analyzed. The slopes in this study and other previous studies are summarized in Table 2. *Raga and Jonas* [1995] determined the log slope (0.44) between the aerosol particles ($3.0 > d > 0.1$ μm) and CCN (SS = 0.85–0.95%). They reported that on one of the polluted days the CCN concentration was lower than aerosol particle concentration because of the presence of a large fraction of carbon from gas/oil rigs. *Hegg et al.* [1996] suggested that CCN concentration at 1% SS was only 10% of the number of dry particles with diameter larger than



	Correlation coefficient (R)	Slope (N_{CCN}/N_{AP})
18 Apr	0.62	0.42
23 Apr	0.83	0.88
26 Apr	0.93	0.72
28 Apr	0.96	1.57

◆ 18 Apr △ 23 Apr ○ 26 Apr □ 28 Apr

Figure 11. (top) Relationship between CCN and aerosol particle ($3.11 > d > 0.12$ μm) concentrations in April 2001. (bottom) Summary of slope and correlation coefficient.

Table 2. Relationship Among N_{AP}, N_{CCN}, and N_C of Previous and Present Studies[a]

Author	Relationship	Notes
	N_{CCN} Versus N_{AP}	
Raga and Jonas [1995]	$N_{CCN} \sim N_{AP}^{0.44}$	N_{CCN} (0.9% SS), N_{AP} ($3.0 > d > 0.1$ μm)
Hegg et al. [1996]	$N_{CCN} \sim 0.1N_{AP}$	Arctic location, N_{CCN} (1% SS), N_{AP} ($d > 0.003$ μm)
Chuang et al. [2000]	$N_{CCN} \sim N_{AP}^{0.63}$	N_{CCN} (0.1% SS), N_{AP} ($d > 0.1$ μm)
This study[b]	$N_{CCN} \sim 0.75N_{AP}$	N_{CCN} (0.3% SS), N_{AP} ($d > 0.1$ μm)
	N_C Versus N_{AP}	
Kaufman et al. [1991]	$N_C \sim N_{AP}^{0.7-0.8}$	derived from Twomey's formula
Jones et al. [1994]	$N_C \sim N_{AP}^{0.26}$	data from *Martin et al.* [1994]
Nakajima et al. [2001]	$N_C \sim N_{AP}^{0.5}$	satellite (AVHRR) derived
Hudson and Yum [2002]	$N_C \sim 0.12N_{AP}$	N_C ($50 > d > 2$ μm), N_{AP} ($d > 0.01$ μm)
This study	$N_C \sim N_{AP}^{0.39}$	N_C ($47 > d > 2$ μm), N_{AP} ($d > 0.1$ μm)
	N_C Versus N_{CCN}	
Hegg et al. [1991]	$N_C \sim 0.71N_{CCN}$	marine stratus, 1% SS
Vong and Covert [1998]	$N_C \sim 0.60N_{CCN}$	N_{CCN} [N_{AP} ($d > 0.08$ μm)], LWC > 0.05 g m^{-3}
Chuang et al. [2000]	$N_C \sim 0.71N_{CCN}$, $N_C \sim N_{CCN}^{0.31}$	stratus cloud, N_{CCN} (0.1% SS)
Hudson and Yum [2001]	$N_C \sim 0.16-0.23\ N_{CCN}$	N_C ($50 > d > 2$ μm), N_{CCN} (1% SS)
Hudson and Yum [2002]	$N_C \sim 0.40\ N_{CCN}$	N_C ($50 > d > 2$ μm), N_{CCN} (0.1% SS)
Hudson and Yum [2002]	$N_C \sim 0.19\ N_{CCN}$	N_C ($50 > d > 2$ μm), N_{CCN} (1% SS)
Yum and Hudson [2002]	$N_C \sim 0.38\ N_{CCN}$	N_C ($50 > d > 2$ μm), N_{CCN} (0.2% SS)
Yum and Hudson [2002]	$N_C \sim 0.21\ N_{CCN}$	N_C ($50 > d > 2$ μm), N_{CCN} (0.4% SS)
Yum et al. [1998]	$N_C \sim 0.38\ N_{CCN}$	N_C ($50 > d > 2$ μm), N_{CCN} (0.2% SS)
Yum et al. [1998]	$N_C \sim 0.54\ N_{CCN}$	N_C ($50 > d > 2$ μm), N_{CCN} (0.4% SS)
This study	$N_C \sim 0.37\ N_{CCN}$, $N_C \sim N_{CCN}^{0.39}$	N_C ($47 > d > 2$ μm), N_{CCN} (0.3% SS)

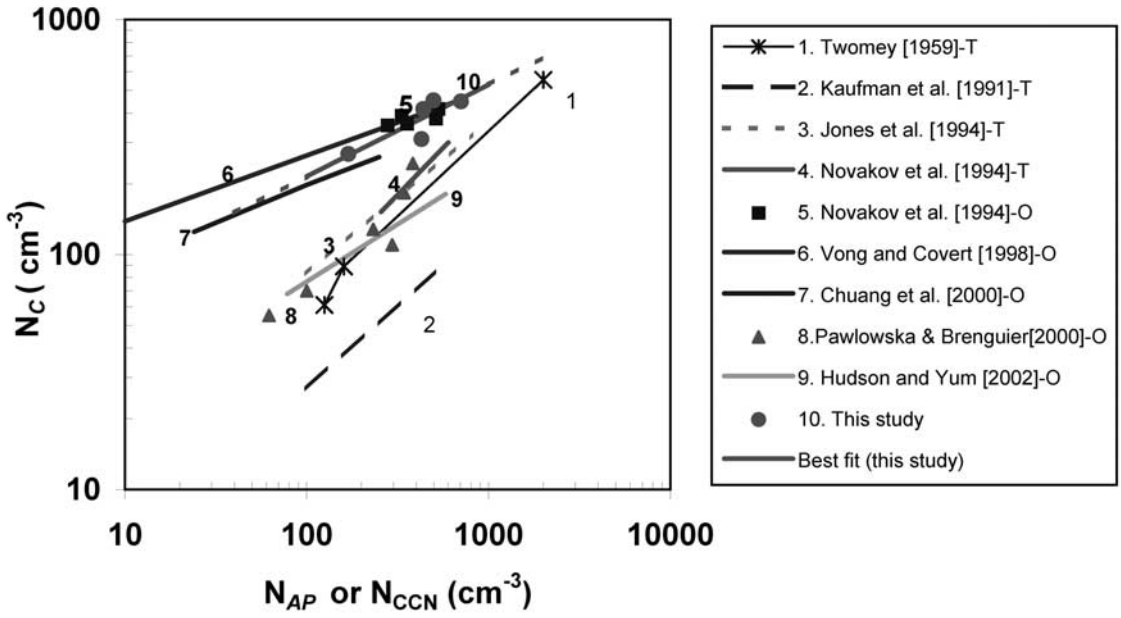
[a]AP, atmospheric particles; AVHRR, advanced very high resolution radiometer; LWC, liquid water content.
[b]Data taken from Asian Atmospheric Particulate Environmental Change Experiment 1 (APEX-E1) (15–24 December 2000) and APEX-E2/Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) (18–28 April 2001) over the sea near the southwest islands area in Japan.

3 nm. On the other hand, *Chuang et al.* [2000] measured aerosol particles larger than 0.1 μm in diameters and CCN at 0.1% SS. They obtained the log slope of 0.63. In this study, N_{AP} is the number concentration of aerosol particles larger than 0.1 μm in diameter, and N_{CCN} is the CCN concentration activated at 0.3% SS. The slope (0.75) of this study was higher than that reported by *Raga and Jonas* [1995] and *Hegg et al.* [1996], but it was comparable to the slope given by *Chuang et al.* [2000]. Hence it is considered that the slope of N_{CCN}/N_{AP} was comparatively high over the sea near the southwest islands of Japan as compared with other areas.

4.3. Effect of Pollutant on Microphysical Properties of Clouds

[22] As described in section 3, the cloud droplet concentrations, effective radius, and LWC were significantly changed with increasing height in both the relatively clean maritime and polluted clouds. *Brenguier et al.* [2000] suggested that the droplet concentrations fixed at the activation level are likely modified farther up by entrainment and mixing processes as well as by drizzle scavenging. They also suggested that the LWC increases adiabatically with height above the cloud bases. *Slingo et al.* [1982] and *Pinto et al.* [2001] suggested that mean effective radius generally increases with height in stratiform clouds. However, *Yum and Hudson* [2001] summarized that entrainment reduces droplet concentrations and evaporation reduces droplet size above the cloud bases. Therefore it will be unrealistic to estimate effect of pollutants on microphysical properties of clouds at upper level or top of the clouds without considering the microphysical processes (e.g., entrainment, evaporation, and coalescence) in the clouds. Hence the cloud droplet concentrations, effective radius, and LWC observed in the lower levels of clouds were taken as important values in each cloud. Similarly, the average concentrations of CCN (N_{CCN}) and aerosol particles ($3.11 > d > 0.12$ μm) measured under the cloud bases were taken as the values for CCN (N_{CCN}) and aerosol particles (N_{AP}), respectively. The relationship between cloud droplet (N_C) and aerosol particle (N_{AP}) concentrations showed that the N_C was increased with increasing N_{AP}. Since the data points were few in numbers, the slope of present study was compared with previous studies (Table 2) to quantify the results. It was found that the log slope ($d\log N_C/d\log N_{AP}$) of the present study (0.39) was larger than the value given by *Jones et al.* [1994] but smaller than the range given by *Kaufman et al.* [1991]. The slope was quite consistent with the value derived from model calculation using satellite data by *Nakajima et al.* [2001].

[23] It is shown in section 4.1 that only a small fraction of aerosol particles serve as CCN at 0.3% supersaturation. Similarly, *Hegg et al.* [1991] and *Gras* [1987, 1989] suggested that, on average, less than half of the CN (400% SS) in the marine atmosphere serve as CCN active at the 1% SS in marine stratiform clouds. Therefore, as a better approach for estimating effect of pollutants on microphysical properties of cloud the relationship between N_{CCN} and N_C is plotted in Figure 12. The solid circles in Figure 12 represent measured data points in this study. The N_C is increased with increasing concentration of N_{CCN}. The line of best fit (solid line) with a correlation coefficient (R) of 0.79 provides the quantitative support for a relationship between N_{CCN} and N_C. The linear slope of this study (0.37, Table 2) was smaller than the slope obtained by *Hegg et al.* [1991], *Vong and Covert* [1998], and *Chuang et al.* [2000]. It was notable that the slope of 0.37 for 0.3% SS was between the values of 0.40 and 0.2 for 0.1 and 1.0% SS



	Authors	N_{AP} or N_{CCN}	N_C
1	Twomey [1959]-T	CCN (125, 160, 2000)	Twomey's formula ($N_C \sim N_{AP}^{2/2+k}$), w= 1m/s
2	Kaufman et al. [1991]-T	CCN (0.3%SS)	Twomey's formula, k= 0.8
3	Jones et al. [1994]-T	mean radius 0.05 μm	N_C=375(1-exp[-2.5e-3xN_{AP}]
4	Novakov et al. [1994]-T	CCN = 0.33xSO_4^{-2} + 166.23	Twomey's formula, k=0.5, w=1m/s
5	Novakov et al. [1994]-O	CCN = 0.33xSO_4^{-2} + 166.23	stratocumulus (d>1 μm)
6	Vong and Covert [1998]-O	CCN (d>0.08 μm)	ground measurement (47>d>2 μm),
7	Chuang et al. [2000]-O	CCN (0.1%SS)	47>d>2 μm
8	Pawlowska and Brenguier[2000]-O	CCN (0.5%SS)	$N_C = N_{mean}$ (LWC>0.9 LWC_{ad}).
9	Hudson and Yum [2002]-O	CCN (0.1%SS)	50>d>2 μm
10	This study	CCN (0.3%SS)	47>d>2 μm

Figure 12. (top) Relationship between CCN concentration (N_{CCN}) and cloud droplet concentration (N_C) in this study and relationship between CCN or atmospheric particles and cloud droplet concentrations measured or calculated with *Twomey*'s [1959] formula in previous studies. (bottom) Summary of conditions of measurements. Suffixes *T* and *O* in the legend represent theoretical and observed data of N_C, respectively.

given by *Hudson and Yum* [2002]. The slope and SS were both intermediate to the *Hudson and Yum* [2002] slopes and SS. Further, the slope was comparable to the slope of 0.38 given by *Yum and Hudson* [2002] and *Yum et al.* [1998] for 0.2% SS. Similarly, the log slope (0.39) was comparable to that given by *Chuang et al.* [2000].

[24] Moreover, to compare the result with previous studies, a relation between measured and theoretical data of N_{CCN} and N_C from previous studies is plotted in Figure 12. *Twomey* [1959] calculated the concentration of droplets formed at updraft velocity of 1 m s^{-1} for the maritime, modified maritime, and continental atmospheres by using Twomey's formula ($N_C \sim N_{CCN}^{2/2+k}$; k is the slope of the cumulative CCN spectrum from $N = CS^k$, where N is the concentration at a given supersaturation S, C is the concentration at 1% supersaturation, and k is the slope of the cumulative log-log distribution). He estimated the CCN concentration as 125, 160, and 2000 cm^{-3} in the maritime, modified maritime, and continental atmospheres, respectively, and derived supersaturation in clouds. Calculated values showed that the cloud droplet concentration increased with increasing CCN concentration (Figure 12). Later on, in order to assess the values and uncertainty in the parameter k, *Kaufman et al.* [1991] used some published measurements of k and CCN concentration at 0.3 and 1% SS. They used the average CCN concentration (N_{CCN} = 300 ± 200) for maritime and continental air mass for a supersaturation of 0.3% and k = 0.8 ± 0.15 to derive the cloud droplet concentration. Similarly, from a lognormal size distribution of aerosol particles, *Jones et al.* [1994] calculated the cloud droplet concentration using the equation given in Figure 12. Further, *Novakov et al.* [1994] presented theoretical results together with measured mean droplet concentrations for cumulus and stratocumulus clouds. Only two cases of *Novakov et al.* [1994] (one theoretical and one measured) are shown in Figure 12.

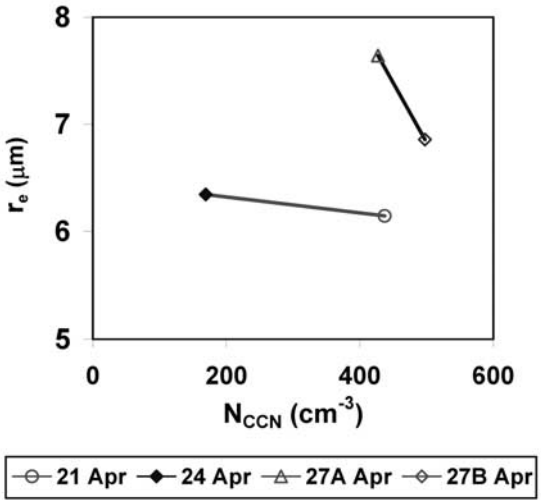


Figure 13. Relationship between CCN concentration and effective radius at similar LWC.

Their theoretical result, obtained with *Twomey*'s [1959] formula for the updraft velocity of 1 m s^{-1} and k parameter of 0.5, is shown by the dotted line in Figure 12. The measured value of N_C ($d > 1$ μm) in stratocumulus clouds, as a function of CCN concentration (at 0.5% SS) derived from mass concentration of non-sea-salt sulfate and a regression relationship, is shown by squares in Figure 12. *Novakov et al.* [1994] reported that the measured concentration of N_C was very high as compared to theoretical concentration. However, it is interesting to note that the data points from Novakov et al. are close and almost similar to data points of this study. *Vong and Covert* [1998] measured the cloud droplet and aerosol number concentration (as N_{CCN}) for high LWC (>0.25 g m^{-3}) and low LWC (0.25 > LWC > 0.05 g m^{-3}) in clouds at 10 m above the ground level on the coast of Washington State. They obtained the linear slope of 0.45 for low LWC. *Chuang et al.* [2000] derived the relation between cloud droplet and CCN concentrations under the cloud bases at 0.1% SS. The relationship between CCN (0.5% SS) and mean droplet concentrations for LWC > $0.9LWC_{adiabatic}$ measured during ACE-2 suggested that the droplet concentrations were dependent upon the aerosol properties within the boundary layer [*Pawlowska and Brenguier*, 2000]. Recently, *Hudson and Yum* [2002] found linear relationships between CCN and cloud droplet concentrations under various levels of pollution over the Indian Ocean. They obtained the linear slope of 0.40 between cloud droplets and CCN concentrations activated at 0.1% supersaturation. The slopes given by a few researchers were similar to each other, although the measured slope was lower than the theoretical one as shown in Figure 12. One possible explanation of lower slope is that the clouds are strongly affected by in-cloud microphysical processes (e.g., entrainment and inhomogeneous mixing) that can influence the response of cloud droplet concentrations to changes in CCN concentrations under the cloud bases. Although the measured slope was lower than the theoretical slope, N_C showed positive relation with N_{CCN} in all the cases. Ideally, CCN should be defined with respect to maximum supersaturation within natural clouds because these SS_{max} drastically change number concentration of the cloud droplets [*Hudson and Yum*, 2001]. SS in the cloud chamber may not be the same as SS in the real clouds. In the case of low supersaturation in clouds, only soluble and large aerosol particles activate to form cloud droplets. On the other hand, at high cloud supersaturation, even aerosol particles smaller and/or less soluble than those measured with a CCN counter can be activated to form cloud droplets in the atmosphere. In Figure 12, N_C is more often less than N_{CCN}, but in a few cases, N_C is greater than N_{CCN}. One of the possible causes of $N_C < N_{CCN}$ is that the SS in the atmosphere was lower than the SS in the cloud chamber. Another possible cause of $N_C < N_{CCN}$ may be microphysical process such as entrainment, mixing, coalescence, and/or drizzle formation. Similarly, $N_C > N_{CCN}$ is considered to be due to higher SS in the atmosphere than in the cloud chamber. Hence it is concluded from the above results that the cloud droplet concentration increased with increasing concentration of CCN under cloud bases.

[25] *Yum and Hudson* [2001] showed a positive relation between droplet concentration and mean diameter for the clouds with monotonically increasing liquid water content. In addition, *Pinto et al.* [2001] suggested that droplet size decreases with increasing droplet concentration but increases with increasing LWC. As described above, cloud droplet concentrations near cloud base increased with increasing CCN concentrations under cloud bases. Hence there is a possibility of effect of CCN concentrations on effective radius of cloud droplets near cloud bases. It is not easy to discuss the effect of CCN concentrations on effective radius near cloud bases (at lowest penetration level) using Figure 8b since LWC is entirely too sensitive to small distances from cloud bases and it strongly influences r_e in clouds. Fortunately, two sets of r_e were observed in different clouds with similar LWC at respective levels of ~50 m (LWC: ~0.2 g m^{-3}) and ~500 m (LWC: ~0.3 g m^{-3}) above the cloud bases. Hence the relationship between r_e and CCN concentrations under the cloud bases was analyzed at similar LWC (Figure 13). The r_e showed a negative relation with CCN under the cloud bases at similar LWC in the cases of ~50 and ~500 m above the cloud bases. *Hudson and Li* [1995] observed low concentrations of CCN near clean clouds. Further, they demonstrated that the cleaner clouds had low concentrations of large droplets. *Albrecht* [1989] suggested that the decrease of cloud droplet sizes due to the increase of aerosol concentrations would suppress the formation of drizzle. Thus it is considered that effective radii near cloud bases decreased with increasing CCN concentrations under the cloud bases at similar LWC.

5. Conclusions

[26] Measurement of SO_2, aerosol particles, and CCN by aircraft observations strongly suggested that the atmosphere over the sea near the southwest islands of Japan was often influenced by polluted air transported from east Asia. It was found that the SO_2 concentrations were as low as 0.3 ppb in the relatively clean maritime atmosphere and as high as 1–10 ppb in the polluted atmosphere. The CN ($d > 0.01$ μm) concentration was as high as 3000–5000 cm^{-3} and AP ($3.11 > d > 0.12$ μm) concentration was as high as 800–1700 cm^{-3} in the boundary layer under the inflow of

pollutants from the east Asian countries such as China, Korea, and Japan. An excellent correlation between concentrations of SO_2 and aerosol particles in the polluted atmosphere suggested that most of the SO_2 might be converted to SO_4^{2-} during the long-range transport. Concentration of CCN activated at 0.3% supersaturation (SS) was as high as 800–2000 cm^{-3} in the polluted atmosphere. The ratio of CCN to CN concentrations was lower than 0.3 in the relatively clean maritime atmosphere, whereas it was ~0.5 in the continentally/anthropogenically influenced air in the boundary layer. Further, the slope of N_{CCN} to N_{AP} in this study is higher than those of other previous studies. The excellent correlation between CCN and aerosol particle concentrations strongly suggested that a heavy loading of air pollutants greatly contributed to CCN in the marine atmosphere perturbed by anthropogenic pollutants.

[27] Mean cloud droplet concentrations (N_C) near the cloud bases in the continentally influenced clouds were ~2 times those in the relatively clean maritime clouds. The relation of $N_C \sim N_{CCN}^{0.39}$ was obtained from the best fit curve of the log-log relation between N_C and N_{CCN}. The positive slope (0.39) suggested that the increase in CCN concentration under the cloud bases increases N_C in clouds. Moreover, it was inferred that high CCN concentration under the cloud bases decreases the effective radius at similar LWC.

[28] This study strongly suggests that the anthropogenic pollutants from east Asia severely increase CCN concentrations and modify the microphysical properties of clouds over the sea near the southwest islands of Japan. The observational results of this study contribute to increased useful information on the actual state of CCN distributions and on the effects of continental pollutants on CCN distributions and microphysical properties near the cloud bases over the ocean in east Asia. Further, the relationship between concentrations of aerosol particles and cloud droplets over the ocean examined in this study will give useful information for a better understanding of precipitation mechanism and/or climate change.

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