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The Influence of Aerosol Source Region on Size-resolved Hygroscopicity During the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) Campaign

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Abstract

Aerosol hygroscopic properties were measured by a tandem differential mobility analyzer (TDMA) system during the Aerosol Characterization Experiment (ACE)-Asia campaign from 31 March to 1 May 2001. Two high flow differential mobility analyzers (DMAs) were used to maximize the count rate on board the Center for Interdisciplinary Remotely Piloted Aircraft (CIRPAS) Twin Otter aircraft. Hygroscopic growth factor distributions of particles having initial dry nanoparticle diameters of 0.040, 0.059, 0.086, 0.126, 0.186, 0.273, 0.400, and 0.586 μm were measured during 19 research flights. Data collected during 12 of those flights were used to investigate aerosol mixing state and the influence of aerosol source region on size-resolved hygroscopicity. The uniformity in size-resolved hygroscopicity was quantified to facilitate comparison between measurements made in different air masses. Hygroscopic growth factors are strongly dependent on source region and sizes. Mean hygroscopic growth factors were observed to be greatest when the air mass origin was from the south. The mean growth factors for continental sources decreased with initial size from 1.47 to 1.27 for 0.040 μm and 0.586 μm, but increased with initial size from 1.44 to 1.8 for 0.040 μm and 0.400 μm dry diameters for marine sources.

Key words : Nano aerosol, Hygroscopic growth factor, DMA/TDMA, Aerosol sources

1. INTRODUCTION

Aerosol particles play an important role in the atmosphere. The amount of water uptake by an aerosol particle determines its role in cloud formation processes as well as influencing its optical properties, due to changes in both particle size and phase (Intergovernmental Panels on Climate Change (IPCC, 2001)). The Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) campaign conducted in the spring of 2001 over eastern Asia was designed to assess the influence of atmospheric aerosol on Earth's climate system (Huebert *et al.*, 2003). In order to determine the impacts of aerosols on climate change, it is necessary to ensure that observations reflect the actual spatial and temporal heterogeneity. ACE-Asia incorporates aircraft-, ship-, ground-, and satellite-based measurements to investigate the physical, chemical, and optical properties of the aerosol. Of greatest interest were dust

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aerosols generated in the Gobi Desert and pollution aerosols originating from the major industrial regions in China and Korea. The Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter aircraft was employed to measure vertical and spatial distributions of aerosol concentrations and properties with a number of instruments. Similar aircraft measurements during the ACE-2 study, a predecessor campaign to ACE-Asia, were described by Collins *et al.* (2000). Gao *et al.* (2003) described the impact of aerosol hygroscopicity on light scattering based on data collected by three nephelometers operated on board the Twin Otter during ACE-Asia. The transport patterns of aerosols are closely related with the meteorological flow (Merrill and Kim, 2004). It is important to identify source region of aerosol for understanding the aerosol climatology of a region.

A TDMA was operated on board Twin Otter aircraft to measure size-resolved hygroscopic growth of the aerosol. The primary aim for operation of a TDMA during the study is to investigate both the size-resolved composition of particles and the association of different classes of compounds depending on the transport pathways. Hygroscopicity properties are characterized in terms of transport sources. In addition to the goal of characterizing aerosol mixing state, the TDMA measurements were useful for identification of non-hygroscopic submicron dust particles.

2. MEASUREMENT AREAS

The Twin Otter was employed during the project to characterize dust plumes and pollution typical of the study region. The Twin Otter completed 19 flights from the Iwakuni Marine Corps Air Station (IMCAS), Japan during the period from March 31 to May 1, 2001. Fig. 1 shows the flight area of the Twin Otter during ACE-Asia over eastern Asia and the sections used in mass trajectory analysis. During a typical flight, the Twin Otter often passed through different aerosol source regions. The area shown in Fig. 1 is divided in two, with polluted air masses

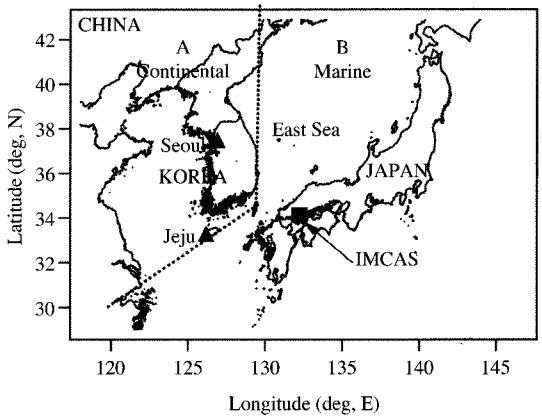

Fig. 1. The flight area of the Twin Otter during ACE-Asia over eastern Asia and the sections used in mass trajectory analysis.

mixed with dust from the Mongolia dust region to the left and marine air masses from the central and northern Pacific to the right. 12 flights during which the collected TDMA data were of the highest quality are analyzed here. These flights were on April 8, 9, 12, 13, 17, 19, 20, 23, 25, 26, 27 and 28 over the East Sea and around Jeju island. Flight routes are presented in detail by Bahreini *et al.* (2003).

3. IN SITU MEASUREMENTS

3.1 Aerosol hygroscopic growth factor measurement

Hygroscopic properties of aerosols have been examined using humidified TDMA systems in several areas (Gasparini *et al.*, 2004; Swietlicki *et al.*, 2000, 1999; i.e. McMurry *et al.*, 1996). A humidified TDMA consists of two DMAs in series, between which the aerosol is exposed to an elevated relative humidity (RH). The voltage applied to the first DMA (DMA1) is fixed to select a narrow size range of a polydisperse particle. Following exposure to high RH, the initially selected monodisperse aerosol enters the second DMA (DMA2) in which the applied voltage is continuously varied to characterize the changes in the size of the humidified

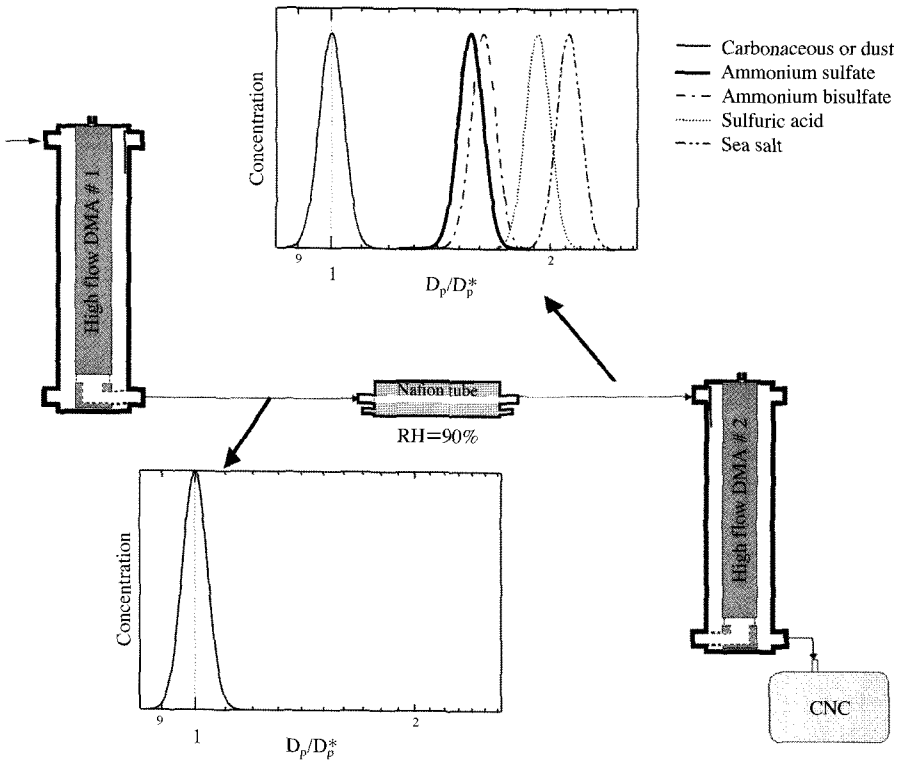

Fig. 2. Schematic of the TDMA theory.

particles. By interacting with water vapor, particle size changes based on its hygroscopic properties. By combining the known aerosol size separated by DMA1 with the size distribution selected by DMA2, the hygroscopic growth factor distribution can be determined. Hygroscopic growth factor (D_p/D_p^*) is the ratio of the diameter of a particle following exposure to 90% RH, D_p, to its dry diameter, D_p^*. Fig. 2 shows a schematic of the TDMA theory. The composition of an ambient aerosol can be inferred through a combination of a measured hygroscopic growth factor distribution and the expected hygroscopicity of pure aerosol types such as ammonium sulfate and sea salt. Fig. 3 shows a schematic of the TDMA instrument system operated during the ACE-Asia campaign. The TDMA system consists primarily of two DMAs, a Condensation Nuclei Counter (CNC), and a humidity conditioner. There are also a computer, pumps, and many control valves etc. Hygroscopic growth factor distributions with dry diameters of 0.040, 0.059, 0.086, 0.126, 0.186, 0.273, 0.400, and 0.586 μm are measured sequentially over a period of 45 minutes. We used Aerosol Dynamics, Inc. high flow DMAs to maximize the flow rate and therefore the count rate.

3.2 Example measurement

One set of flight data is shown in Fig. 4 to illustrate the distributions used in the subsequent analyses of data from the 12 research flights considered in this study. The top graph in Fig. 4 shows the aircraft altitude and TDMA RH measured on 23 April 2001. The time hereafter is local standard time. Lower graphs show hygroscopic growth factor distributions for dry particle diameters of 0.040, 0.059, 0.086, 0.126, 0.186, 0.273, 0.400, and 0.586 μm at eight different measurement legs. The aircraft flew ascents and descents and multiple altitude level legs from low altitude to 2 km for most of this flight. The presence of more hygroscopic 0.040- and 0.059-

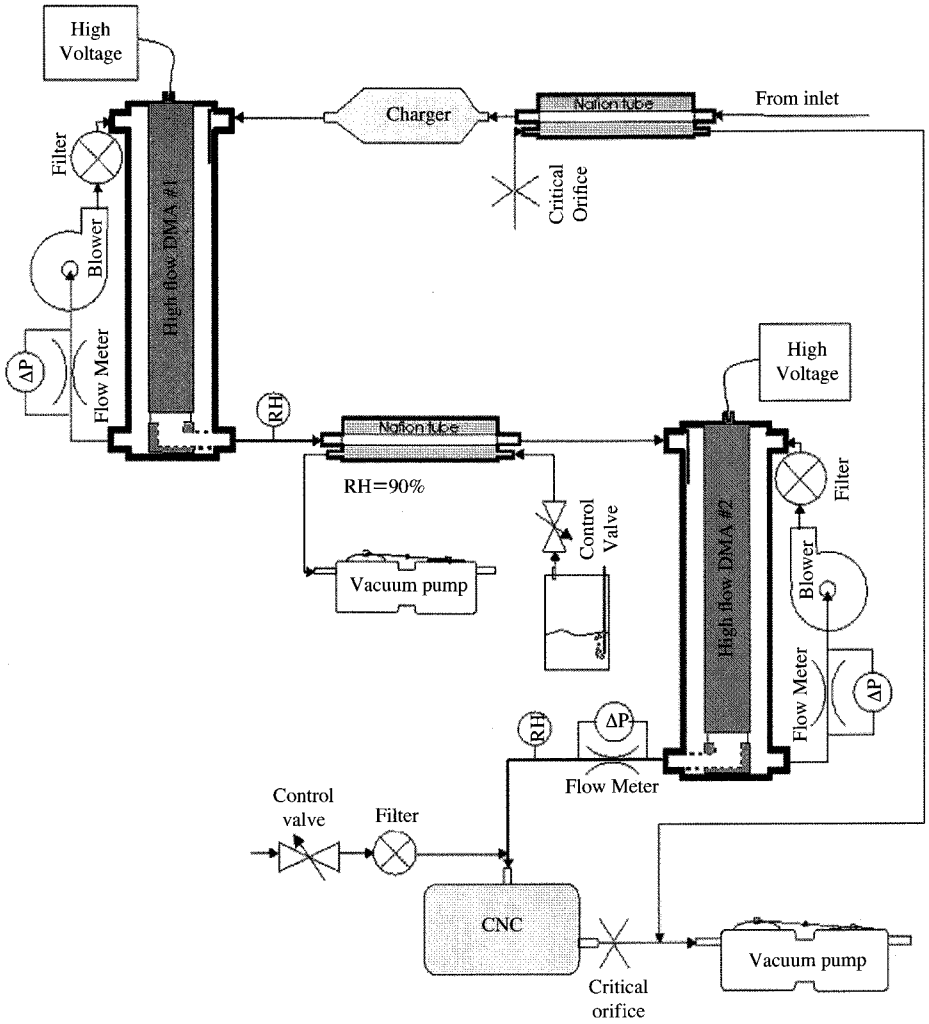

Fig. 3. Schematic of the TDMA system operated in ACE-Asia campaign.

μm particles during sampling intervals suggests recent nucleation of sulfuric acid particles. Most hygroscopic growth factor distributions measured on this date are monomodal and bimodal, and it suggests that the aerosol was internally or externally mixed by the relatively long transit time from source regions in China. Although hygroscopic growth factors varied as a function of time and size, most distributions were centered at approximately 1.4 to 1.7, and it suggests the aerosol was most likely composed of a mixture of sulfate with some less soluble species such as organics.

3.3 Parameterizations of growth factors

The lognormal distribution often provides a good fit of aerosol size and hygroscopicity distributions, and is regularly used in atmospheric applications (Seinfeld and Pandis, 1998). Computer software was developed to fit each hygroscopic growth factor distribution with between one and four lognormals. Each lognormal is prescribed by a geometric mean diameter (or equivalently, hygroscopic growth factor), a mode concentration, and the geometric standard deviation of the mode through the following expression,

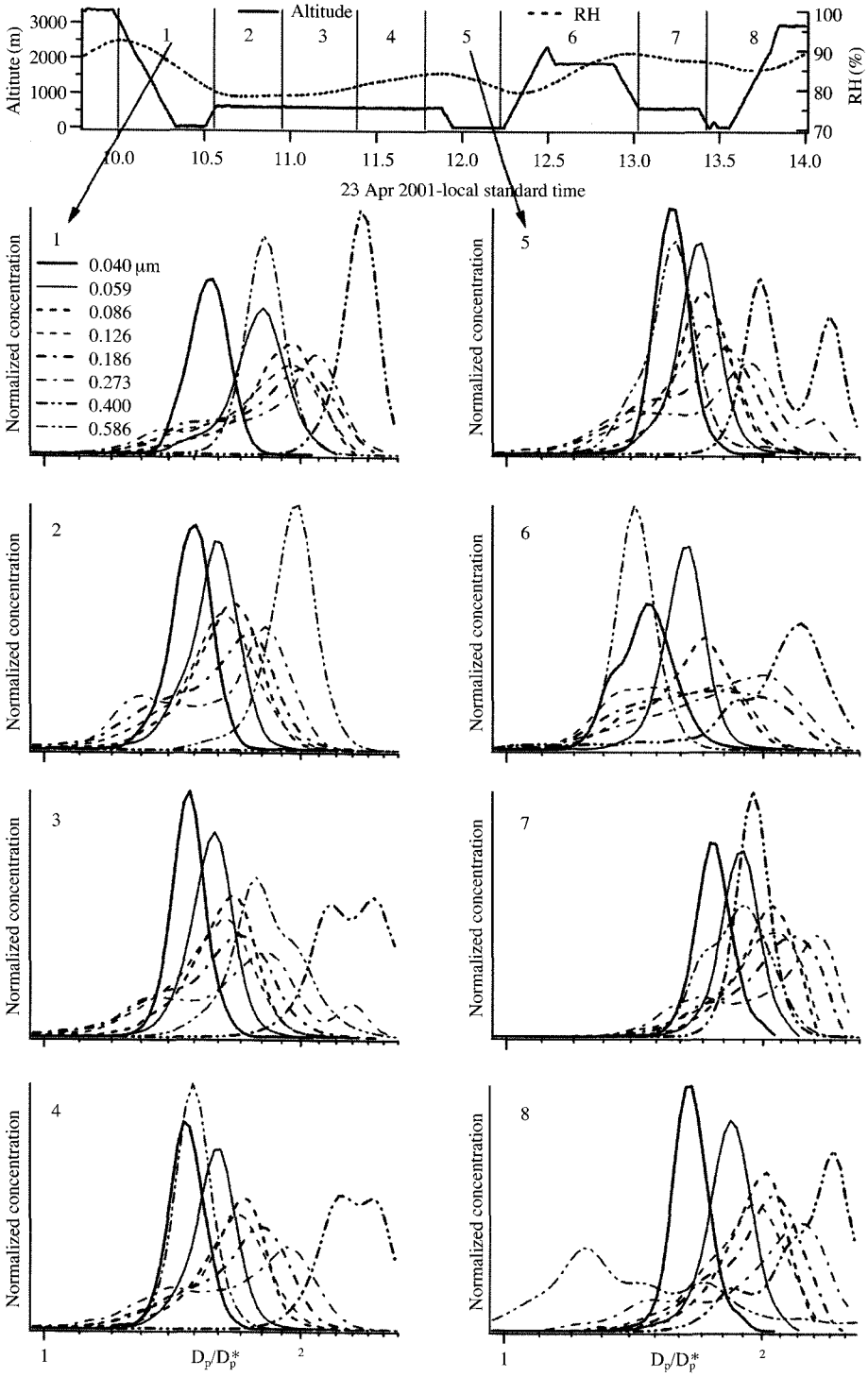

Fig. 4. Aircraft altitude and TDMA RH (upper) and hygroscopic growth factors distributions measured by the TDMA system (bottom) on 23 April 2001.

$$\frac{dN_i}{d\log D_p} = \frac{N_i}{\sqrt{2\pi}\cdot\log(\sigma_{g,i})} \exp\left[-\frac{(\log D_p - \log D_{pg,i})^2}{2\cdot(\log\sigma_{g,i})^2}\right] \quad (1)$$

where N_i is the total number concentration, $D_{pg,i}$ is the geometric mean diameter, and $\sigma_{g,i}$ is the geometric standard deviation for mode i.

4. RESULTS AND DISCUSSIONS

4.1 Average hygroscopic properties

Fig. 5 shows the statistics of measured hygroscopic growth factor distributions during 12 of the 19 research flights. The pie charts in the second column show the frequency of one, two, or some other number of modes in a hygroscopic growth factor distribution segregated by dry diameter. These classifications were based on the number of lognormal curve fits to growth factor distributions. The number fraction, geometric mean, and standard deviation of the lognormal distributions were used to quantitatively describe the hygroscopic growth factor distributions. The hygroscopic growth factor distributions of both 0.040- and 0.059-μm dry diameter were mainly monomodal, whereas multiple modes were frequently observed in the growth factor distributions for larger particles.

The plots in the right column of Fig. 5 show histograms of the central hygroscopic growth factor of the every mode for each particle size analyzed. The dashed vertical line in each histogram represents the theoretical hygroscopic growth factor of ammonium bisulfate at 90% RH, calculated based on the work of Tang and Munkelwitz (1994). Compared to measurements made in the Houston, Texas metropolitan area by Gasparini *et al.* (2004), there are relatively few non-hygroscopic modes for particles smaller than 0.273 μm. Hygroscopic growth factor distributions for particles with dry diameters at least 0.273 μm did occasionally possess minimally hygroscopic and non-hygroscopic modes, which were probably indicative of larger non-hygroscopic dust particles during some of the flights. Swietlicki *et al.* (1999) measured hygroscopic growth factors in northern England from 35 to 265 nm particles, and measured average growth factors of 1.11～1.15 for less hygroscopic particles, 1.38～1.69 for more hygroscopic particles and 2.08～2.21 for sea salt particles. Observed growth factors for 50 nm and 150 nm at Pasadena, CA by Cocker *et al.* (2001) ranged between 1.0 and 1.6. Growth factors during this study were higher than those urban aerosols. The internal mixing of dust particles, sulfates, nitrates, OC, BC may contribute to high hygroscopic growth factors.

4.2 Variability of hygroscopic properties with source regions

Distinct dust layers were frequently observed during the ACE-Asia campaign. The composition, source region, and concentration of the dust aerosols within each layer often differ from those at other heights. Non-hygroscopic dust particles are often found in layers in the higher altitude, while hygroscopic sea salt particles are typically present only close to the surface. The source region is critical for aerosol composition and consequently hygroscopic growth.

72-hour backward trajectories of parcels terminating at each measurement level were computed to investigate hygroscopic properties for the different air masses at twelve-hour intervals for entire periods using NOAA HYSPLIT model (Draxler and Rolph, 2003). These trajectories are classified mainly from the marine and continental. Marine sources are from 9, 19, 20, 23, 25, and 26 April, and continental sources are from 8, 12, 13, 17, 27, and 28 April. Examples of two different backward trajectories with different air mass source region are shown in Fig. 6. Anthropogenic emissions from major desert regions in Northern areas may alter the properties of the aerosol particles by mixing with soluble components. This type of perturbation would result in an increased hygroscopicity of the aerosol. But aerosols from marine sources show more hygroscopic growth factor than that of continental by the source region and soluble aerosol. Comparison of aerosol hygroscopic growth factor with those from previous measurements is not shown because of the lack of data

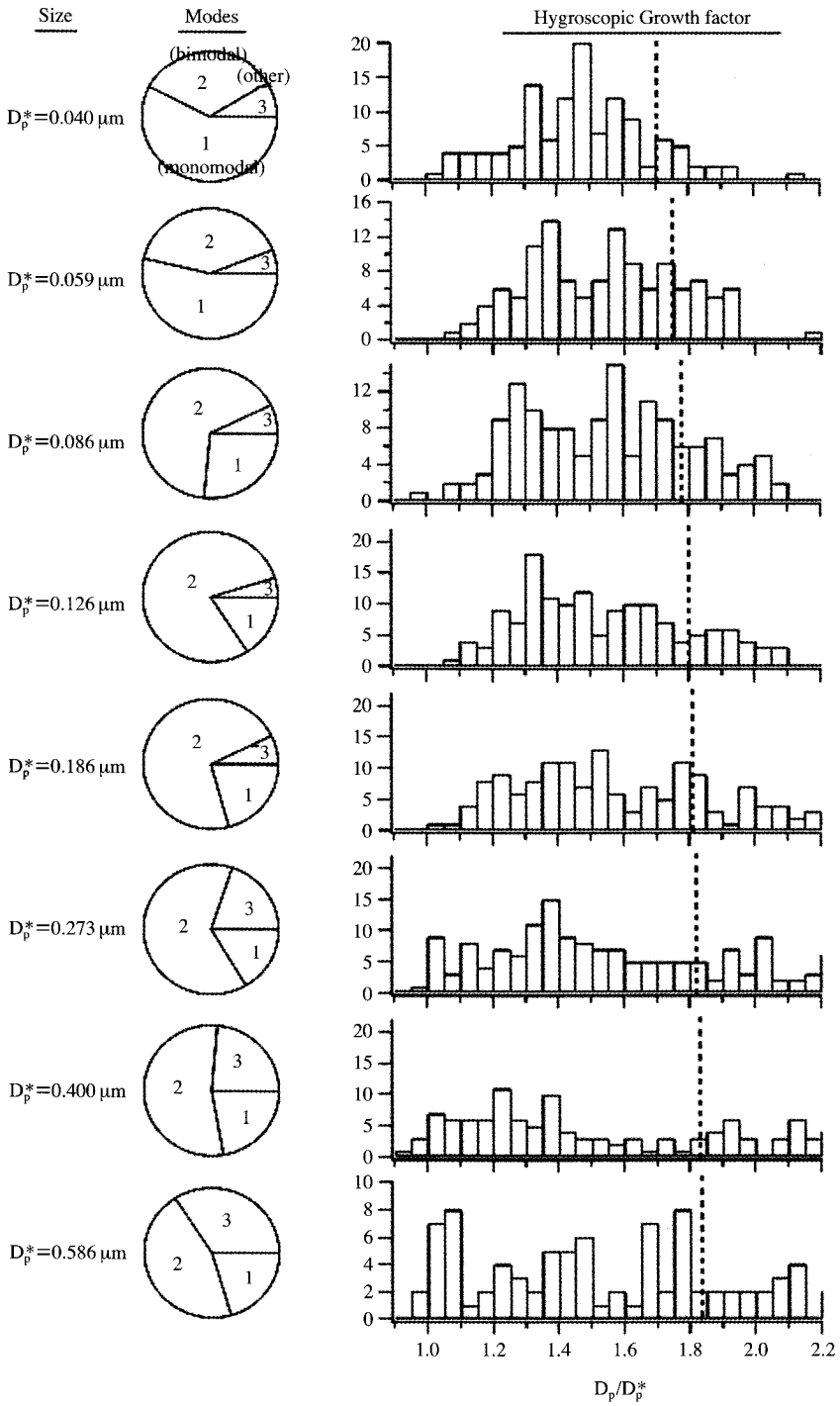

Fig. 5. Frequency of modes and histograms of the central growth factor of the every mode measured throughout the study. Dry diameter is listed in the first column, the fraction of distributions that were either monomodal, bimodal, or trimodal is shown in the pie charts in the second column and central hygroscopic growth factor of the every mode is shown in the right column.

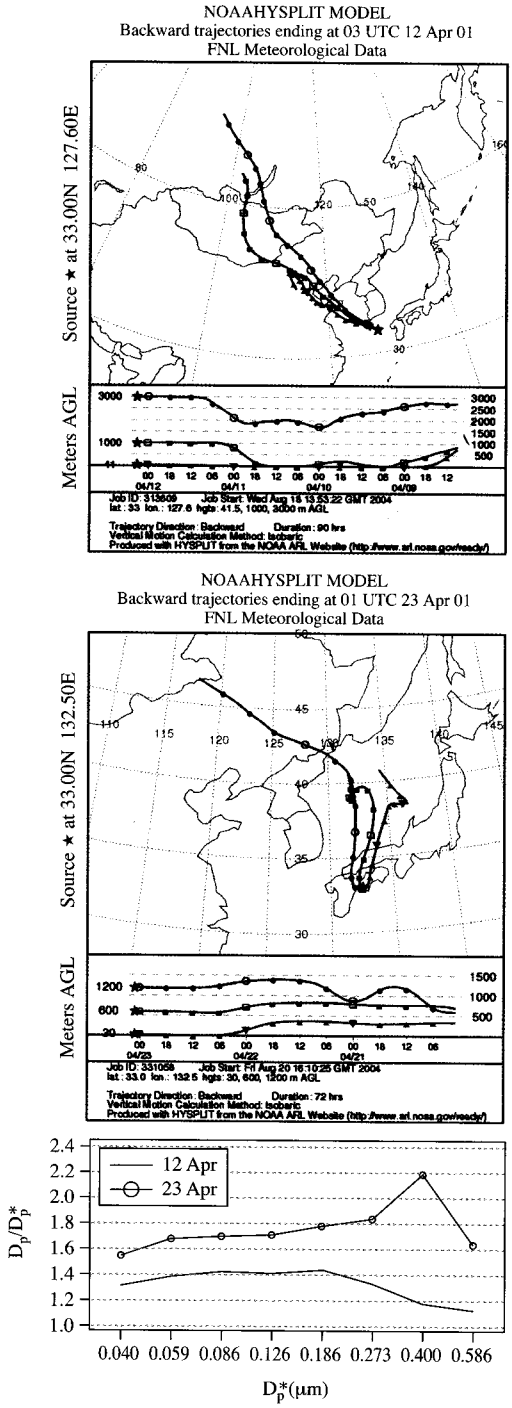

Fig. 6. The examples of two different backward trajectories calculated every six hours during the measurements (upper) and mean hygroscopic growth factors (bottom).

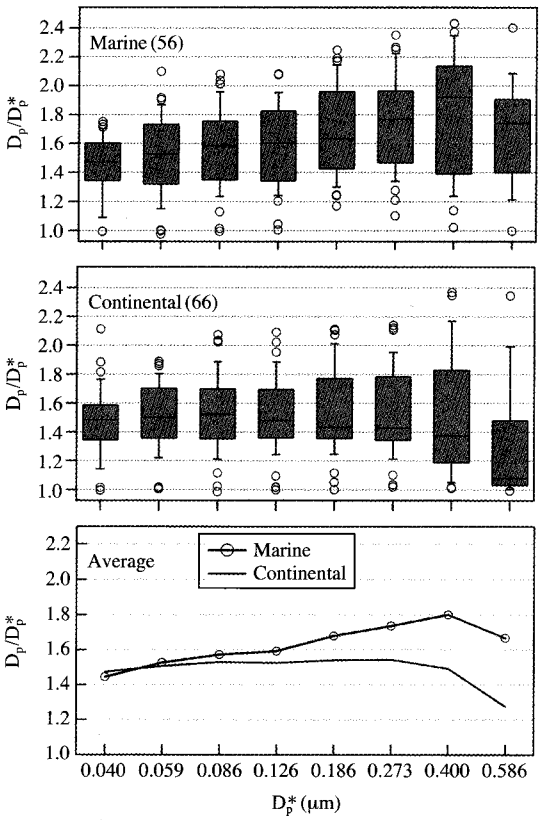

Fig. 7. Box plots and mean growth factors as a function of dry diameter separated by backward trajectory classification. The numbers in parenthesis denote the number of TDMA loops performed under each classification.

according to air mass trajectories.

Fig. 7 contains box plots showing the central growth factors segregated by airflow sources. The ends of each box represent the 25th and 75th percentiles and caps on the whiskers denote the 10th and 90th percentiles. The line across each box is the median. Consistent with measurements of the humidity dependence of light scattering by Gao *et al.* (2003), the aerosol from the marine sources was typically more hygroscopic than that from the continental sources. They explained this dependence by noting that aerosol samples originating from the north had similar fractions of anthropogenic, biomass, and dust components, but aerosols from the south had a larger fraction of sea salt species and

internally- or externally-mixed with by sulfate. This is supported by the observation here that the 0.586-μm particles were less hygroscopic than those of the 0.273-μm particles only in the uppermost level considered. Seinfeld *et al.* (2004) reported the similar observations of the source dependence of sulfate and coarse aerosol mass in Asia. The differences in hygroscopic properties in two category is highest when the size is increasing peak at 0.586 μm suggesting main component of these size are sea salt and dust.

5. SUMMARY AND CONCLUSION

Measurements of size-resolved aerosol hygroscopicity were made using a tandem differential mobility analyzer system on board the CIRPAS Twin Otter aircraft during the ACE-Asia campaign from March 31 to May 1, 2001. Hygroscopic properties of particles with dry diameters of 0.040, 0.059, 0.086, 0.126, 0.186, 0.273, 0.400, and 0.586 μm were measured over time intervals of approximately 45 minutes. Mean hygroscopic growth factors at 90% RH typically exceeded 1.4, suggesting the soluble fraction of the aerosol was relatively high except for 0.586 μm diameter for continental sources. The absence of multiple distinct modes in most distributions suggests the aerosol was generally internally-mixed. Despite the lack of multiple distinct modes, asymmetry in the measured growth factor distributions of larger particles measured indicates aerosol populations with similar, but not identical, composition were often present. The breadth of modes within hygroscopic growth factor distributions, and that of the entire distribution were used to quantify the compositional homogeneity of the aerosol. Overall, hygroscopicity was observed to be higher flows from south. One cause of the observed sources dependence is the frequent presence of hygroscopic particles that had been coated with soluble compounds such as sulfate.

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