J. KOSAE Vol. 21, No. E 1 (2005) pp. 23~35
Journal of Korean Society for Atmospheric Environment

Size Distribution Characteristics of Particulate Mass and Ion Components at Gosan, Korea from 2002 to 2003

J.S. Han, K.J. Moon*, S.J. Lee, J.E. Kim[1)] and Y.J. Kim[1)]

Department of Atmospheric Chemistry, National Institute of Environmental Research (NIER), Environmental Research Complex, Kungseodong, Seogu Inchun 404-170, Korea

[1)]ADvanced Environmental Monitoring Research Center (ADEMRC), Gwangju Institute of Science and Technology (GIST), 1 Oryong-dong, Buk-gu, Gwangju 500-712, Korea

(Received 15 January 2005, accepted 15 March 2005)

Abstract

Size distribution of particulate water-soluble ion components was measured at Gosan, Korea using a micro-orifice uniform deposit impactor (MOUDI). Sulfate, ammonium, and nitrate showed peaks in three size ranges; Sulfate and ammonium were of dominant species measured in the fine mode ($D_p < 1.8$ μm). One peak was observed in the condensation mode (0.218~0.532 μm), and the other peak was obtained in the droplet mode (0.532~1.8 μm). Considering the fact that the equivalent ratios of ammonium to sulfate ranged from 0.5 to 1.0 in these size ranges, it is inferred that they formed sufficiently neutralized compounds such as $(NH_4)_2SO_4$ and $(NH_4)_3H(SO_4)_2$ during the long-range transport of anthropogenic pollutants. On the other hand, nitrate was distributed mainly in the coarse mode (3.1~6.2 μm) combined with soil and sea salt.

Two sets of MOUDI samples were collected in each season. One sample was collected when the concentrations of criteria air pollutants were relatively high, but the other represented relatively clean air quality. The concentrations of sulfate and ammonium particles in droplet mode were the highest in winter and the lowest in summer. When the air quality was bad, the increase of nitrate was observed in the condensation mode (0.218~0.282 μm). It thus suggests that the nitrate particles were produced through gas phase reaction of nitric acid with ammonia. Chloride depletion was remarkably high in summer due to the high temperature and relative humidity.

Key words : MOUDI, Size distributions, Ion components, Neutralization, Chloride depletion, Gosan

1. Introduction

Particulate matter is a mixture of components that are formed by a wide variety of mechanisms associated with both natural and anthropogenic origins (Alves *et al.*, 2000). In general, particles up to 0.1 μm in aerodynamic diameter mainly originate from either atmospheric gas-to-particle conversion or combustion processes. These particles coagulate rapidly to form larger particles. On the other hand, particles in the size range above 1 μm originate from mechanical suspension processes in the lower troposphere (Seinfeld and Pandis, 1998; Tuch *et al.*,

* Corresponding author.
Tel: +82-(0)32-560-7332, E-mail : iamiyan@me.go.kr

1997). Especially, secondary aerosols containing sulfate, nitrate, and ammonium occupy the great portions of sub-micron aerosols. Recent studies have shown that secondary inorganic aerosols normally have four modes in size distribution with three modes in the size range smaller than 2 μm (Zhuang *et al.*, 1999a; Keywood *et al.*, 1999). The smallest mode extending from about 0.005 to 0.1 μm, is called the nuclei mode. It is produced by the condensation of primary pollutants or by the homogeneous condensation of secondary aerosol. The larger mode with the mass median aerodynamic diameter at about 0.2 μm, is called the condensation mode. It is not only produced by the gas phase reaction condensing hot vapors produced by combustion processes, but also made by the condensation of nuclei mode particles in the atmosphere. The other mode, with a peak at about 0.7 μm, is called the droplet mode. John *et al.* (1990) suggested the formation of the droplet mode from the condensation mode. This mode is usually formed by the processes related to the sulfate production in the ambient air (Meng and Seinfeld, 1994). Finally, the coarse mode normally has a peak at 3~5 μm in aerodynamic diameter, which is mainly generated by various mechanical processes.

The size distribution and chemical composition of atmospheric aerosols are important factors to determine the influence of anthropogenic pollutants. Various chemical components can usually have astonishingly similar size distributions in long-range transported aerosol (Han *et al*, 2004; Hillamo *et al.*, 1993) while the size distribution of specific chemical components can reflect the influence of particular anthropogenic sources (Keywood *et al.*, 2000). However, little work has been done on the chemical characterization of size-segregated aerosols of Asian plume. Therefore, size distribution characteristics of major ion components collected by a micro-orifice uniform deposit impactor (MOUDI) at Gosan, Jeju Island, Korea are mainly discussed in this paper to suggest the information on the size-resolved chemical characterization of Asian plume.

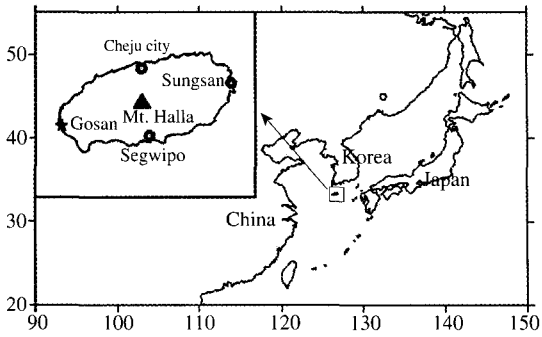

Fig. 1. Location of Gosan sampling site.

2. Experiment

2. 1 Sampling site and period

As shown in Fig. 1, Jeju is located in the boundary of China, Korea, and Japan, which is an ideal location to assess the long-range transport of anthropogenic and natural aerosols in East Asia region. The climate is temperate, tending to have distinctive four seasons. The predominant wind direction is north or northwest in spring, fall and winter and southeast in summer under the influence of Asian monsoon. Aerosol sampling was conducted at Gosan (33° 17′N, 126° 10′E, 70 m ASL) on the western tip of Jeju Island. The Gosan site has been a representative background monitoring site of many previous major studies related to the long range transport of air pollutants in East Asia (Han *et al.*, 2004; Moon *et al.*, 1998; Carmichael *et al.*, 1996). It was served as a super site of ACE-Asia program (Heubert *et al.*, 2003) and will become again a super site of ABC project (Ramanathan *et al.*, 2003). Atmospheric aerosol sampling was performed in four seasons; April 2002, September 2002, February 2003, and June 2003.

2. 2 Sampling and analysis

In this study, one set of 8-stage MOUDI (MSP Co., MSP-100) was operated to collect size-segregated samples of ambient aerosols at Gosan. The equivalent aerodynamic cut-off diameters of 8 stages are 0.218, 0.282, 0.532, 1.0, 1.8, 3.1, 6.2, 9.9,

and 18 μm, respectively. The MOUDI was installed on the roof of a sampling trailer at Gosan, which was located about 3 m above the ground. The MOUDI was operated at a flow rate of 30 L pm. The atmospheric aerosol samples were collected on 47 mm Teflon substrates (Zefluor™, 2.0 μm pore size, Gelman Sciences) and 37 mm Teflon filter (Teflo™, 2.0 μm pore size, Gelman Sciences) was used as a backup filter. Each sampling started at 9:00 AM and continued for more than 24 hours depending on the meteorological condition. Total 21 sets of samples were collected during the entire measurement periods. After sampling, the sampled filters were dried in a desiccator, and the mass of them were measured by a micro balance. Then, the filters were extracted with 1 mL of Methanol and 19 mL of distilled deionized water for 30 min by shaker and ultrasonicator, respectively. Then the extracts were filtrated using 1 μm cellulose filter. The eight water-soluble inorganic ions (SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Mg^{2+} and Ca^{2+}) of the collected particles were analyzed using ion chromatography (Dionex, DX-120) with an electric conductivity detector (Dionex CD-20). During the measurement period, concentrations of criteria pollutants; SO_2, NO_2, O_3, CO and PM_{10} were continuously measured at the Gosan background air pollution monitoring station. In addition, meteorological parameters such as wind, ambient temperature, relative humidity, and precipitation were measured at the Gosan meteorological observatory. These data were used to interpret the behavior and formation mechanisms of the atmospheric aerosols collected in this study.

3. RESULT AND DISCUSSION

3. 1 Meteorological conditions and criteria air pollutants

Only 8 sets among the 21 sets of MOUDI samples were selected to represent several episodic cases over the entire measurement period and applied to the following analysis. At this time, the concentrations of criteria pollutants during each sampling period were used in order to choose the polluted and non-polluted periods in each season. Average concentrations of PM_{10} and other criteria pollutants for the selected MOUDI sampling period are summarized in Table 1.

The variation of meteorological conditions and criteria air pollutants data during the eight sampling periods are shown in Fig. 2. Local wind direction was predominantly northwesterly and northerly in winter, fall, and spring, while the southerly and southeasterly wind was mainly observed under the influence of Asian monsoon in summer. Wind speed was faster in spring and winter than other seasons. An Asian dust (AD) outbreak occurred from 8 to 10 April 2002 resulting in elevated concentration of PM_{10} mass up to a maximum of 259 $\mu g/m^3$ on 9 April. The average concentration of PM_{10} mass in April 2002 was 3.6 times higher than that observed in June 2003 due to the influence of Asian Dust storms. SO_2 had the highest concentration of 5.0 ppb in February 2003.

3. 2 Particulate mass and ionic species concentrations

Average concentrations of mass and ion compo-

Table 1. Measurement periods of MOUDI.

Season	Sampling period	PM_{10} ($\mu g/m^3$)	SO_2 (ppb)	O_3 (ppb)	CO (ppm)	Remark
Spring	2002.4.7~4.10	179	1.25	49.2	0.47	Asian dust period
	2002.4.11~4.12	87.1	3.18	65.4	0.56	Non-Asian dust period
Summer	2003.6.16	51.4	1.00	39.2	0.60	Pollution period
	2003.6.13~6.15	30.7	0.92	34.2	0.42	Non-polluted period
Fall	2002.9.2~9.6	55.7	1.21	47.8	0.32	Pollution period
	2002.9.7~9.11	30.5	1.18	47.3	0.20	Non-polluted period
Winter	2003.2.18~2.20	68.7	6.74	32.0	0.58	Pollution period
	2003.2.14~2.17	28.9	4.71	29.6	0.45	Non-polluted period

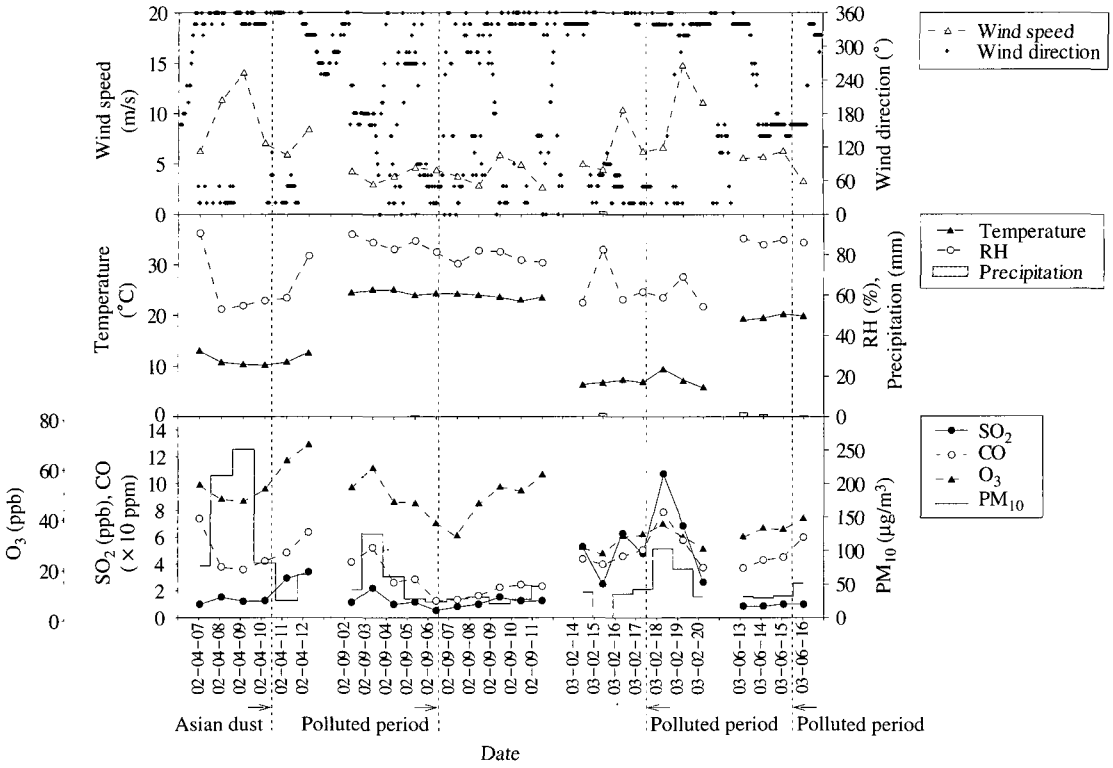

Fig. 2. Meteorological parameters (wind speed, wind direction, temperature, relative humidity, and precipitation) and criteria air pollutants (SO_2, O_3, PM_{10}, and CO) concentrations measured at Gosan during the measurement period.

nents in the three size modes are summarized in Table 2. The size ranges are divided based on the observed occurrence of sulfate modes (Seinfeld and Pandis, 1998) and the measured size distributions of aerosol chemical components. In this study, condensational, droplet, and coarse modes correspond to the size range of 0.218~0.532, 0.532~1.8, and 3.1~6.2 μm, respectively. Particle mass during other measurement periods except for the AD event was mostly distributed in the droplet mode with average mass concentrations ranging from 32.1 to 43.6 $\mu g/m^3$. More than 65% of sulfate, ammonium and potassium were present in the droplet mode, while nitrate particles were almost equally distributed in both the droplet (40.8%) and coarse modes (45.9%). Soil and sea salt components such as calcium, magnesium, sodium, and chloride were found mostly in the coarse mode. The concentrations of sulfate, ammonium, nitrate, and potassium in the droplet mode were higher in winter than in other seasons implying the influence of increased consumption of heating fuel and long-range transport of aerosol to Gosan.

3. 3 Size distribution of particle mass

Size distributions of particulate mass were observed to be either bimodal or trimodal regardless of sampling periods, as shown in Fig. 3. Especially, the particulate mass was bimodally distributed in winter, which is similar to the typical bimodal distribution observed in urban areas (Cheng and Tsai, 2000). Furthermore, condensation mode was not observed in both pollution and non-pollution periods in winter, implying that these particles were

Table 2. Average concentrations of mass and ionic species in three different size modes for each season.

(unit in (μg/m³).

Season	Mode	Mass	SO_4^{2-}	NO_3^-	NH_4^+	Cl^-	Na^+	K^+	Mg^{2+}	Ca^{2+}
Spring	Condensation	6.72	1.91	0.18	0.72	0.00	0.13	0.08	0.05	0.09
	Droplet	25.62	5.63	1.58	1.17	0.04	0.21	0.42	0.04	0.12
	Coarse	127.74	1.37	3.30	0.12	1.47	1.31	0.21	0.04	0.25
	Sum	160.08	8.91	5.06	2.02	1.52	1.65	0.72	0.13	0.45
Summer	Condensation	6.56	1.79	1.00	0.91	0.00	0.00	0.05	0.00	0.00
	Droplet	18.95	6.28	0.15	1.14	0.01	0.03	0.24	0.01	0.01
	Coarse	6.59	0.42	0.51	0.04	0.40	0.48	0.03	0.07	0.12
	Sum	32.10	8.48	1.67	2.09	0.41	0.51	0.31	0.08	0.13
Fall	Condensation	3.08	0.99	0.05	0.61	0.03	0.11	0.02	0.00	0.00
	Droplet	19.59	6.63	1.00	2.06	0.16	0.31	0.37	0.05	0.06
	Coarse	14.14	0.47	1.88	0.12	3.39	1.56	0.09	0.25	0.35
	Sum	36.82	8.09	2.93	2.78	3.58	1.98	0.48	0.30	0.41
Winter	Condensation	8.80	3.06	0.82	1.29	0.14	0.01	0.14	0.00	0.00
	Droplet	25.54	9.55	3.52	3.13	0.27	0.18	0.67	0.05	0.08
	Coarse	9.20	0.57	1.34	0.08	1.06	0.60	0.04	0.09	0.27
	Sum	43.55	13.18	5.67	4.51	1.47	0.80	0.85	0.14	0.35
Total	Condensation	6.29 (9.2%)	1.94 (20.0%)	0.51 (13.4%)	0.88 (31.0%)	0.04 (2.5%)	0.06 (5.2%)	0.07 (12.4%)	0.01 (7.4%)	0.02 (6.8%)
	Droplet	22.43 (32.9%)	7.02 (72.7%)	1.56 (40.8%)	1.87 (65.8%)	0.12 (7.0%)	0.18 (14.8%)	0.42 (72.1%)	0.04 (23.7%)	0.07 (20.0%)
	Coarse	39.42 (57.9%)	0.71 (7.3%)	1.76 (45.9%)	0.09 (3.3%)	1.58 (90.5%)	0.99 (80.0%)	0.09 (15.5%)	0.11 (68.9%)	0.25 (73.1%)
	Sum	58.14	9.66	3.83	2.85	1.75	1.24	0.59	0.16	0.34

Note 1: Condensation mode : 0.218~0.532 μm; Droplet mode : 0.532~1.8 μm; and Coarse mode : 3.1~6.2 μm.
2: Parenthesis ( ) represents the percent fraction of each mode.

sufficiently aged enough to be transformed the condensation mode particle into the droplet mode one. Considering the dominant northerly windward in the same period and the fact that Gosan is a relatively clean area free from the industrial pollution sources, it is also inferred that the remarkable occurrence of the droplet mode particles in winter is associated with high relative humidity above marine areas (Zhuang *et al.*, 1999a) during long-range transport of particulate pollutants. The droplet mode diameter varied from 0.73 μm in winter to 1.4 μm in summer due to the high relative humidity (86%) condensation in summer season. Coarse mode mass was the highest in spring due to the influence of AD storm. Coarse mode particle mass concentration (Fig. 3(a)) during the AD period peaked with the mode diameter was about 7 μm. During the pollution periods, the remarkable increase of the droplet mode mass was commonly observed. In addition, condensation mode mass also increased in the pollution period of fall under the lowest wind speed and stagnant air mass conditions, implying that relatively close local sources could partially con-

Table 3. The average size distribution of equivalent ion concentration. **(μeq/m³)**

	Condensation mode	Droplet mode	Coarse mode
Anion	0.050	0.175	0.088
SO_4^{2-}	0.040 (80.9%)	0.008 (16.6%)	0.001 (2.5%)
NO_3^-	0.146 (83.6%)	0.025 (14.4%)	0.003 (2.0%)
Cl^-	0.015 (16.8%)	0.028 (32.4%)	0.045 (50.9%)
Cation	0.056	0.129	0.072
NH_4^+	0.049 (87.8%)	0.003 (5.0%)	0.002 (3.3%)
Na^+	0.001 (1.8%)	0.001 (2.1%)	0.104 (80.4%)
K^+	0.008 (6.2%)	0.011 (8.4%)	0.003 (2.4%)
Mg^{2+}	0.003 (2.6%)	0.005 (7.2%)	0.043 (59.8%)
Ca^{2+}	0.002 (3.2%)	0.009 (12.7%)	0.012 (17.0%)

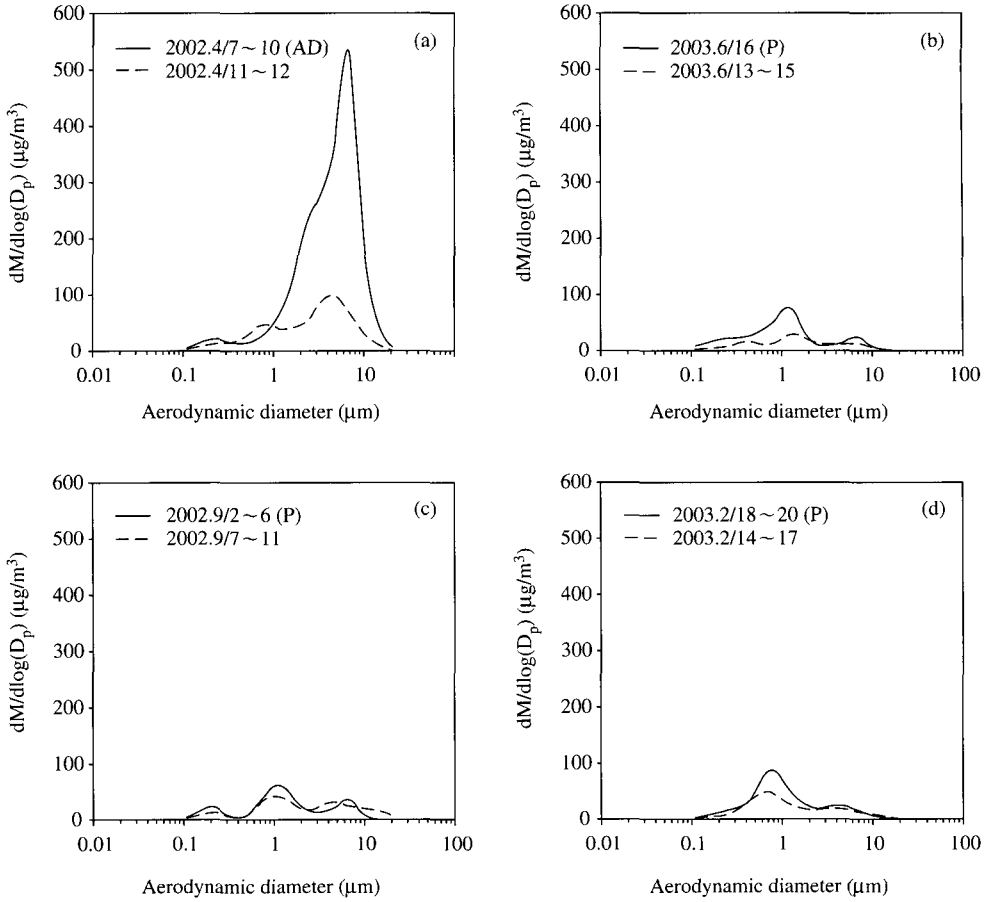

Fig. 3. Size distributions of mass concentration ((a) spring, (b) summer, (c) fall, (d) winter); AD means Asian dust outbreaks, and P means Pollution period.

tribute to the ambient aerosols.

3. 4 Size distributions of water-soluble ion components

The equivalent concentration of each ion component observed in the three size modes are summarized in Table 3. Sulfate and ammonium accounted for more than 80% of total anion and cation concentrations in both the condensation and droplet modes. On the other hand, chloride and sodium in the coarse mode hold more than 50% of total anion and cation concentrations. Moreover, nitrate and sum of magnesium and calcium individually occupied about 30% in the coarse mode. The equivalent mass concentrations of anion and cation are shown in Fig. 4 for each measurement period. Similar shapes and concentration levels were observed in both anion and cation distribution profiles. This means that the measured anion components such as sulfate, nitrate, and chloride were nearly combined with cation species like ammonium, sodium, potassium, calcium, and magnesium. The difference between anion and cation equivalent concentrations was mainly observed in the droplet mode. The gap increased during the pollution period in the summer and winter, implying that it was primarily caused by the undetermined ion components including hydrogen.

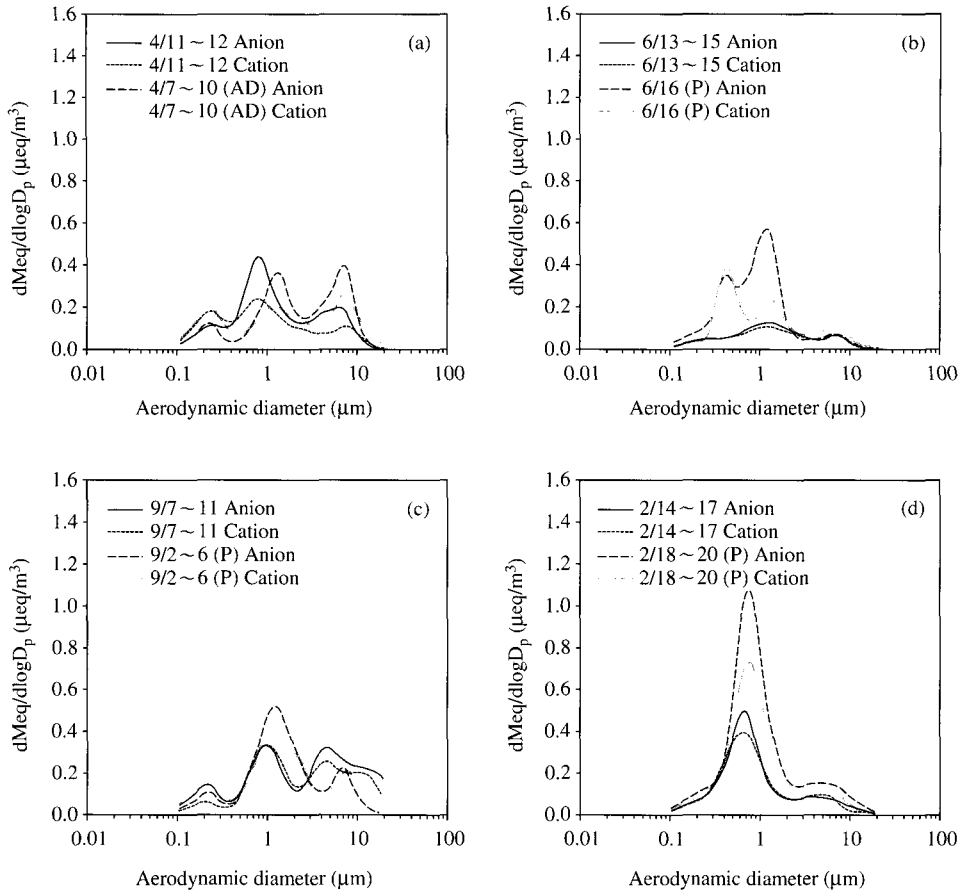

Fig. 4. Size distribution of anion and cation equivalent concentrations ((a) spring, (b) summer, (c) fall, (d) winter).

Therefore, it is inferred that the acidic pollutants such as sulfuric acid and nitric acid could influence the fine aerosol (0.532~1.8 μm) during the pollution periods.

Size distribution was estimated as shown in Fig. 5 using the equivalent size distribution of water-soluble ion components.

Size distributions of sulfate, ammonium, and potassium particles varied from unimodal to trimodal. However, the droplet mode commonly dominated the size distribution of these three ions. Generally, the droplet mode cannot be explained by primary emission, gas-phase nucleation, and condensation (Zhuang *et al.*, 1999a). It is usually formed by the activation of the condensation mode particles, followed by aqueous-phase chemistry and water evaporation (Seinfeld and Pandis, 1998). Considering there is no large industrial emission sources aroused at Gosan and the rainy days data are excluded in this analysis, it is inferred that the droplet mode particles could be formed by the transformation of anthropogenic pollutants emitted from distance sources during the long-range transport.

Ammonium can not only be accumulated in the droplet mode when ammonia vapor reacts or condenses on an acidic particle sulfate, but also form the condensation mode particles when heteromolecular nucleation of ammonia gas with some acidic gases occurs (Zhuang *et al.*, 1999a). At this time, ammonia tends to react with sulfuric acid or

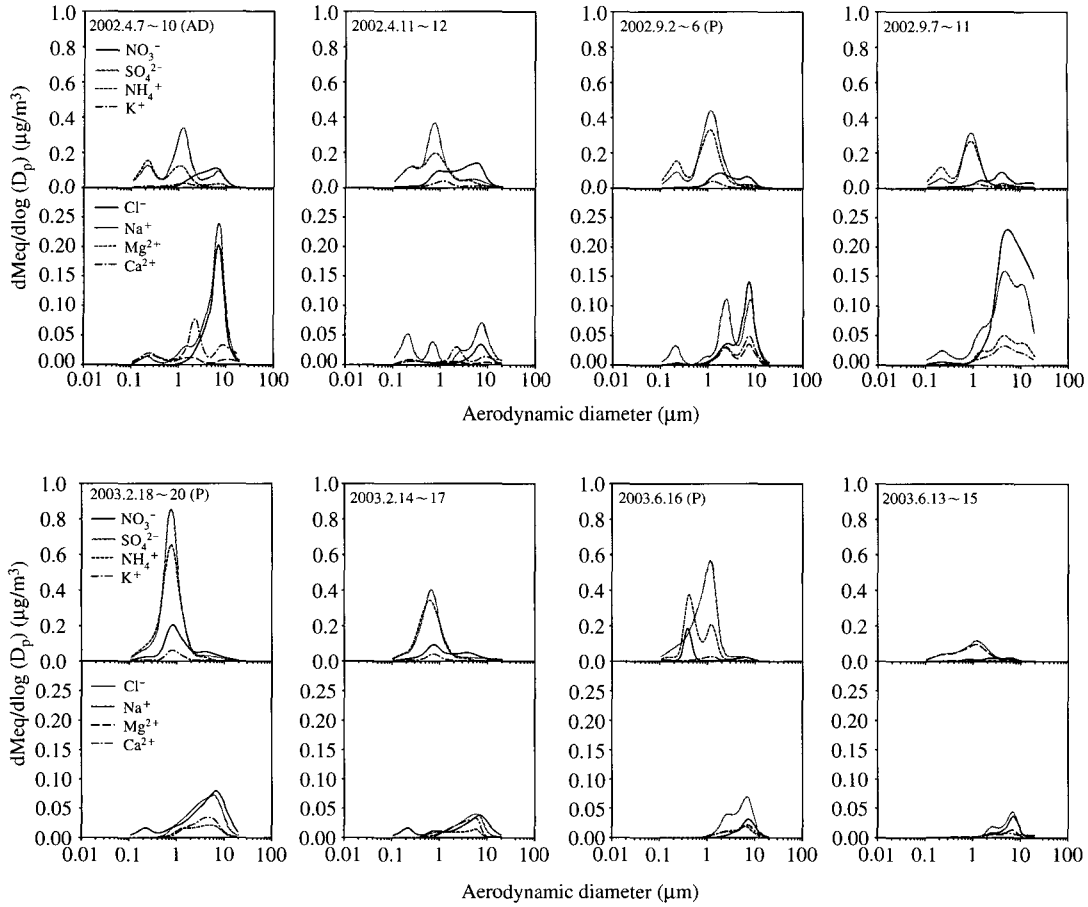

Fig. 5. Size distribution of equivalent concentrations of particulate ion components.

sulfate because ammonium sulfate is most stable among the products of the reaction. Concentrations of droplet mode sulfate and ammonium were the highest in winter, and existed in sufficiently neutralized states. In spring and fall, condensation mode sulfate and ammonium were also observed implying that these components originated from relatively close source regions because the condensation mode particles easily grow up affected by the high relative humidity at a coastal area (Zhuang *et al.*, 1999a). Existing form of ion components was estimated by stoichiometric equivalent ratios of ammonium containing particles. As shown in Fig. 6a, the equivalent ratios of NH_4^+/SO_4^{2-} had values above 0.5 in the fine mode indicating that $(NH_4)_2SO_4$ (ratio = 1.0) and $(NH_4)_3H(SO_4)_2$ (ratio = 0.75) mainly contributed to the fine particles.

The fine mode potassium (0.5 ~ 1.5 μm or < 1.1 μm) has been reported in previous works on the size distribution of ion components in East Asia (Topping *et al.*, 2004; Matsumoto *et al.*, 1998). Arimoto *et al.* (1995) and Cheng *et al.* (2000) suggested the soluble potassium as a tracer of biomass burning because it was known that large amounts of biomass in China were consumed by open burning. In this study, the droplet mode potassium had the highest value in winter. On the other hand, most of sea salt or soil components, such as chloride, sodium, magnesium, and calcium, were mainly distributed in the coarse mode. At this time, it is inferred that sea

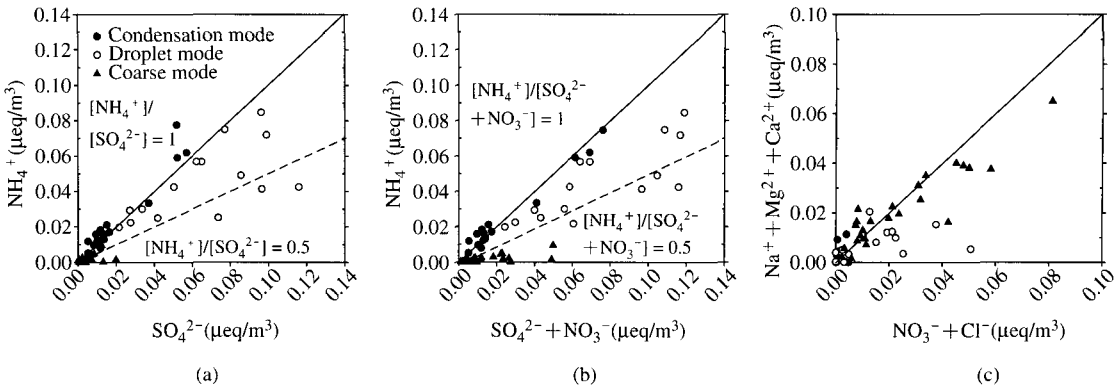

Fig. 6. Equivalent molar ratio of major ion components in three size modes.

salt could exist in combination with nitrate or sulfate considering that the equivalent ratio of chloride and sodium didn't always correspond one to one and that nitrate and sulfate were considerably distributed not only in the fine particles, but also in the coarse particles. For the soil components, it is difficult to estimate the existing form because the concentrations of carbonate and oxygen were not measured in this study. But the fact that the equivalent ratio of $(Na^+ + Mg^{2+} + Ca^{2+})/(NO_3^- + Cl^-)$ was about 1.0 in the coarse mode particles as shown in Fig. 6c suggests that not only sea salt but also soil could be transformed into the compound including nitrate. Therefore, coarse mode nitrate and sulfate could be mainly formed by the reaction of gas-phase nitric acid with sea salt or soil particles into several forms such as $NaNO_3$, $Mg(NO_3)_2$, and $Ca(NO_3)_2$.

3. 5 Asian Dust storm period

AD outbreaks were observed from 8 to 10 April 2002. In this period, average mass concentration of PM_{10} was 179 μg/m^3, which is more than 2 times higher than that observed from 11 to 12 April after the AD events. In addition, Ca^{2+} and Mg^{2+} also had higher concentrations due to huge influence of soil dust. The highest loading of Ca^{2+}, 0.47 μg/m^3, was found during this period. Nitrate also had highest concentration in the coarse mode implying the association with soil dust particles. Sea salt particles in the coarse size range were also prevalent as shown in Fig. 7 during the AD period due to the influence of the higher wind speeds.

3. 6 Pollution periods

Except for the AD period, the elevated concentrations of gaseous pollutants and PM_{10} were observed as summarized in Table 1, from 2 to 6 September 2002, from 18 to 20 February 2003, and on 16 June 2003. The average concentration of PM_{10} in these periods was 58.6 μg/m^3, which is about 2 times higher than the other periods value of 30.0 μg/m^3. During these pollution periods, the droplet mode sulfate and ammonium had much higher concentrations than those in non-pollution periods, as shown in Fig. 8. Moreover, the considerable increase of nitrate was also observed in the droplet mode. This elevation of secondary aerosol components in the droplet mode suggests the influence of anthropogenic pollutants transported from the emission sources. Especially, the condensation mode nitrate and ammonium were sharply increased in summer pollution period as shown in Fig. 5. The high concentration of condensation mode nitrate generally means that it is emitted from the relatively close region.

3. 7 Chloride depletion

During the sampling periods in April 2002 and June 2003, concentration of sodium ion was higher

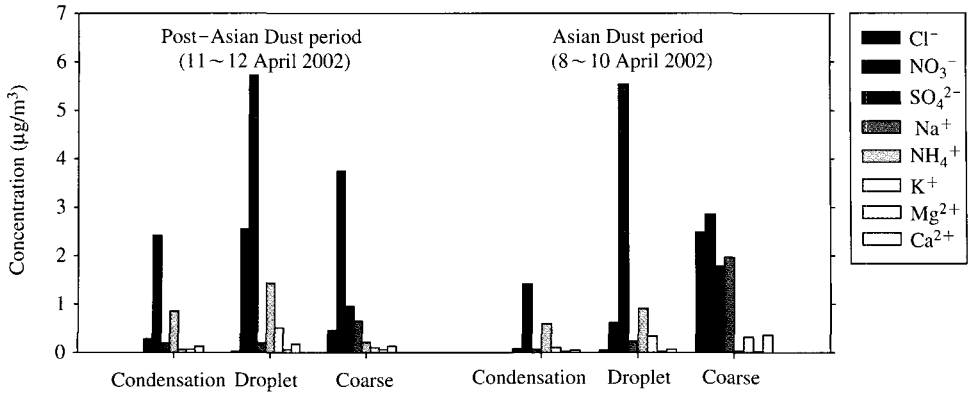

Fig. 7. Mass concentration of chemical components measured in the AD and post-AD period.

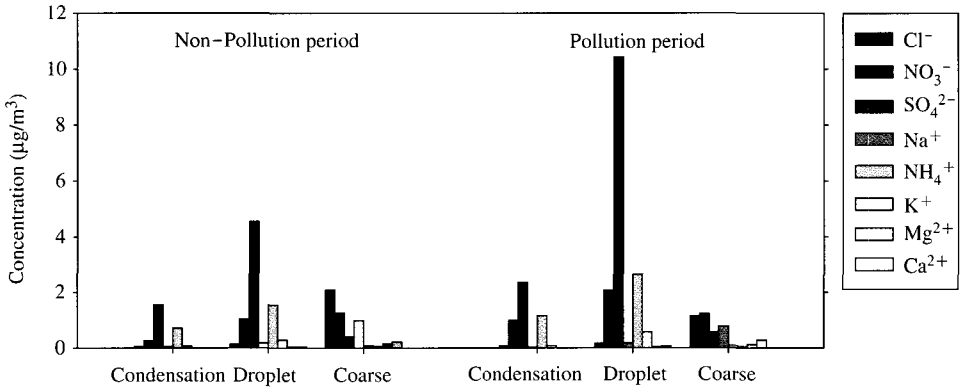

Fig. 8. Mass concentration of chemical components measured in the pollution periods and non-pollution periods.

than that of chloride. This result implies that chloride depletion could be considerably arisen during these periods. Fig. 9 shows the size distribution of chloride depletion during all measurement periods. The percentage of chloride depletion is calculated as:

$$Cl_{dep}(\%) = ([Cl^-]_{sea\ salt} - [Cl^-])/[Cl^-]_{sea\ salt} \times 100\%$$

where $[Cl^-]_{sea\ salt} = 1.174 \times [Na^+]$. $[Cl^-]$ and $[Na^+]$ are the measured equivalent concentrations of chloride and sodium (Zhuang *et al*, 1999b). Overall, the extent of chloride depletion decreased with increasing particle size. The loss of chloride was also considerably influenced by the location of the sulfate peak. The amount of chloride depletion revealed the high percentage in the size range of 1~3 μm near by the peak of droplet mode sulfate (0.9~2 μm). This trend can be explained by the evaporation of NH_3 together with HCl (Yao *et al.*, 2003; Pakkanen, 1996). The total amount of chloride depletion and ammonia evaporation was the largest in summer and was the smallest in winter. The result shows well that the loss of chloride significantly depends on the meteorological condition such as relative humidity and temperature (see Fig. 2) (Chang *et al.*, 2000). Furthermore, the amount of chloride depletion was larger in pollution periods because of the large amount of sea salt

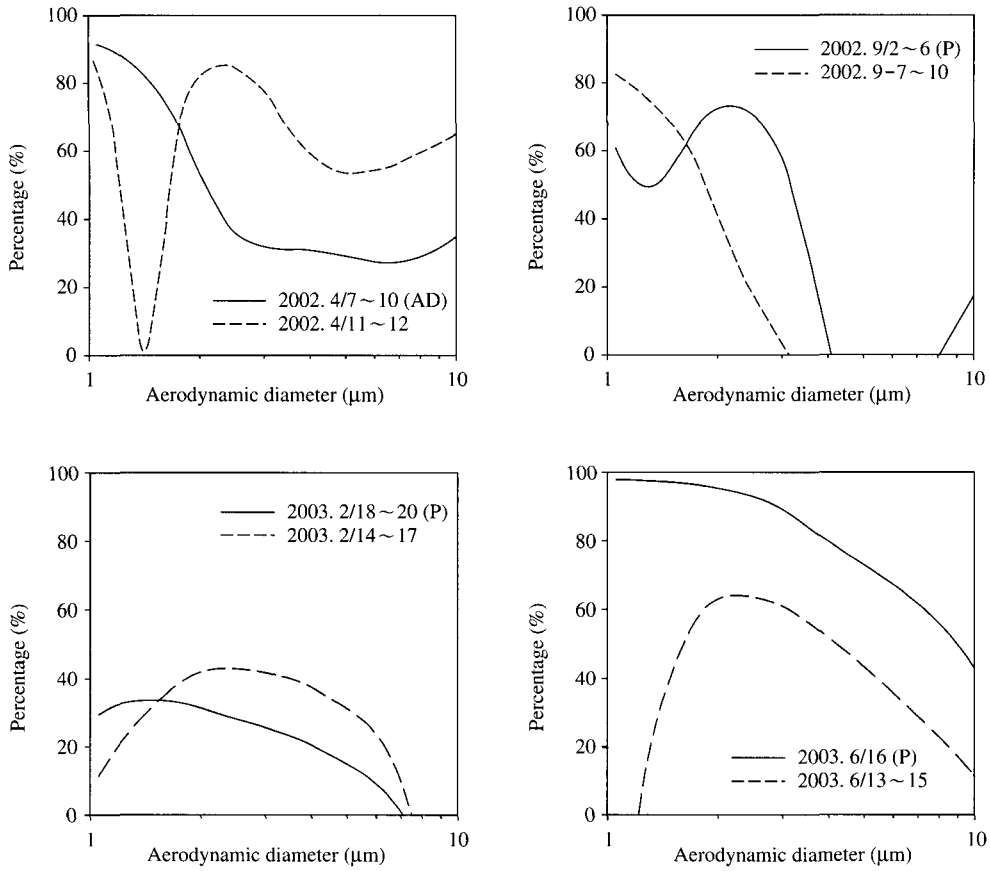

Fig. 9. Size distribution of the percentage of chloride depletion.

compound such as NH_4Cl than in non-pollution periods in summer.

4. Conclusion

Eight sets of MOUDI samples were collected to analyze the size distribution of particulate chemical composition at Gosan in Jeju from April 2002 to June 2003. Aerosol mass and major ion components primarily showed trimodal size distribution including condensation, droplet, and coarse modes, corresponding to the size ranges 0.218~0.532 μm, 0.532~1.8 μm, and 3.1~6.2 μm, respectively. Mass and secondary aerosol components were mainly distributed in the droplet mode. Because the droplet mode is normally formed by the secondary reaction of primary pollutants, it can be inferred that the droplet mode mass and sulfate could be formed during long-range transport of anthropogenic pollutants. Equivalent ratio and the very similar distribution in sulfate and ammonium particles imply that these ion components were transported together. On the other hand, sea salt and soil components were mainly distributed in the coarse mode, partially combined with the coarse mode nitrate and sulfate. The concentration of droplet mode ammonium sulfate was higher in winter than those in other seasons. On the other hand, chloride depletion and ammonia evaporation was chiefly

observed in summer by the influence of the high temperature and relative humidity. During the AD period, the concentrations of soil and sea salt components were high in droplet and coarse modes. Finally, sulfate, ammonium, and nitrate particles in the droplet mode size range sharply increased during several pollution periods, implying the transport of anthropogenic pollutants.

ACKNOWLEDGEMENT

This work was supported by the project of Ministry of Environment, Korea; "Long Range Transport of Toxic Trace Substances: Identification and Impact Analysis" and in part by the Korea Science and Engineering Foundation (KOSEF) through the Advanced Environmental Monitoring Research Center (ADEMRC) at Gwangju Institute of Science and Technology.

REFERENCES

Alves, C.A., C.A. Pio, and A.C. Duarte (2000) Particle size distributed organic compounds in a forest atmosphere., Environ. Sci. Technol., 25, 2133-2140

Bae, G.N., J.H. Ji, and K.C. Moon (1999) Collection characteristics of a MOUDI cascade impactor for coarse particles, KOSAE, 15(6), 799-804.

Carmichael, G.R., Y. Zhang, L. Chen, M.S. Hong, and H. Ueda (1996) Seasonal variation of aerosol composition at Cheju Island, Korea, Atmospheric Environment, 30, 2407-2416.

Chang, M.C., C. Soiutas, S. Kim, H. Gong Jr., and W.S. Linn (2000) Reduction of nitrate losses from filter and impactor samplers by means of concentration enrichment, Atmospheric Environment, 34, 85-98.

Chang, K.F., G.C. Fang, C.S. Lu, and H.L. Bai (2001) Particle size distribution and anion content at a traffic site in Sha-Lu, Taiwan, Chemosphere, 45, 791-799.

Cheng, M.T. and Y.I. Tsai (2000) Characterization of visibility and atmospheric aerosols in urban, suburban, and remote areas, the Science of the Total Environ., 263, 101-114.

Han, J.S., K.J. Moon, J.Y. Ahn, Y.D. Hong, Y.J. Kim, S.Y. Ryu, S.S. Cliff, and T.A. Cahill (2004) Characteristics of ion components and trace elements of fine particles at Gosan, Korea in spring time from 2001 to 2002, Environ. Monitoring and Assessment, 92, 73-93.

Han, J.S., K.J. Moon, S.Y. Ryu, Y.J. Kim, and K.D. Perry, Source estimation of anthropogenic aerosols collected by a DRUM sampler during spring of 2002 at Gosan, Korea, Atmospheric Environment, accepted 19 January 2005.

Hillamo, R.E., V.M. Kerminen, W. Maenhaut, J.L. Jaffrezo, S.C. Balachandran, and I. Davidson (1993) Size distributions of atmospheric trace elements at Dye 3, Greenland-I. Distribution characteristics and dry deposition velocities, Atmospheric Environment 27A, 2787-2802.

Keywood, M.D., G.P. Ayers, J.L. Gras, R.W. Gillett, and D.D. Cohen (1999) Relationships between size segregated mass concentration data and ultrafine particle number concentrations in urban areas, 1999, Atmos. Environ., 33, 2907-2913.

Keywood, M.D., G.P. Ayers, J.L. Gras, and R.W. Gillett (2000) Size distribution and sources of aerosol in Launceston, Australia, during Winter 1997, Air & Waste Manage. Assoc., 50, 418-427.

Marple, V.A., K.L. Rubow, and S.M. Behm (1991) A Microorifice Uniform Deposit Impactor (MOUDI): description, calibration, and use, Aerosol Sci. and Tech., 14, 434-336.

Matsumoto, K., I. Nagao, H. Tanaka, H. Miyaji, T. Iida, and Y. Ikebe (1998) Seasonal characteristics of organic and inorganic species and their size distributions in atmospheric aerosols over the northwest pacific ocean, Atmospheric Environment, 31, 1931-1946.

Meng, Z. and J.H., Seinfeld (1994) On the source of the submicrometer droplet mode of urban and regional aerosols, Aerosol Science and Technology 20, 253-265.

Moon, K. *et al.* (1998) Study on long-range transport of air pollutants in Northeastern Asia (III) (in Korean). Report to National Institute of Environment Research, Korea, Report No. BSG557-6191, Korea Institute of Science and Technology, Seoul, Korea.

Pakkanen, T.A. (1996) Study of formation of coarse particle nitrate aerosol, Atmospheric Environment, 30, 2475-2482.

Park, S.U. and L.S. Chang (2000) A numberical simulation of a Yellow Sand event observed in Korea during 14~23 April, Journal of Applied Meteorology 13, 863-870.

Ramanathan, V., G. Carmichael, P.J. Crutzen, B. Holben, K. Prather, J. Prospero, and D. Savoie, J. Schauer (2003) *Regional Aeorosol-Chemistry-Climate Observatories for the Indo-Asia-Pacific Region*, Proposal submitted to NOAA.

Seinfeld, J.H. and S.N. Pandis (1998) Atmospheric Chemistry and Physics from Air Pollution to Climate Change, John Wiley & Sons, Inc., New York, 429-451.

Topping, D., H. Coe, G. McFiggans, R. Burgess, J. Allan, M.R. Alfarra, K. Bower, T.W. Choularton, S. Decesari, and M.C. Facchini (2004) Aerosol chemical characteristics from sampling conducted on the Island of Jeju, Korea during ACE Asia, Atmospheric Environment, 38, 2111-2123.

Tuch, T., P. Brand, H.E. Wichmann, and J. Heyder (1997) Variation of particle number and mass concentration various size ranges of ambient aerosols in eastern Germany, Atmos. Environ., 31, 4193-4197.

Twomey, S. (1975) Comparison of constrained linear inversion and an iterative nonlinear algorithm applied to the indirect estimation of particle size distributions, J. of Compu. Phys., 18, 188-200.

Yao, X., M. Fang, and C.K. Chan (2003) The size dependence of chloride depletion in fine and coarse sea-salt particles, Atmospheric Environment, 37, 743-751.

Zhuang, H., C.K. Chan, M. Fang, and A.S. Wexler (1999a) Formation of nitrate and non-sea-salt sulfate on coarse particles, Atmospheric Environment, 33, 4223-4233

Zhuang, H., C.K. Chan, M. Fang, and A.S. Wexler (1999b) Size distributions of particulate sulfate, nitrate, and ammonium at a coastal site in Hong Kong, Atmospheric Environment, 33, 843-853.