Aerosol composition change between 1992 and 2002 at Gosan, Korea

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[1] TSP composition data and backward trajectory analysis results at Gosan, Korea, for 10 years between March 1992 and February 2002 are studied to understand the site characteristics and the relationship between the aerosol composition and meteorological conditions. The average non-sea-salt-(nss)-sulfate concentration (6.74 $\mu g\ m^{-3}$) is higher than those observed at other background areas in the world. It was demonstrated that the observed high level of sulfate is due to transport from outside the site. The concentrations of nss-sulfate/ammonium/nss-potassium, sodium/chloride/magnesium, and nss-calcium/nitrate show a strong relationship, suggesting their common emissions sources and/or transport pattern. It is likely that the concentration ratio of nss-sulfate to nitrate is decreasing because of the increase of the nitrate concentration. On the basis of various measurement and emission estimate studies, it is suggested that this trend is mainly caused by the emission trend change in China. About half of the air mass trajectories are from northern China, and a quarter are from southern China. On the basis of cluster analysis, it was found that when air parcels moved from China, the concentrations of nss-sulfate, ammonium, and nitrate are the highest. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0325 Atmospheric Composition and Structure: Evolution of the atmosphere; 0335 Atmospheric Composition and Structure: Ion chemistry of the atmosphere (2419, 2427); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); *KEYWORDS:* long-term trend, Gosan, TSP

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1. Introduction

[2] Northeast Asia, including Korea, China, and Japan, is characterized by high emission of both anthropogenic and natural ambient trace species. Among them, China emits an overwhelming fraction [*Streets and Waldhoff*, 2000; *Streets et al.*, 2000]. Thus it is important to understand the effects of the emissions and transport of ambient trace species from northeast Asia, especially, China, on the regional air quality and global environment.

[3] Jeju Island is located at about 100 km south of the Korean Peninsula, about 500 km west of China (Jiangsu province), and about 200 km east of the Japanese Islands (Kyushu), as shown in Figure 1. Jeju Island is one of the cleanest areas in Korea, with low emissions of air pollutants [*Kim et al.*, 1998a]. Thus Jeju Island is an excellent location to study the transport and transformation of ambient trace species in northeast Asia and to study the impact of continental outflow.

[4] Gosan is located at the far western edge of the island, the least developed area on the island on the grounds of a meteorological station. Several intensive and routine measurement studies including PEM-West A and B have been carried out at Gosan [*Arimoto et al.*, 1996; *Chen et al.*, 1997; *Kim et al.*, 1998a, 1998b, 1999a, 1999b; *Lee et al.*, 2001]. Gosan was one of the supersites during the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia). *Huebert et al.* [2003] claimed that aerosols observed at Gosan were frequently impacted by Asian desert dust storms and anthropogenic sources located in China, Korea, and Japan depending on its air mass history.

[5] Long-term measurement data of aerosol composition, especially with meteorological conditions, are important in understanding the characteristics of a site. Since Gosan was a supersite during ACE-Asia, it is important to

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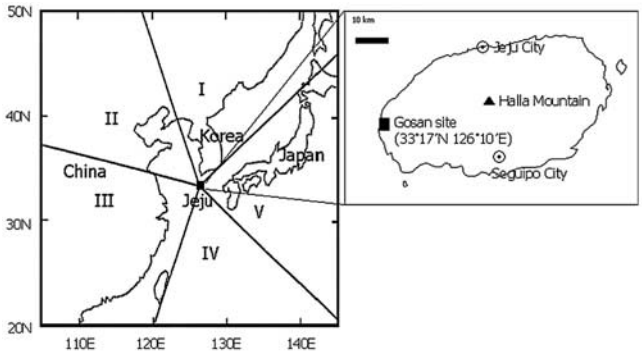

Figure 1. Map of the study site, Gosan, Jeju Island, Korea, and the sectors used for the sector analysis.

understand the site characteristics. In this work, the aerosol ion composition data measured at Gosan, Jeju Island, and Korea and backward trajectory analysis data between March 1992 and February 2002 are used to study the site characteristics and the transport pattern of aerosol in the region. Variations of aerosol composition are analyzed. Then, annual trends of the nitrate and sulfate concentrations, important acidic components of aerosols, are presented and discussed using nonparametric and long-term statistical analysis methods. Also, cluster analysis is carried out to understand the relationship between the ion composition and air parcel trajectory.

2. Sampling and Analysis

[6] Gosan is located on the western tip of Jeju Island (126°10′E, 33°17′N), as shown in Figure 1. A trailer containing the TSP sampler is situated about 10 m inland from the cliff, which is about 70 m above sea level. The sampling inlet is located about 6 m above the ground. The Jeju upper air meteorological station is located 100 m northeast of the site and measures upper air meteorological parameters twice a day and other meteorological parameters.

[7] Particles were collected by a high-volume tape sampler (Kimoto Electric Co., model 195A), which is an automatic sampling system with roll type PTFE filters (Sumitomo Electric, 100 mm × 10 m). Particles were collected for either 6 or 24 hours, and then a new clean filter surface was moved to the sampling area on which particles were collected. The flow rate was about 170 LPM. All the data presented in this work are the 24-hour-averaged results. The annual mean concentration is calculated from the data between March of one year and February of the next year.

[8] Eight ions were analyzed; NH_4^+ was analyzed by the indophenol method with a UV-visible spectrophotometer, and Na^+, K^+, Ca^{2+}, and Mg^{2+} were analyzed by atomic absorption spectroscopy. Anions (SO_4^{2-}, NO_3^-, and Cl^-) were analyzed by anion chromatography. Non-sea-salt-(nss)-K^+, nss-Ca^{2+}, nss-Mg^{2+}, and nss-SO_4^{2-} concentrations are estimated by assuming that all Na^+ are from sea salt and subtracting the sea salt composition. Details on the ion analysis and quality assurance/quality control (QA/QC) are given by *Kim et al.* [1998a].

[9] To ensure the quality of the data, three steps of quality control procedures were taken. First, an instrument quality check was carried out. Second, the sampling and analysis QA/QC procedures were checked. Third, ion balance was used to check the validity of the data. The data for which the ratio of the sum of the equivalent cation concentrations to the equivalent anion concentrations was within 30% are used for further data analysis. The criterion of 30% was chosen since it was found from the rain sample analysis and aerosol elemental analysis results that organic ions and carbonates could be up to 30% of the total anion concentrations at Gosan. Organic acids may be a nonnegligible part of the anions during summer, when biogenic emissions of organic compounds are high, and the carbonates during spring, when dust storms are frequent. However, these were not analyzed in this work. Among the total data sets of 1531, 145 data sets (about 9.5%) are discarded on the basis of these three quality checks. In this paper, the remaining 1386 data sets are used for further analysis.

3. Results and Discussion

3.1. Long-Term Trend of Aerosol Composition

[10] Table 1 shows the mean concentrations of the analyzed ions. Among the ions, the mean concentration of nss-sulfate is the highest (6.74 μg m^{-3}). More than 90% of the total sulfate are nss-sulfate. This shows that the contribution of sea salts to TSP sulfate is small although the Gosan site is located at the seashore. This agrees with the results of other

Table 1. Comparison of the Ion Concentrations in This Study With Other Study Results[a]

References	Place	Period	Particle Size	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}	SO_4^{2-}	NO_3^-	Cl^-	nss-K^+	nss-Ca^{2+}	nss-Mg^{2+}	nss-SO_4^{2-}
This study	Gosan, Korea	March 1992 to Feb. 2002	TSP	1.50	1.83	0.41	0.52	0.27	7.20	1.58	1.82	0.34	0.45	0.07	6.74
Kim et al. [1998b]	Gosan, Korea	July 1994 to Aug. 1994	TSP	0.89	4.95	1.34	0.54	0.36	12.10	1.78	3.16	1.17	0.36	−0.23	10.86
Hatakeyama [1993]	Nagano, Japan	March 1994	TSP	0.66	0.15	0.08	0.27	0.04	2.99	0.48	0.12				
Hatakeyama [1993]	Okinawa, Japan	March 1994	TSP	0.31	5.90	0.27	0.64	0.70	4.12	0.95	9.78				2.65
Mukai et al. [1990]	Oki, Japan	Dec. 1983 to May 1988	PM_{10}	0.51	0.43	0.13	0.15	0.11	3.59	0.11	0.05				
Lee et al. [2001]	Gosan, Korea	1996–1997	$PM_{2.5}$	1.48	0.64	0.26	0.15	0.05	5.13	0.67	0.55				
Lee et al. [2001]	Gangwha, Korea	1996–1997	$PM_{2.5}$	3.61	0.56	0.58	0.37	0.46	5.98	3.35	0.57				
Chow et al. [1994]	San Nicolas, United States		$PM_{2.5}$	0.68					2.77	0.46					

[a]Units are μg m^{-3}.

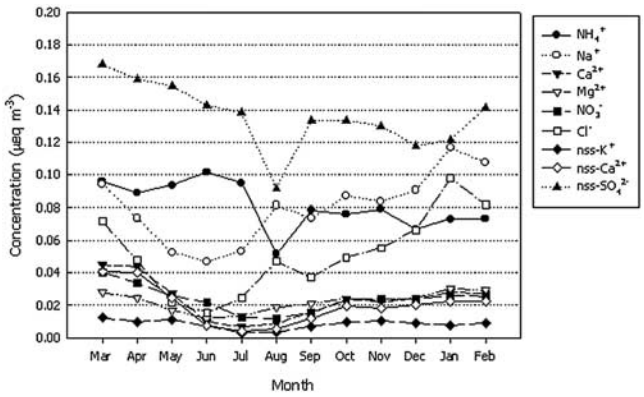

Figure 2. Variations of the monthly mean ionic concentrations.

studies using different samplers for TSP [*Kim et al.*, 1998a, 1998b] and $PM_{2.5}$ [*Lee et al.*, 2001] at Gosan. Since the contribution of biogenic emissions of sulfur species to nss-sulfate at Gosan is small, about 10% [*Arimoto et al.*, 1996], nss-sulfate measured at Gosan should have anthropogenic origins. Since the ambient concentration of SO_2 at Gosan is low, about 1 ppb [*Kim et al.*, 1998a], and other factors for the accumulation of ambient trace species are not significant, such as land-sea breeze [*Kim et al.*, 1995], it is likely that the majority of the observed nss-sulfate should be from outside Gosan. A similar conclusion was observed for the measurement of carbonaceous species at Gosan [*Kim et al.*, 1999a] and by ACE-Asia measurement strategy [*Huebert et al.*, 2003].

[11] The mean ratio of the sum of the cation concentrations (0.222 μeq m^{-3}) to the sum of the anion concentration (0.227 μeq m^{-3}) is 0.98. Major anions are sulfate and chloride, and major cations are ammonium and sodium. The equivalent ratios of ammonium to nss-sulfate are, for most data, below 1 with a mean equivalent ratio value of 0.60. Thus nss-sulfate in TSP at Gosan is not sufficiently neutralized by ammonium. On the contrary, the equivalent ratio is close to 1 (about 0.87) for $PM_{2.5}$ particles at Gosan [*Lee et al.*, 2001] because of preferential distribution of ammonium on the fine fraction of the particles [*Kim et al.*, 1999b].

[12] The results of this study are compared with the results of other studies in Table 1. The mean concentration of nss-sulfate (6.74 μg m^{-3}) of this study is higher than that at other marine background sites in the world. The concentration of nss-sulfate is 3.20–4.80 μg m^{-3} at Okinawa, Japan, 0.37–0.70 μg m^{-3} at Midway, in the Pacific, and 0.37–0.70 μg m^{-3} at Oahu, Hawaii [*Arimoto et al.*, 1997]. The mean concentration of sulfate of this study (7.20 μg m^{-3}) is higher than that at Nagano, Japan (0.57–2.99 μg m^{-3}, *Hatakeyama* [1993]), a mountainous background area, or San Nicolas Islands, United States (2.77 μg m^{-3}, *Chow et al.* [1994]), a marine background area of the Los Angeles region. Thus, considering the ambient level of SO_2 at Gosan and the emission strength of sulfur species in Jeju, it also suggests that the Gosan site is affected by the outside.

[13] Figure 2 shows the variation of the monthly mean concentrations of ions at Gosan. The concentrations of nss-sulfate, nss-potassium, nss-calcium, and nitrate peak in March and are the highest during spring. The high concentrations of these species in spring are related to the strong continental outflows associated with cold front passage, while the low concentration in summer is related to warm southerly flows that transport low-latitude marine air to Jeju as described by *Chen et al.* [1997] and high precipitation at Gosan. During spring, frequent migratory anticyclones are related to the passage of cold fronts and dust storms; both are favorable to continental outflow. The concentrations of sulfate and ammonium drop in August because the prevailing wind direction is southeast and there is higher precipitation in August. *Mukai et al.* [1990] also observed the concentration of sulfate drops during the wet season at Oki Island, located in the western part of the Japanese main island. The concentration of ammonium peaks between June and July because of high emissions during summertime, from, for example, animal excrement and applications of fertilizer. The concentrations of sodium and chloride peak in winter because the wind speed is higher during winter. The sea-salt fraction is more influenced by wind speed than by other conditions, as identified by *Chen et al.* [1997]. Calcium and nitrate show high concentrations due to dust storms in spring. The concentration of potassium also is high in spring because potassium is influenced by both soil particles and anthropogenic air pollutants, as will be discussed next.

[14] To further understand the relationship among the ions in TSP, correlation coefficients were estimated and are shown in Table 2. Sodium, magnesium, and chloride are highly correlated to each other because of their common origin (sea salt). Nss-sulfate and ammonium are highly correlated (r = 0.87). This high correlation is also observed for fine particles at Gosan, 0.79 [*Lee et al.*, 2001]. These two species are known to be predominantly in the fine particles at this site [*Kim et al.*, 1999b] and neutralize each other in the fine particle fraction [*Lee et al.*, 2001]. Nss-potassium is better correlated with nss-sulfate (r = 0.68) than with nss-calcium (r = 0.43). It is known that nss-potassium is mainly emitted from either crustal origins or biomass burning. Also, it was found that the nss-potassium measured at Gosan in spring 1999 has a bimodal distribution [*Kim et al.*, 1999b]. So there is the high possibility that nss-potassium originated from anthropogenic sources or/and is transported with anthropogenic species in the region simultaneously rather than from soil particles as suggested by *Kim et al.* [1998a].

[15] The negative value of the correlation coefficient between chloride and nss-sulfate (r = −0.21) suggested that acidic particles caused chloride loss. The chloride loss by sulfate particles probably occurred on the filter during the sampling [*Mukai et al.*, 1990]. Nss-calcium and nitrate are well correlated (r = 0.63). Figure 3 shows the variation of the concentration of nitrate and calcium by wind direction at 850-hPa height between 1992 and 2001. The concentrations

Table 2. Correlation Coefficients Among Ionic Compounds

	NH_4^+	Na^+	Mg^{2+}	NO_3^-	Cl^-	nss-K^+	nss-Ca^{2+}
Na^+	−0.17						
Mg^{2+}	−0.09	0.80					
NO_3^-	0.20	0.34	0.44				
Cl^-	−0.25	0.87	0.73	0.22			
nss-K^+	0.52	0.09	0.22	0.47	−0.05		
nss-Ca^{2+}	0.09	0.26	0.45	0.63	0.26	0.43	
nss-SO_4^{2-}	0.87	−0.02	0.12	0.32	−0.21	0.68	0.34

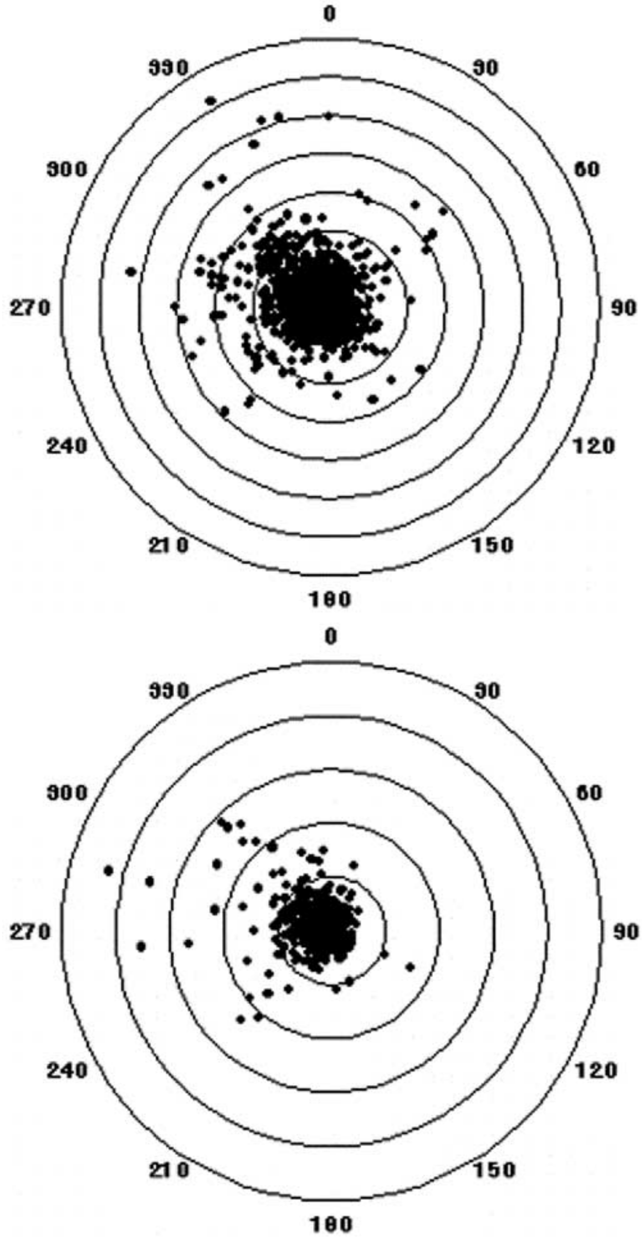

Figure 3. Variations of (top) nitrate and (bottom) calcium concentrations between 1992 and 2001 by wind direction at 850 hPa (concentration scale 2 μg m^{-3}).

of nitrate and calcium are high when the wind came from the west (210°–330°). Therefore it seems that the calcium component of soil reacts with NO_y while transported to Gosan [*Underwood et al.*, 2001], and thus nitrate is mainly in the coarse particle fraction. The same trend has been observed in Beijing, China. *Yao et al.* [2003] measured aerosol size distribution and their ionic concentrations in 2001 and 2002 and found that nitrates are found in both fine and coarse mode particles. Further, when nitrate was in coarse mode, nitrate is well correlated with calcium ions. *Kim et al.* [2004] analyzed both $PM_{2.5}$ and TSP data at Gosan between 1998 and 2002 and found that nitrate in TSP is in good correlation with crustal species such as nss-calcium while nitrate in $PM_{2.5}$ is in good correlation with anthropogenic species such as nss-sulfate or ammonium. Thus nitrate might have two distinct patterns depending on transport characteristics.

[16] Among the ionic species, representative anthropogenic acidic components are nitrate and nss-sulfate. The trend of the annual mean concentrations of nitrate and nss-sulfate was estimated by the linear regression method. The concentration of nitrate is increasing gradually while the concentration of nss-sulfate is slightly decreasing or stable during the period. Linear regression equations are $y = 2.05 \times 10^{-3}x + 0.0159$ (R^2 = 0.658) for nitrate and $y = -2.50 \times 10^{-3}x + 0.154$ (R^2 = 0.370) for nss-sulfate, respectively.

[17] However, the linear regression method might not indicate well the trend of fluctuant cases with seasonal variations and meteorological effects. Thus a nonparametric method called the Mann-Kendall method was applied to the data. It has been widely used in long-term analysis such as water quality management [*Yue et al.*, 2002]. In particular, the seasonal Mann-Kendall test method can account for seasonal variation. Thus the method is well suited to analyze the long-term trend of the annual concentrations. Briefly describing the Mann-Kendall test, first, it judges the existence or nonexistence of a trend. Second, it estimates the magnitude of the trend, if any. Then, the statistical significance is verified by calculating p value using a table of normal distribution function.

[18] Using this method, it is found that the equivalent concentration ratio of nss-sulfate to nitrate (S/N equivalent ratio) is statistically decreasing with a rate of -0.7047 yr^{-1} because the concentration of nitrate is increasing. The concentration of nss-sulfate shows no trend, while the concentration of nitrate is increasing with the rate of 0.00167 μeq m^{-3} yr^{-1}. Thus both methods report the same trend.

[19] The measurement data of S/N equivalent ratio are compared with the ratio values based on the emission data of NO_x and SO_2 in the region, as shown in Figure 4. It was found that the emission ratio values for China are closer to

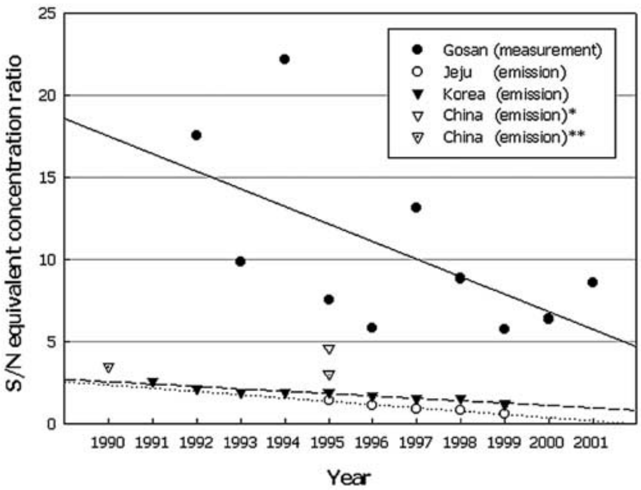

Figure 4. Linear trend analysis of annual average equivalent ratio of nss-sulfate to nitrate. Emission data are the equivalent ratios of sulfur dioxide to the oxides of nitrogen. Korean data are from *Ministry of Environment, Korea* [2001]. One asterisk, from *Lee et al.* [2001]. Two asterisks, from *Streets and Waldhoff* [2000].

Table 3. Result of the Number by Sector and by Cluster

Sector	Cluster 1	2	3	4	5	Total	(Fraction)
I	2	0	153	0	2	157	(0.12)
II	10	25	314	0	309	658	(0.51)
III	3	186	0	0	146	335	(0.26)
IV	0	25	0	48	0	73	(0.06)
V	0	0	0	73	0	73	(0.06)
Total	15	236	467	121	457	1296	
(Fraction)	(0.01)	(0.18)	(0.36)	(0.09)	(0.35)		

the observed ratio values than the emission ratios for either Jeju Island or Korea. Thus it can be suggested that the observed S/N ratios are affected by China rather than Jeju itself or Korea. In other words, there is a possibility that the observed S/N ratio follows the emission trend change in the region, especially in China. Evidently, it is a first estimation, and there are several limitations in comparing emission data to the measurement data directly. It is necessary to consider the conversion rate of gaseous phase to particle phase (SO_2 to sulfate and of NO_x to nitrate), deposition rate, and transport pattern. However, other measurement and modeling studies also support the decreasing trend of S/N ratio in the region, and that trend is mainly caused by the emission trend change in China.

[20] *Takahashi and Fujita* [2000] analyzed rain samples in Japan between 1987 and 1996 and found that the equivalent ratio of N/S is increasing in the rain samples collected in western Japan. They explained this observation by the fact that since the 1980s, SO_2 and NO_x emissions in east Asia have rapidly increased and the increase rate for NO_x emissions in the region was larger than that for SO_2 emissions. *Prospero et al.* [2003] reported that the anthropogenic sulfate and nitrate aerosol concentrations at Midway Island in the central North Pacific almost doubled from 1981 to the mid-1990s but stabilized since the late 1990s. They suggested that this trend is closely related to the emissions from Asia, especially China. *Streets et al.* [2000] reported that the estimated Asian SO_2 emissions (40–45 Tg by the year 2020) are not growing nearly as fast as was thought likely in the early 1990s (2020 emissions as high as 80–110 Tg). *Streets and Waldhoff* [2000] related that growth in NO_x emissions between 1990 and 1995 was larger than for SO_2 in China (including Hong Kong and Taiwan). On the basis of these estimations and observation, it is likely that NO_x emissions in Asia, especially in China, are growing rapidly while SO_2 emissions are stabilized or decreasing.

3.2. Relationship Between Ion Composition and Air Parcel Trajectory

[21] Backward trajectory analysis was carried out for the sampling days using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model (available at http://www.arl.noaa.gov/ready/hysplit4.html, NOAA Air Resources Laboratory, Silver Spring, Maryland, United States) to understand the relationship between the observed aerosol composition and air movement. This model is a complete system for computing trajectories to complex dispersion and deposition simulations using puff or particle approaches. Description of the model is given by *Draxler and Hess* [1997].

[22] Two sets of meteorological data were used; the CDC1 archive data were used for the sampling days till May 1997, and FNL archive data were used for the days since June 1997. By comparing the results from two data sets for the same period, it was found that the results are almost identical. The model vertical velocity method that calculates vertical motion by the meteorological model's vertical velocity fields was used to calculate trajectories. Among 1386 sampling days, 1296 cases of backward trajectory analysis results are obtained. Three trajectories starting at 500, 1500, and 3000 m, respectively, were estimated. On the basis of the starting height of 1500 m, the lowest height above which the effects from local meteorological conditions or topography are small, the trajectories are classified into five sectors based on the sectors proposed by *Kim et al.* [1998a] as indicated in Figure 1. Sector I consists of Korea including eastern Russia, sector II consists of northern China including Mongolia, sector III consists of southern China including Taiwan, sector IV consists of the Pacific area, and sector V consists of Japan. An air parcel trajectory is assigned to the sector in which the trajectory stayed the majority of the time.

[23] The number of days for each sector is shown in Table 3. About half of the air masses come from sector II, northern China, and about a quarter come from sector III, southern China. Thus it is evident that the air masses observed at Gosan are mainly from China, especially from northern China.

[24] Figure 5 shows the variation of the mean concentrations of ions by sector. The concentrations of anthropogenic species at all sectors except sector IV are comparable to each other. When air parcels moved from the Pacific, the concentrations of nss-sulfate and ammonium are lower than from other sectors. It is clear that the aerosol concentrations at Gosan when air parcels moved from the Pacific are the regional marine background levels. When air parcels moved from Japan, the concentrations of sulfate and ammonium are comparable to those when air parcels moved from other sectors probably because of the sulfur emissions from Sakurajima volcano, located at Kagoshima Island, southern Kyushu, and the neutralization between sulfate and ammonium. Note that the amount of sulfur emissions from volcanoes in Japan is similar to that from anthropogenic emissions.

[25] It has been reported that the concentrations of sulfate and ammonium were higher when the air parcels moved

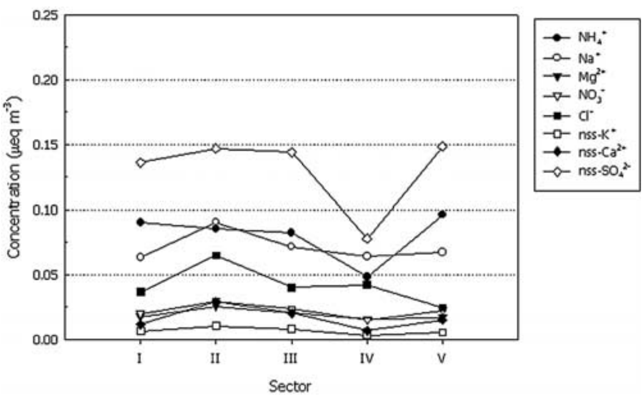

Figure 5. Variation of the ionic mean concentrations by sector.

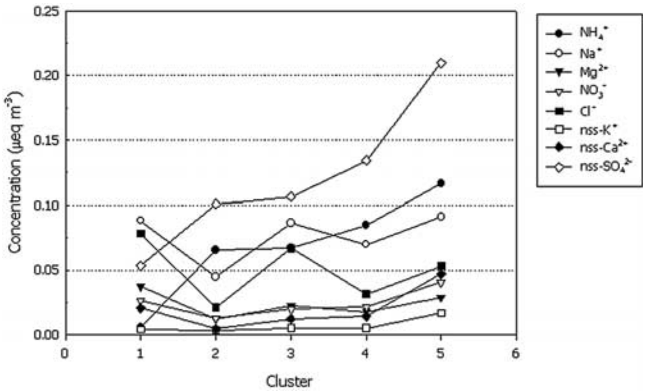

Figure 6. Variation of the ionic mean concentrations by cluster.

from southern China (sector III) than when they moved from northern China at Gosan for $PM_{2.5}$ [*Lee et al.*, 2001]. The concentrations of carbonaceous species were also higher when air parcels moved from southern China than when air parcels moved from northern China [*Kim et al.*, 1999a]. However, the results of this study do not show that trend. One possible reason for that is that there are several cases in which air parcels just passed by Taiwan but not mainland China till they reached Jeju Island. Thus some cases are classified as sector III that mostly pass over the Pacific and the Yellow Sea and thus lowered the average concentrations of anthropogenic species.

[26] When air parcels moved from northern China, the concentrations of sodium and chloride are higher since Gosan is located at the western edge of Jeju and air parcels from northern China passed above the ocean. The concentration of nss-calcium is higher when air parcels moved from northern China, indicating more dust particles when air parcels are from northern China.

[27] To fully understand the relationship between the concentration of ions and air parcel movement, cluster analysis was carried out. The K-means method is used, which is a nonhierarchical clustering method, to discover natural groupings of the items or variables [*Johnson and Wichern*, 1999].

[28] Table 3 also shows the cluster analysis results, and Figure 6 shows the variation of the mean concentrations by clusters. Log-transformed data are used in order that the variables affect the result equally. Since in the cluster analysis, both sector and ionic concentrations are treated equally, the sector number (1, 2, 3, 4, and 5) affects cluster more than the concentration (real concentration). Thus, by normalizing them, this kind of artifact can be eliminated.

[29] Cluster 1 does not have many cases. The cases in cluster 2 are mainly from southern China (sector III), and cluster 3 is exclusively from Korea (sector I) and northern China (sector II). Cluster 4 is from the Pacific (sector IV) and Japan (sector V), and cluster 5 is from both northern China (sector II) and southern China (sector III). Classifying roughly, cluster 2 is from southern China and the Pacific, cluster 3 is from Korea, cluster 5 is from China, and cluster 4 is from the Pacific and Japan. Among all the cases (1296), clusters 3 (36%) and 5 (35.3%) are major clusters.

[30] When air parcels moved from cluster 5 (sectors II and III), the concentrations of ammonium, nss-sulfate, and nitrate are the highest. The high concentrations of anthropogenic species in cluster 4 including sector IV and V are due to the high concentrations of anthropogenic species in sector V. All of the trajectories in sector V were classified into cluster 4. Both sectors II and III are equally distributed into clusters 2 and 5, respectively. To sum it up, when air parcels were from sectors II or III, the concentrations of major anthropogenic species were the highest.

4. Summary

[31] The fate of ambient trace species in northeast Asia is important in both the regional and global environment because of their huge emission amounts. To understand the fate of ambient trace species in the region, it is essential to quantify continental outflow of aerosols. A long-term trend analysis is an essential part of the site characterization. Furthermore, combined with transport pattern, it can provide important information on the transport pattern of ambient trace species in the region. In this work, aerosol composition data measured at Gosan and backward trajectory analysis results between March 1992 and February 2002 are used to study the site characteristics and transport pattern of aerosol in the region.

[32] Daily concentrations of eight major ions in TSP were used in this study. After a three-step quality check, from 1531 daily sample data, 1386 data are used. The average non-sea-salt-(nss)-sulfate concentration (6.74 $\mu g\ m^{-3}$) is higher than those observed at other background areas in the world. From various data analysis results it was concluded that the observed high levels of sulfate are due to transport from outside the site. The concentrations of both anthropogenic species and soil components show maxima during spring except ammonium. This can be explained by the strong continental outflow associated with cold front passage. The concentration of ammonium is the highest during June and July because of the strong agricultural activities during summer in the region. However, during August, because of higher precipitation and the prevailing wind from the southeast, the concentrations of anthropogenic and soil species show minimum levels.

[33] Among the ionic species, nss-sulfate, ammonium, and nss-potassium show a close relationship. Also sodium, chloride, and magnesium and nss-calcium and nitrate show strong correlations, suggesting common emission sources (sea salt for sodium, chloride, and magnesium, for example) or transport pattern. Interestingly, nss-calcium and nitrate show high concentrations when the wind blows from the west. It is likely that nss-calcium in soil reacts with NO_y during transport.

[34] The long-term trend of the concentration ratio of nss-sulfate to nitrate based on the results from a couple of statistical methods is decreasing at Gosan because of the increase of the nitrate concentrations. On the basis of the results of other measurement and emission estimate studies, it can be suggested that this trend reflects the emission pattern change in the region, especially in China.

[35] Backward trajectory analysis was carried out using HYSPLIT4 with the NOAA meteorological data. About half of the trajectories came from northern China, and a quarter came from southern China. On the basis of a cluster

analysis, it was shown that when air parcels were from China, the concentrations of anthropogenic and soil species were higher.

[36] The results given in this study can be used as a basis for further characterization of the transport pattern in the region based on the data obtained during ACE-Asia.

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