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Simultaneous Measurements of Hygroscopic Property and Cloud Condensation Nucleus Activity of Aerosol Particles of Marine Biogenic Origin

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Introduction

Some inorganic and organic compounds formed and released by marine biological activity are emitted into the atmosphere as gases and aerosol particles. Among these compounds, those emitted as gases can be converted to condensable species via photochemical reaction processes in the atmosphere, and might therefore engender the formation and growth of atmospheric aerosol particles. Although the formation of sulfate aerosols from marine-derived dimethyl sulfide (DMS) is understood well (e.g., Seinfeld and Pandis 1997), the possible contribution of secondary organic aerosol formation from marine-derived volatile organic compounds (Meskhindze and Nenes 2006, 2007) is not understood sufficiently. Furthermore, primary organic materials could be released to the atmosphere in association with sea salt particles as a result of the bursting of bubbles on the ocean surface (e.g., Blanchard 1964). (Although they were not part of this project (A01-K1) at the time of the proposal, the release is also an important process, as demonstrated by recent studies (e.g., Facchini *et al*. 2008).) These primary and secondary aerosol components of marine biogenic origin might affect the atmospheric aerosol concentrations and properties, although the actual significance remains unclear.

Aerosol particles' capability to act as the nuclei of cloud droplets, i.e., cloud condensation nuclei (CCN) activity, and the particle hygroscopicity, which strongly influences the CCN activity, are related to the CCN number concentrations. They might therefore affect the lifetime and the optical property of clouds and precipitation over the ocean. Consequently, elucidating hygroscopicity and CCN activity is important for assessing the climatic effects of aerosols. If marine biological activity affects the Earth's climate through its influence on the formation and growth of atmospheric aerosols, such activity can constitute a key process of the interaction between the atmosphere and the biosphere (Charlson *et al*. 1987). However, our understanding of this interaction via the formation and growth of aerosols, from elemental to overall processes, remains limited.

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The PI of the project A01-K1 has been using a hygroscopicity tandem differential mobility analyzer (HTDMA) for analyses of the hygroscopic properties of laboratory-generated and atmospheric aerosol particles (Mochida and Kawamura 2004; Mochida *et al*. 2006, 2008). Using this technique, changes of the particle diameter resulting from the uptake of water vapor can be measured quantitatively. A CCN counter (CCNC) measures the number concentration of particles becoming large droplets under supersaturated water vapor conditions (relative humidity (RH) > 100%), whereas the HTDMA measures the particle diameter growth under subsaturated water vapor conditions (RH < 100%). Despite the difference in the RH ranges, the hygroscopic growth data obtained using the HTDMA, which are closely related to the bulk hygroscopicity of the particles, are useful for assessing the CCN activity under supersaturated water vapor conditions (e.g., Brechtel and Kreidenweis 2000; Petters and Kreidenweis 2007). At the time of the proposal of this research project, the development of a part of a new HTDMA had been started. In the project, this newly built HTDMA at Nagoya University was used to conduct atmospheric observations of the hygroscopic growth of particles, with some customization depending on the field campaigns.

The objectives of this research project were to investigate the influence of the formation of growth of aerosol particles of a marine biological origin based on measurements of hygroscopicity and CCN activity of aerosol particles, which is closely related to the formation of cloud droplets, and to elucidate the link between marine biological activity and cloud processes. More concretely, the specific objectives were (1) to measure changes in the hygroscopicity and CCN activity associated with particle formation and growth, (2) to estimate the ratios of inorganics to organics in the particles, (3) to estimate the particles' surface tension, (4) to clarify the external mixing state of the particles, (5) to make a general assessment in view of marine biological activity, and (6) to propose important processes to be studied using a model. In this project, two atmospheric observations and analyses of the collected data were assigned as the activities of this project. Based on atmospheric aerosol observations conducted at Cape Hedo, Okinawa, and over the western North Pacific and on analyses of the data, results related to some of the objectives described above were acquired. Regarding the objectives described above, the operation of an HTDMA coupled with a CCN counter was proposed, as specified in the title of this project. Although measurements in these two observations were performed using the HTDMA coupled to a CCN counter, the main part of this report concerns the measurements of hygroscopic growth factors of aerosols and the subsequent analysis. This is partly because measurements of particle hygroscopicity and CCN activity were performed in collaboration with other research institutions and under other research funding. It should be noted that, although this summary was prepared by a single author, the studies of this project were conducted along with several collaborators and contributors, as described in Acknowledgements.

Atmospheric Measurements Using HTDMA

Instrumental setup and aerosol observations at Cape Hedo, Okinawa

Measurements of atmospheric aerosol particles using a HTDMA connected to a CCNC were conducted at the Cape Hedo Atmosphere and Aerosol Monitoring Station of the National Institute for Environmental Studies (NIES-CHAAMS) in Okinawa, Japan. This site is located in the northwestern part of Okinawa Island,

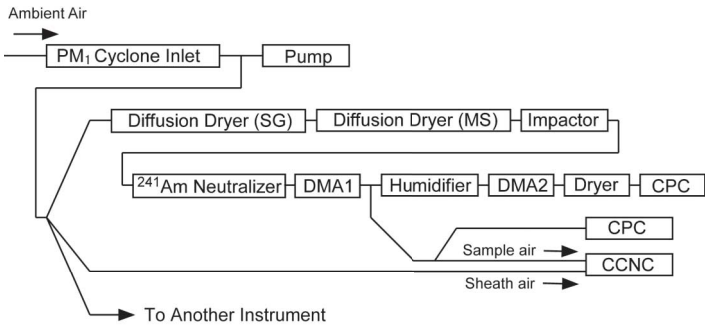

Fig. 1. Schematic diagram depicting the instrumental arrangement used for atmospheric measurements (HTDMA coupled to the CCNC). The instrument was used for the observation of this project until 1 April 2008, and that after 3 April in a related research project. Modified from Mochida *et al.* (2010) by permission of American Geophysical Union (AGU). Copyright 2010 AGU.

where the flow of polluted air from eastern Asia has been studied (Takami *et al.* 2007). Atmospheric measurements were performed in March and April, 2008. A schematic diagram of the measurement system, including the HTDMA coupled to the CCNC, is presented in Fig. 1. During field measurements, the atmospheric aerosols were aspirated at a PM1 cyclone inlet placed on the rooftop of the station building. Part of the sample aerosol was dehumidified by passage through two diffusion-type scrubbers, each filled with silica gel (SG) and a molecular sieve (MS). The sample aerosol was passed through an ^{241}Am aerosol neutralizer to establish a charge equilibrium. Then it was introduced to the first DMA (DMA1) column in the HTDMA. Electrical voltage was applied to DMA1 to classify aerosols and to select charged particles in a specific narrow-diameter range. A part of the sample aerosol was passed through Nafion tubing downstream of the DMA1. To supply water vapor to the sample aerosol, air for humidification was passed between the outside wall of the Nafion tubing and the inner wall of its surrounding housing. The humidity condition of the air used to humidify the sample aerosol was controlled, using mass flow controllers, by mixing the dry air and the humidified air through a Nafion humidification device with water outside the Nafion tubing. The sample aerosol, with RH controlled to about 85%, was then introduced to the second DMA (DMA2) column of the HTDMA. The humidity condition of the sheath air supplied to the DMA2 column was also controlled using a Nafion humidification device so that the RH inside the DMA2 column was about 85%. Under this condition, DMA2 was operated under a scanning mobility particle sizer (SMPS) mode. The voltage applied to DMA2 was scanned, and the particles transferred through DMA2 were counted using a condensation particle counter (CPC, Model 3022; TSI Inc.). Another Nafion tube was placed between DMA2 and the CPC so that the humidified sample aerosol was dehumidified again. This was to reduce possible unfavorable condensation of water inside the CPC. Note that the experimental setup is reported for a study under a related research project at the same location (Mochida *et al.* 2010).

The voltage applied to DMA1 was changed every 5 min to select the dry mobility diameters of atmospheric aerosol particles subjected to the measurement of

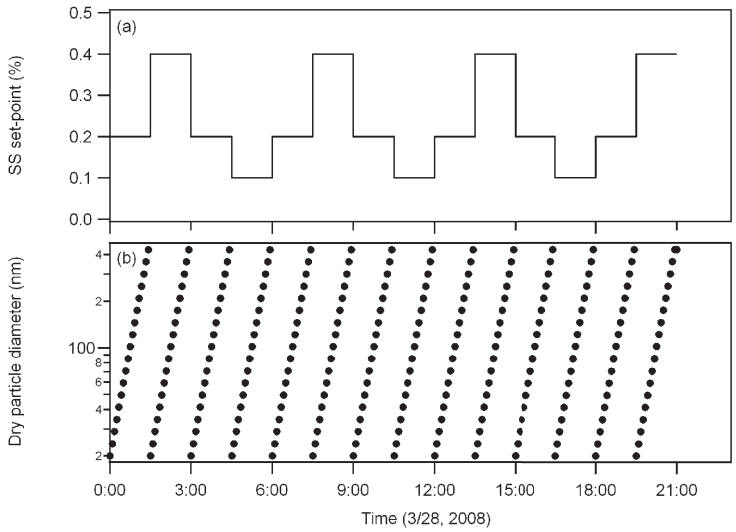

Fig. 2. Example of (a) the setting of the CCNC and (b) the dry mobility diameter setting of DMA1 in the HTDMA, for observations conducted at Cape Hedo, Okinawa, on 28 March 2008. The HTDMA data collected for diameters of 24.1–359 nm were used for the analyses. The CCNC were operated in collaboration with related research projects. The collected CCNC data have not been analyzed for the characterization of the CCN activity.

the hygroscopic growth (Fig. 2). Dry diameters used for analyses in this study were 24.1, 28.9, 34.6, 41.4, 49.6, 59.4, 71, 85.1, 102, 122, 146, 175, 209, 250, 300, and 359 nm. These diameters are uniformly spaced on a log scale. A set of data of the particle hygroscopic growth was collected from small to large diameters every 1.5 h (Fig. 2(a)). When this project was proposed, it had been planned to use short DMAs for the two DMAs in the HTDMA and to measure particles of <10 nm. However, longer DMAs were used for the measurements in this study. Consequently, analyses were conducted for larger (≥24.1 nm) particles.

During the measurements, the sample aerosol was split downstream of DMA1 in the HTDMA. The split flow was introduced to the CCNC (CCN-100; Droplet Measurement Technologies) and the CPC (Model 3775; TSI Inc.), as shown in Fig. 1.

Observations over the northwestern North Pacific

During the KH-08-2 cruise of R/V Hakuho Maru, which is owned by the Japan Agency for Marine-Earth Science and Technology (JAMSTEC), atmospheric aerosol measurements conducted using the HTDMA connected to the CCNC were performed. Measurements for this project were conducted during the leg 1 cruise in the northwestern North Pacific region in August 2008. The atmospheric aerosol was aspirated at the inlet above the bridge of R/V Hakuho Maru. The sample aerosol was transferred to the room of the research vessel through sampling tubing. Particles larger than 1 μm were removed using a PM1 cyclone connected in-line. Then, the sample aerosol was introduced to a series of instruments and sampling lines composing the HTDMA, which include two diffusion-type scrubbers connected in series (filled respectively with SG and MS). The

setup of the instruments including the HTDMA resembled that used for measurement at Cape Hedo, Okinawa (Fig. 1). However, the instrumental setup had some important differences. For example, a different model of CPC (Model 3775; TSI Inc.) was used for HTDMA during the R/V Hakuho Maru cruise. The experimental setup is reported for a study under a related project (Mochida *et al*. 2011).

Near the threshold diameters for CCN activation, hygroscopic growth was measured using smaller diameter intervals. These results are the same as those for aerosol measurements at Cape Hedo, Okinawa, obtained after 3 April 2008 (Mochida *et al*. 2010) and during the KH08-2 leg 2 cruise over the western North Pacific (Mochida *et al*. 2011). Consequently, in the case of the study for the northwestern North Pacific region, the hygroscopic growth was measured for the dry diameter range, which is the same as that for the study at Cape Hedo (24.1–359 nm), but for a greater number of diameters. For instance, the additional dry mobility diameters when CCN was measured at 0.2% supersaturation (SS) setting were 51.4, 53.3, 55.2, 57.3, 61.5, 63.8, 66.1, 68.5, 73.7, 76.4, 79.1, 82.0, 88.2, 91.4, 94.7, 98.2, 106, and 109 nm. They were selected so that dry particle diameters for the measurements are uniformly spaced in a log scale in the range of 49.6–109 nm. Different dry diameter sets were applied for the time periods of CCN measurements with SS settings of 0.1% and 0.4%. The additional diameters for the 0.1% SS setting were shifted to a larger diameter range (88.2–188 nm). In contrast, the additional diameters for the SS setting of 0.4% were shifted to a smaller diameter range (35.9–76.4 nm). The SS presented above are the set values, which have not been corrected based on the measurement of CCN efficiency spectra of pure ammonium sulfate particles (Mochida *et al*. 2010). These additional measurements necessitated a longer acquisition time for the series of dry mobility diameters (3 h per cycle) although they provided information related to the particle hygroscopicity for a wider variety of sizes.

Data Analysis

Based on data collected from the measurements at Cape Hedo, Okinawa, and over the northwestern North Pacific, the hygroscopic growth factors, defined as the ratio of the diameter of an aerosol particle under a humidified condition $d_p(\mathrm{RH})$ to that under a dry condition $d_{p,dry}$, were investigated for the sampled atmospheric aerosol particles:

$$\text{Hygroscopic growth factor} = d_p(\mathrm{RH})/d_{p,dry}. \quad (1)$$

The diameters used for the calculation ($d_p(\mathrm{RH})$ and $d_{p,dry}$) were electrical mobility diameters. As described above, the hygroscopic growth factors were obtained at *ca*. 85% RH. In the case of atmospheric particles, the hygroscopic growth factor can differ among particles. Therefore, if a sample atmospheric aerosol is a mixture of several particle groups with similar hygroscopicity, then bi-modal or multi-modal distributions are expected to be observed.

In this project, inversion to consider the widths of the transfer functions of DMAs (e.g., Stolzenburg 1988) in the HTDMA were not performed for the calculations of the distributions of hygroscopic growth factors. Furthermore, issues that demand further assessment, such as disturbance of the RH in the DMA2 column, remain. (At the time of the acquisition of the hygroscopic growth data, the RH of the sample and sheath air supplied to the top of the DMA2 column and that of the sheath air exiting from the bottom of the DMA2 were measured and recorded.) Multiply-charged particles are expected to exist in

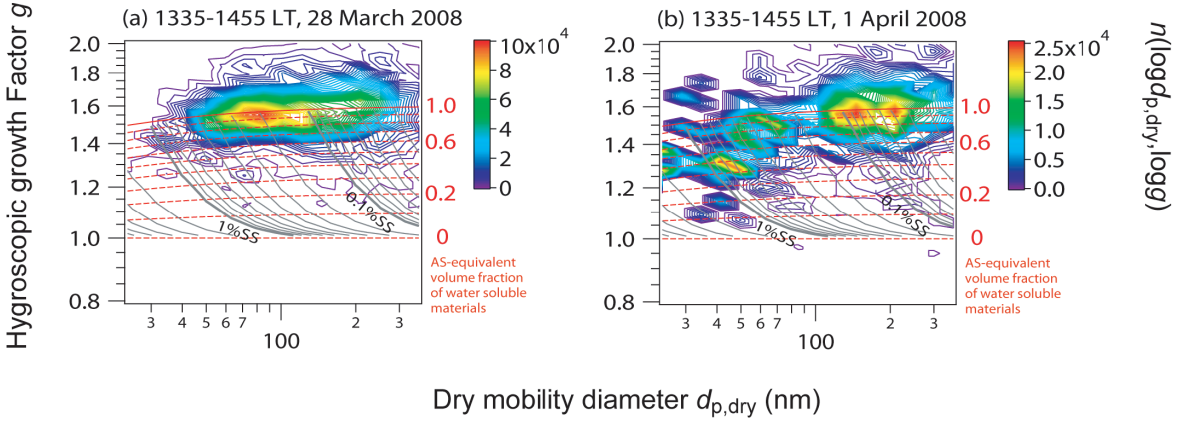

Fig. 3. Hygroscopic growth factor distributions for different dry mobility diameters: (a) distributions at 1335–1455 LT on 28 March 2008, and (b) distributions at 1335–1455 LT on 1 April 2008. Red dashed lines represent the ammonium sulfate (AS) equivalent volume fraction, as calculated based on a core-shell model assuming the particle to be an insoluble core and a shell of ammonium sulfate aqueous solution in the humidified condition. Gray solid lines represent the SS required for the CCN activation. The fraction of aerosol particles above the SS isopleth line is predicted to activate under the SS condition. These SS isopleths were also calculated assuming core-shell particles with ammonium sulfate as the soluble shell component. See Mochida *et al.* (2010) for details related to calculations of the AS-equivalent volume fraction and the SS isopleths. The AS-equivalent volume fraction of water-soluble materials, the SS isopleths, and the label of the color contours have been corrected for this report after completion of the project. Furthermore, correction factors to adjust the sizing between DMA1 and DMA2 were updated using the values from the work reported earlier (Mochida *et al.* 2010). The concept of the SS isopleths is explained in Gasparini *et al.* (2006). This figure is presented herein to outline the results of this project; it was prepared based on a preliminary analysis and might not be identical to finalized results in the future.

the aerosol downstream of the aerosol neutralizer according to the Boltzmann distribution (Wiedensohler 1988; TSI Inc. 2006). Correction for the presence of multiply-charged particles (Duplissy *et al.* 2009) was not performed in this study.

For the aerosols over Cape Hedo, Okinawa, distributions of hygroscopic growth factors for dry mobility diameters of 24.1–359 nm were drawn based on the collected data for 16 diameters. In the case of the aerosols over the northwestern North Pacific region, the size-resolved hygroscopic growth factor distributions were obtained for the same size range but based on data for a larger number of dry diameters in addition to the 16 diameters.

Results of Atmospheric Observations

Hygroscopic growth of aerosol particles over Okinawa

Results obtained from the hygroscopic growth measurements of aerosols at Cape Hedo, Okinawa, and over the northwestern North Pacific are described briefly. Based on the analysis of the observational data collected at Cape Hedo, Okinawa, distributions of hygroscopic growth factors of atmospheric aerosol particles from the Aitken mode (<100 nm) to the accumulation mode (>100 nm) on 28 and 31 March, and on 1 April 2008 were obtained (Fig. 3). More data were collected using the HTDMA before 28 March, but they have not been analyzed for the characterization of the aerosols at Cape Hedo. Hygroscopic growth factors observed at the set point of 85% RH were characterized mainly by unimodal distributions. The hygroscopic growth factors of most of the observed particles were similar to those of pure ammonium sulfate particles, which indicates that the observed atmospheric particles have high hygroscopicity. For particles in which soluble and insoluble components are mixed, the higher fraction of the water-soluble fraction engenders higher hygroscopicity of the particles if the hygroscopicity of the water-soluble fraction is assumed to be constant. (The core-shell model treats this issue in a simplified manner.) The hygroscopic growth factors of aerosol particles observed at Cape Hedo, Okinawa, suggest that the volume fractions of the water-soluble component in the particles were large if the soluble fraction is composed mainly of ammonium sulfate or materials whose bulk hygroscopicity is equivalent to that of ammonium sulfate. Note that data collected using the CCNC in collaboration with related projects were not analyzed for the characterization of the CCN activity.

Hygroscopic growth of aerosol particles over the northwestern North Pacific

Based on atmospheric measurements obtained over the northwestern North Pacific on board the R/V Hakuho Maru, measurements of marine aerosol particles using the HTDMA connected to the CCNC were conducted for the atmosphere over the remote ocean where the concentrations of aerosol particles are, in general, expected to be low. Distributions of hygroscopic growth factors in the dry diameter range from the Aitken to the accumulation modes were obtained, although analyses were preliminary (data not shown). Similarly to the HTDMA data collected at Cape Hedo, Okinawa, data collected during this cruise are expected to be useful for assessing the particle composition qualitatively. However, the obtained hygroscopic growth factor distributions include those probably influenced by the ship's exhaust, which might be attributable in part to the long measurement cycle necessary to obtain the size-resolved hygroscopic growth factor distributions (3 h). Therefore, care should be taken to distinguish the distributions of hygroscopic growth factors of marine aerosol particles from those emitted from the research vessel. It is worth noting that, in the observa-

tion of the KH08-2 leg 1 cruise of R/V Hakuho Maru, data were also collected using a CCNC and a CPC connected to the HTDMA as a collaborative study. Calculations of the size-dependence of the number fraction of the CCN (so-called CCN efficiency spectra), which represent the CCN activity of particles, have been attempted for SS settings of 0.1%, 0.2%, and 0.4% (data not shown). Therefore, the hygroscopic growth factor distributions obtained in this project were for the period including the period of the measurement of the CCN activity.

Study Outcomes

From the two atmospheric observations at Cape Hedo and over the northwestern North Pacific, data used to infer the chemical composition of aerosol particles qualitatively were obtained, with an approximation that the major components of particles are water-soluble and water-insoluble components and with an assumption of the hygroscopicity of the water-soluble component. Because the bulk hygroscopicity of the particulate matter, which is represented by the hygroscopic growth factors, is closely related to the CCN activity of the particles, the obtained distributions of hygroscopic growth factors are expected to be applicable to additional analysis of the CCN in the studied aerosols (Gasparini *et al*. 2006; Mochida *et al*. 2010). For example, using the core-shell model, the CCN activity of aerosol particles at various supersaturation conditions can be predicted from hygroscopicity growth factor distributions as presented by the SS isopleths in Fig. 3. As shown in the figure, the CCN activity of a particle is larger if the diameter or the hygroscopic growth factor is larger.

As explained above, inversion to incorporate consideration of the transfer functions of two DMAs in the HTDMA was not performed for calculation of the distributions of hygroscopic growth factors from the observational data of this project. Therefore, the obtained hygroscopic growth factors might have apparent distributions that are presumably broader than the actual distributions, as in the case of the examples presented in the paper by Mochida *et al*. (2008). Inversion of the HTDMA data using MATLAB (The Mathworks Inc.) is currently possible based on a study relevant to this research project (Mochida *et al*. 2010, including auxiliary materials).

Analyses of data collected from the two atmospheric observations are preliminary. Some subjects related to the research objectives, such as the comparison of the particle CCN activity with the marine biological activity (objective 5 in the Introduction section), were not achieved in this project. However, the setup of the instrument and the assessment of the measurement method contributed to the subsequent atmospheric observations performed primarily under a related research project in which the PI of A01-K1 was involved (Mochida *et al*. 2010, 2011). The objectives of this study include one newly accomplished in a related project (analysis of the surface tension of aerosol particles (objective 3 in the Introduction) based on a comparison of the measured CCN activation diameters with those predicted from hygroscopic growth factors measured using HTDMA). Furthermore, the objectives of this project (A01-K1) include those assessed similarly in the related project. For example, the AS equivalent volume fraction was assessed based on the hygroscopic growth factor distributions at Cape Hedo after 3 April 2008 (relevant to objective 2 in the Introduction). Details of these analyses were presented in two earlier reports (Mochida *et al*. 2010, 2011).

Summary and Future Outlook

Through this research project, informa-

tion related to the hygroscopic property of atmospheric aerosol particles at Cape Hedo and over the northwestern North Pacific was obtained. Although the results are based on analyses performed preliminarily, and although some of the objectives presented at the time of the proposal have not been accomplished, information related to the mixing state of atmospheric aerosols has been obtained based on the acquisition of hygroscopic growth factors for various particle diameters from the Aitken to the accumulation modes. This type of information, which is unobtainable using the general methods of bulk/size-resolved aerosol sampling and subsequent chemical analyses such as ion chromatography, is insufficient even at present. Furthermore, atmospheric measurements performed primarily for this project supported atmospheric measurements conducted at Cape Hedo and onboard R/V Hakuho Maru under a related research project.

The hygroscopic property of aerosol particles over the ocean is still an important research subject, as demonstrated by a recent study on hygroscopicity and CCN activity (Good *et al*. 2010). Although more than two years have passed since the end of the project term, key issues remain to be resolved. For instance, studies that particularly address smaller particles are needed. New particle formation was not evident, at least from the data analyzed for the marine atmospheric aerosols under this project. However, if measurements of the hygroscopic growth of small particles are performed not at the marine boundary layer, where new particle formation is suppressed because of the presence of sea salts, but in environments with lower concentrations of particles (e.g., in the free troposphere), it is expected that important knowledge related to the properties and the composition of small particles, whose chemical compositions are difficult to analyze directly, will be obtained. Furthermore, the detection of externally-mixed aerosol particles in the marine atmosphere remains a subject for further research. Based on measurements of hygroscopic growth factors over the North Pacific during spring, Massling *et al*. (2007) estimated that sea salt particles were mixed externally with other particles. Additional investigations conducted during seasons with large emissions of sea salts, or in areas with large sea-salt emissions, are important. The data are expected to be valuable for the development of models dealing with sea-salt particles and to assess, for example, the role of sea-salt aerosols in cloud droplet formation. Moreover, the data are important to assess the presence of organics of a marine biological origin emitted with sea salts (e.g., O'Dowd *et al*. 2004; Keene *et al*. 2007; Facchini *et al*. 2008) and to assess the possible influence of the condensation of secondary components. For these studies, the assessment of the effect of the surface tension of organics (Fruentes *et al*. 2011) is a key subject to be investigated. Further, comparison of the particle CCN activity with the indices of marine biological activity, which has not been accomplished in this project, remains as an important research subject.

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in a collaborative study. The PI also thanks Mr. Akio Shigai of the Institute for Advanced Research, Nagoya University, the captain and crew of the R/V Hakuho Maru, and other researchers and staff who supported this project. Author affiliations are shown as at the time the work was completed.

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