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Major factors affecting the ambient particulate nitrate level at Gosan, Korea

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Abstract

Major factors that determine the ambient particulate nitrate concentration at Gosan, Jeju Island, Korea are identified based on the inorganic ionic and elemental data in TSP and $PM_{2.5}$ measured between March 1998 and February 2003. It was found that the nitrate in TSP was highly correlated with crustal species such as nss-Ca^{2+} and nss-Mg^{2+}, while nitrate in $PM_{2.5}$ was highly correlated with anthropogenic species such as NH_4^+. In the high nitrate cases in TSP, the concentrations of crustal species and NH_4^+ were high and the air parcels moved mostly from Mongolia. In the high nitrate cases in $PM_{2.5}$, the concentration of NH_4^+ was high and the air parcels moves mostly from southern China. It was found that the air parcels were mainly from the Pacific Ocean for the low nitrate cases. Still there was significant number of the low nitrate cases with air parcels from Mongolia for both TSP and $PM_{2.5}$. In these cases, the concentrations of crustal species were low for TSP and those of NH_4^+ were low for $PM_{2.5}$. Therefore, the most important factors that determined the level of nitrate were the concentration of crustal species for TSP and the concentration of NH_4^+ for $PM_{2.5}$ irrespective of air parcel movement.

Keywords: TSP; $PM_{2.5}$; Nitrate concentration; Correlation; Factor analysis; Backward trajectory analysis

1. Introduction

Northeast Asia including Korea, China, and Japan is characterized by high emissions of anthropogenic air pollutants. Among them, China emits an overwhelming fraction (van Aardenne et al., 1999; Streets et al., 2000). The emissions of the key air pollutants such as SO_2 and NO_x from China have been expected to increase because of the continuous industrialization (Foell et al., 1995; Streets et al., 2000). Especially, the emission of NO_x from China is drastically increasing. Streets and Waldhoff (2000) predicted the NO_x emission from China will increase by 122% from 1995 to 2020. Recently, He et al. (2007) found a continuous increase of NO_2 concentration during the past decade with a sharp linear increase rate of 14.1–20.5%/year after the year 2000 in satellite observation data, and 10.8%/year between 2000 and 2003 from the modeling study.

NO_x is transformed to nitric acid and nitrate by physical and chemical transformations during transportation in the air. Since Korea is located on the downwind area of China, it has been suggested that the ambient nitrate concentration and nitrate deposition increase due to the long-range transport from China. For example,

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Park and Lee (2003) estimated that about half of nitrogen deposition in Korea between 1994 and 1998 was originated from outside. Among them, nitrate aerosols accounted for 80%. The ambient nitrate level is intricately related to the ambient O_3 and fine particle concentrations. Therefore, it is important to identify the major factors that determine the ambient nitrate level in this region.

Jeju Island is located at about 100 km south of Korean Peninsula, about 500 km west of China (Jiangsu province), and about 200 km east of Japanese Islands (Kyushu) as shown in Fig. 1, and it is one of the cleanest areas in Korea with low emissions of air pollutants (Kim et al., 1998). Therefore, it is an excellent location to study the transport and transformation of ambient trace species in northeast Asia. Gosan is located on the western tip of Jeju Island (126°10′E, 33°17′N), the least developed area in the island on the grounds of a meteorological station. Several intensive and routine measurements studies including Pacific Exploratory Mission-West A and B (PEM-West A and B), Aerosol Characterization Experiment-Asia (ACE-Asia), and Atmospheric Brown Clouds-East Asian Regional Experiment 2005 (ABC-EARX 2005) have been carried out at Gosan (Arimoto et al., 1996; Bae et al., 2007; Chen et al., 1997; Huebert et al., 2003; Park et al., 2004).

In the previous studies of Kim et al. (2003, 2004), the general characteristics of the nitrate concentration at Gosan were studied for the period of 1998 and 2002. According to their results, the nitrate concentration was high in spring and winter, and low in summer. The main reasons for the low concentrations in summer were frequent rainfalls and major air parcel trajectories being from the clean North Pacific. Also, they reported that the high nitrate concentration cases mainly occurred when the air parcels moved from Mongolia and China, and the low nitrate concentration cases occurred mostly when the air parcels moved through the Pacific Ocean. Another point was most meteorological parameters such as ambient temperature, relative humidity (RH), and wind speed did not show statistically significant relationship with the nitrate concentration (Kim, 2004). However, these results were not concrete enough to identify the major factors affecting nitrate concentration levels in this region. Also, the number of data for those studies were small (less than 235) to obtain quantitative result.

The main goal of this work is to identify the major factors that affect the nitrate concentration observed at Gosan based on a long-term data. The nitrate and other inorganic ions, and element data in TSP and $PM_{2.5}$ measured at Gosan between March 1998 and February 2003 were analyzed. First, the relationship between the nitrate concentrations and those of other ions were studied. Second, the highest 10% of the nitrate concentration cases were further analyzed to find relationship with other parameters such as RH, air parcel trajectories, and the Al and Ca levels. Finally, important parameters for the highest and lowest 10% of the nitrate concentration cases were discussed.

2. Measurement data

At Gosan site, a trailer containing the TSP sampler is situated about 10 m inside of cliff, which is about 70 m

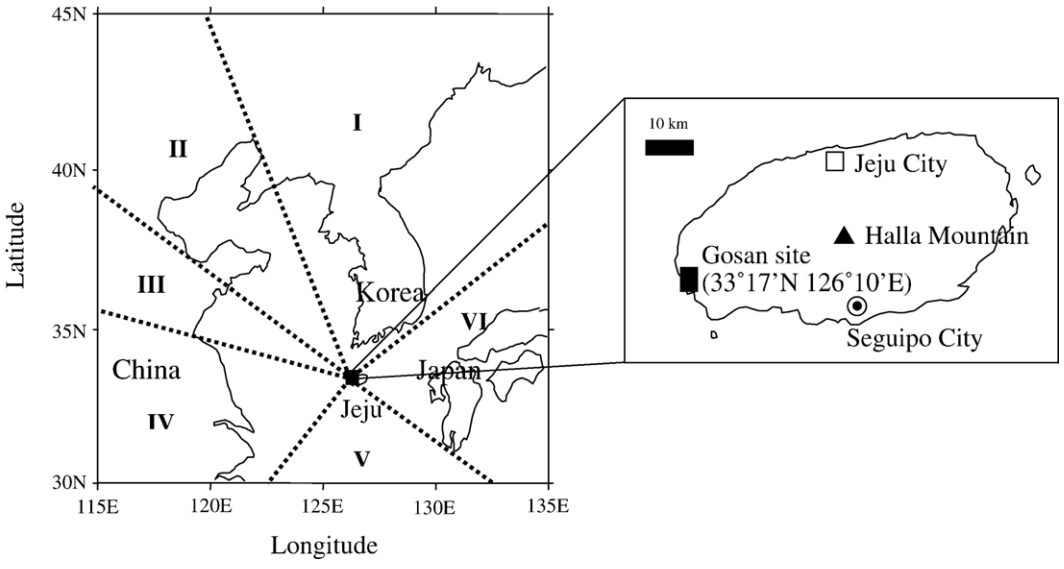

Fig. 1. Map of the measurement site, Gosan, Jeju Island, Korea and the sectors used in backward trajectory analysis.

above sea level. Sampling inlet is located about 6 m above the ground. 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 continuously. TSP 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 h 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. $PM_{2.5}$ particles were collected by a low volume sampler. The sampler consists of a Teflon-coated aluminum cyclone with a cut size 2.5 μm at a flow rate of 16.71 LPM (URG, USA), a Teflon filter holder for 47 mm filters (Sarvillex, USA), a critical orifice (BGI, USA) and a pump (Dayton, USA).

The measurements are consisted of routine and intensive measurements. During routine measurements, TSP samples were taken every 3rd days, and $PM_{2.5}$ samples were taken every 6th days from March 1998 to February 2003. Also, 14 intensive measurements were carried out from March 1998 to February 2003, and during intensive measurements, TSP and $PM_{2.5}$ samples were taken everyday for 1–2 week. Daily sampling started at 0900 LST and lasted for 24 h. All the data presented in this work is the 24 hour averaged results. Details about measurement date and number of data are shown in Table 1.

Table 1
Measurement statistics and the number of samples after QA/QC

Routine measurement				Intensive measurement			
Year	Duration	No. of data		Year	Duration	No. of data	
		TSP	$PM_{2.5}$			TSP	$PM_{2.5}$
1998	3.2–12.27	69	34	1998	3.2–3.31	30	–
					4.1–4.27	20	6
					11.5–11.15	9	9
1999	1.2–12.28	96	48	1999	4.6–4.15	11	10
					6.18–6.25	6	7
2000	1.3–12.28	75	36	2000	3.10–3.15	6	6
					6.9–6.19	11	11
					11.10–11.19	10	10
2001	1.3–12.29	84	30	2001	4.9–4.21	11	11
					8.5–8.14	7	3
					11.5–11.17	11	4
2002	1.4–12.30	93	41	2002	3.2–3.14	10	8
					4.1–4.30	21	16
					12.10–12.19	10	10
2003	1.5–2.28	20	7	2003	–	–	–
Total No. of data		437	196	Total No. of data		173	111
Total No. of TSP data				610			
Total No. of $PM_{2.5}$ data				307			

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+} by an 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 were estimated by assuming all Na^+ are from sea-salt and subtracting the sea-salt composition. Details on the ion analysis were given in Park et al. (2004).

To ensure the quality of the data, three steps of quality control procedures were taken. First, instrument quality check had been 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 with the ratio of the sum of the equivalent cation concentrations to the equivalent anion concentrations being 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 (Park et al., 2004). Organic acids may be a non-negligible part of anions during summer when biogenic emissions of organic compounds are high and carbonates during spring when dust storms are frequent. But these were not analyzed in this work. In case of TSP, among the total data sets of 636, 26 data sets (about 4.1%) were discarded, and in case of $PM_{2.5}$, among the total data sets of 375, 68 data sets (about 18.1%) were discarded based on these three quality checks. In this work, remaining 610 data sets of TSP and 307 data sets of $PM_{2.5}$ are used for further analysis.

Twenty elements, including Al and Ca, were analyzed by Inductively Coupled Plasma (ICP) Spectrophotometer (JOBIN YVON Emission Instruments, Model JY 38S) and polychromator detector. Details on the elemental analysis were given in Park et al. (2003).

3. Results and discussion

3.1. General characteristics

To understand the relationship among the ions in TSP and $PM_{2.5}$, statistical analysis was performed. The Statistical Program for Social Science (SPSS) was used. First, the Pearson correlation coefficients were estimated. For both TSP and $PM_{2.5}$, sodium and chloride were highly correlated to each other. Also anthropogenic species, such as NH_4^+ nss-SO_4^{2-}, and nss-K^+ were highly correlated. However, nitrate in TSP was highly correlated with nss-Ca^{2+} (r=0.65) and nss-Mg^{2+} (r=0.41) rather than NH_4^+ (r=0.27), but nitrate in

$PM_{2.5}$ was highly correlated with NH_4^+ ($r=0.65$) rather than nss-Ca^{2+} ($r=0.19$) and nss-Mg^{2+} ($r=0.18$).

Secondly, factor analysis was carried out using a Varimax factor matrix. Three factors with the eigenvalue over 1 were identified for TSP and $PM_{2.5}$, respectively. For both TSP and $PM_{2.5}$, the first factor was the influence from sea-salts (Na^+ and Cl^-) with 50.2% and 44.0%, respectively. The second factor was the influence from the anthropogenic origins (NH_4^+ and nss-SO_4^{2-}) with 27.1% and 26.5%, respectively. The third factor was the influence from crustal species (nss-Ca^{2+} and nss-Mg^{2+}) with 10.3% and 9.3%, respectively. Nitrate showed distinct feature in this analysis. In case of TSP, nitrate belonged to the third factor (crustal) while for $PM_{2.5}$ it was classified to the second factor (anthropogenic).

In the study of Arimoto et al. (1996), the relationship of nitrate and Al, representative of dust, was low in the source region of Asian dust because the source regions of dust and nitrate in Asia were decidedly different. However, the relationship at Oahu, Hawaii Islands was high ($r=0.75$; $p<0.001$). They suggested that this improved correlation at Oahu was due to the reaction between nitrate and mineral particles during transportation. Thus, the relationship between nitrate and the concentrations of Al and Ca, the representative crustal elements were analyzed to further identify correlation. In TSP, the correlation with Al was 0.464 ($p<0.001$), and the correlation with Ca was 0.498 ($p<0.001$). The relationships between nitrate in $PM_{2.5}$ and Al and Ca were low as expected. The values of the correlation coefficients between nitrate and Al and Ca were 0.076 and 0.133, respectively, both of them were statistically insignificant.

3.2. Characteristics of the high nitrate cases

Generally at Gosan, nitrate showed better relationship with crustal species in TSP while showed better relationship with anthropogenic species in $PM_{2.5}$. To verify whether this behavior was also observed for high nitrate samples, the highest 10% of nitrate concentrations cases for both TSP and $PM_{2.5}$ were analyzed. The numbers of the data were 60 for TSP and 28 for $PM_{2.5}$, respectively. The mean of the high nitrate concentrations were 0.10 and 0.073 μeq m^{-3} for TSP and $PM_{2.5}$, respectively.

3.2.1. Relation to other ions

To understand the relationship between the high nitrate concentrations and other ions in TSP and $PM_{2.5}$, the ions which increased along with nitrate were identified. Here, when the ion concentration of the high nitrate case sample was higher than its mean plus one standard deviation, the ion was defined as increasing with nitrate.

As shown in Fig. 2, for the high nitrate cases in TSP, both nss-Ca^{2+} and nss-Mg^{2+} (26 days, 43%of the total samples), and NH_4^+ (17 days, 28%) increased along with nitrate. In $PM_{2.5}$, NH_4^+ increased along with nitrate (16 days, 57%).

The correlation analysis was performed again with the 60 and 28 high nitrate cases for TSP and $PM_{2.5}$ to compare it with the whole sample results. In case of TSP, the correlations between nitrate and nss-Ca^{2+} and nss-Mg^{2+} were similar to the results for the whole sample, but the correlation with NH_4^+, Na^+ and Cl^- became lower. It is not clear why the correlation between nitrate and NH_4^+ became even lower than the whole sample result though NH_4^+ increased simultaneously for 28% of the high nitrate cases. In case of $PM_{2.5}$, the correlation between nitrate and NH_4^+ became even higher ($r=0.80$) than the results for the whole sample. However, the correlation between nitrate and nss-SO_4^{2-} became lower, and nitrate and nss-Ca^{2+} and nss-Mg^{2+} did not show any correlations at all.

Several measurement studies suggested that nitrate in coarse particles was often associated with crustal species but also associated as well as sea-salts (Lee et al., in press; Ro et al., 2001; Whang and Ro, 2006; Zhuang et al., 1999). However, in our study, nitrate in TSP was associated mainly with crustal species (43% of total high nitrate cases) and rarely associated with sea-salts (7% of total high nitrate cases), even Gosan is located on the coastal area (the western tip of Jeju Island).

3.2.2. Relation with air parcel movement

Backward trajectory analysis was carried out for the high nitrate cases to understand the relationship between the observed aerosol composition and air parcel source and transport. The HYSPLIT4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Rolph, 2003) was used. The FNL (FiNaL run) archive data (NOAA, 2002) were used as the meteorological data and the model vertical velocity method was used to calculate trajectories. Starting time was 00 UTC considering the sampling starting time, and the running time was 96 h. Based on the starting height 1500 m, the trajectories are classified into six sectors as shown in Fig. 1. Sector I consists of Korea including eastern Russia, sector II Mongolia, sector III northern China, sector IV southern China, sector V the Pacific ocean, and the sector VI Japan. An air parcel trajectory is assigned to a sector in which the trajectory stayed the most time.

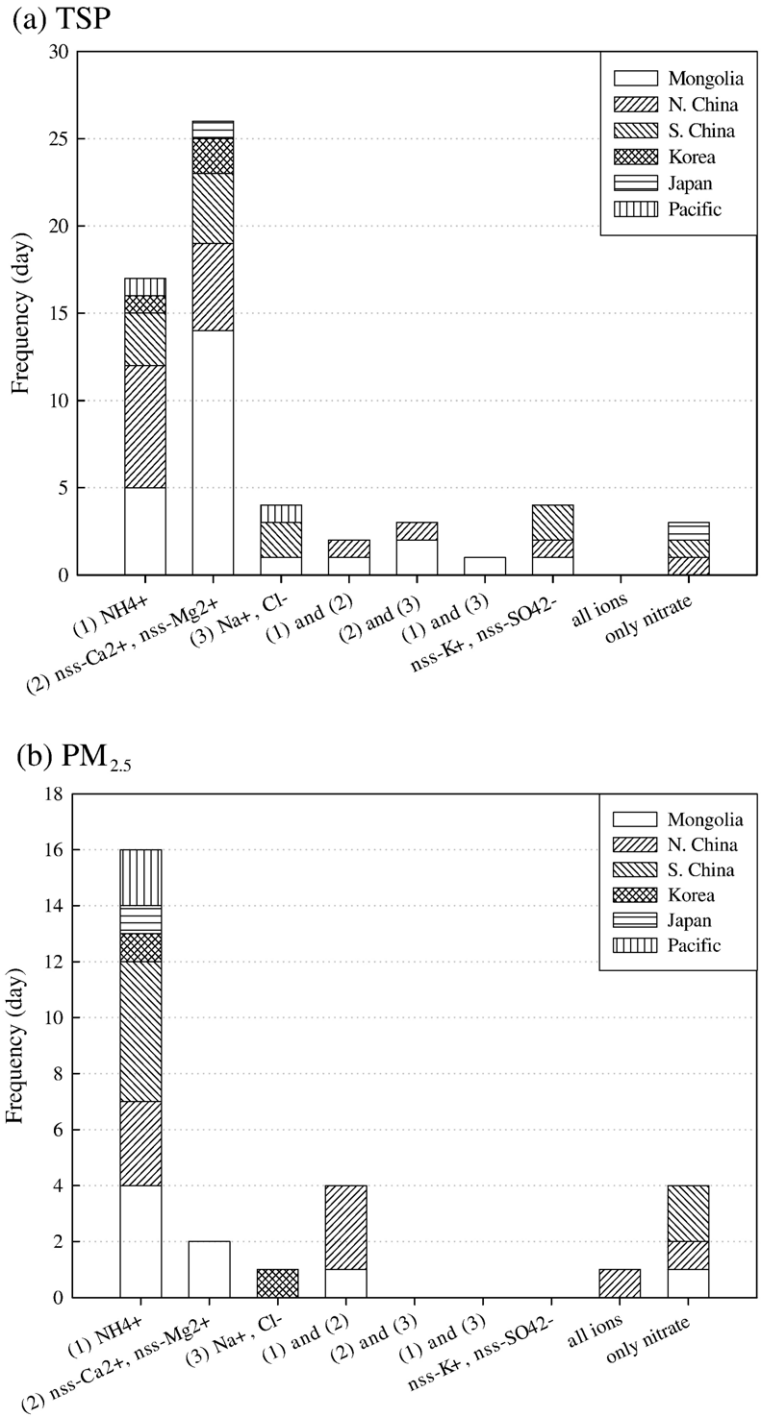

Fig. 2. Number of days of the high nitrate concentration cases with simultaneous increase of other major ions which were also divided into the sectors of air parcel trajectories.

For both TSP and $PM_{2.5}$, when the nitrate concentrations were high with high nss-Ca^{2+} and nss-Mg^{2+} concentrations, the air parcels mainly moved from Mongolia (14 days out of total 26 days, and for 2 days out of total 2 days for TSP and $PM_{2.5}$, respectively). For the high nitrate concentration days with high NH_4^+ concentration, the air parcels mostly moved through northern or southern China (for 10 day s out of total

17 days for TSP, for 8 days out of total 16 days for $PM_{2.5}$) (Fig. 2).

When the air parcels moved from Mongolia, they tended to contain higher crustal species concentrations, because it passes over the arid desert area. According to the studies of Iwasaka et al. (2003a,b) and Trochkine et al. (2003), the aerosols collected over desert areas near Dunhuang were comprised mainly by mineral dust particles, and Si and Ca rich particles were the major. Therefore, when the air parcels moved from Mongolia, the air tended to contain higher crustal species, and, thus, higher chance to react with nitric acid to form nitrate in TSP.

On the other hand, when air parcels moved through China, they tended to contain higher anthropogenic species than crustal species because it passes over the highly industrialized area. Especially, the Huang river region, the most populated and economically and agriculturally active region in China, emits large amounts of NO_x and NH_3 due to strong industrial and agricultural activities (Song and Carmichael, 2001). Therefore, when the air parcels came from China, the air tended to contain high NH_4^+ and, thus, higher chance to form fine ammonium nitrate particles.

3.2.3. Relation with meteorological parameters

According to Krueger et al. (2003), $CaCO_3$ particles can react with HNO_3 to produce $Ca(NO_3)_2$ species in the presence of small amount of moisture on the surface of $CaCO_3$ particles. And the reaction product, $Ca(NO_3)_2$, which is more hygroscopic than $CaCO_3$, absorbs moisture on the surface extensively, and the reaction proceeds until $CaCO_3$ species is consumed. Whang and Ro (2006) also found that reacted $CaCO_3$ particles seem to contain moisture when they were collected.

Therefore, to find out the effect of relative humidity on the high nitrate cases with high nss-Ca^{2+} and nss-Mg^{2+} concentrations in this region, the relationships between high nitrate cases with high nss-Ca^{2+} and nss-Mg^{2+} and relative humidity were analyzed. The relative humidity data were obtained from FNL data which were used to calculate backward trajectories of air parcels. The correlations were 0.306 ($p=0.121$) and 0.500 ($p=0.667$) for TSP and $PM_{2.5}$, respectively. However, for both TSP and $PM_{2.5}$, the values were not statistically significant in the 95% confidence area. Therefore, the effect of relative humidity on high nitrate cases with high nss-Ca^{2+} and nss-Mg^{2+} concentrations was not definitive.

Also, as pointed out in Section 1, it was found that the meteorological parameters did not show close relationship with the nitrate concentration in both TSP and $PM_{2.5}$.

3.3. The factors affecting the high and low nitrate concentrations

To further identify the factors that determine the high and low nitrate concentration cases in TSP and $PM_{2.5}$, the lowest 10% of the nitrate concentration cases were compared to the highest 10% cases. The mean of the nitrate concentrations were 0.007 and 0.002 $\mu eq\ m^{-3}$ for TSP and $PM_{2.5}$, respectively.

The backward trajectory analysis was also performed for the lowest 10% of the nitrate concentration cases. As shown in Fig. 3, the lower nitrate cases occurred mostly when the air parcels moved through the Pacific Ocean for both TSP and $PM_{2.5}$ (55% for the low nitrate cases in TSP, 39% for the low nitrate cases in $PM_{2.5}$). But the air parcels moved from Mongolia also contributed to the low nitrate concentration cases next after the air parcels from the Pacific Ocean (18% for TSP and 29% for $PM_{2.5}$).

As shown in Fig. 2, among the 60 high nitrate cases for TSP, 25 cases (42%) were from Mongolia. Therefore, to find out the reason of the different nitrate concentrations within the same air parcel trajectories, the ion concentrations of highest 10% and lowest 10% nitrate cases were compared as shown in Fig. 4. In case of TSP, the mean concentrations of sea-salts (Na^+ and Cl^-) and crustal species (nss-Ca^{2+} and nss-Mg^{2+}) turned out to be lower ($p<0.01$) than high nitrate cases, and in case of $PM_{2.5}$ the mean concentrations of anthropogenic species such as NH_4^+, nss-SO_4^{2-} and nss-K^+ turned out to be lower ($p<0.01$) when the nitrate concentration was low.

Also, there were low nitrate concentration cases even though the air parcels moved from southern China with severe stagnation over highly polluted area. The ion concentrations of these cases are shown in Fig. 5. On Dec. 24th 2002, the nitrate concentration in TSP was the second lowest of the year. Most ion concentrations were over the averages, but the nss-Ca^{2+} concentration was very low. On Dec. 30th 2002, the nitrate concentration in $PM_{2.5}$ was the lowest of the year, and in this case, the NH_4^+ concentration was not high enough, on the contrary.

In fine particle, nitrate is generally present as in the form of ammonium nitrate (Song and Carmichael, 2001). Especially, the result from the gas/particle equilibrium modeling in the Huang river region in China, NH_4^+ and NO_3^- distributions were closely coupled, and both species accumulated in the fine mode (Song and Carmichael, 2001). On the other hand, the formation of coarse particle nitrate resulting from reaction of nitric acid or its precursors with sea-salt or soil dust has been observed in several studies (Lee et al., in press; Mamane and Gottlieb, 1992; Ro et al., 2001; Underwood et al., 2001; Whang and Ro, 2006; Zhang and Iwasaka, 1999;

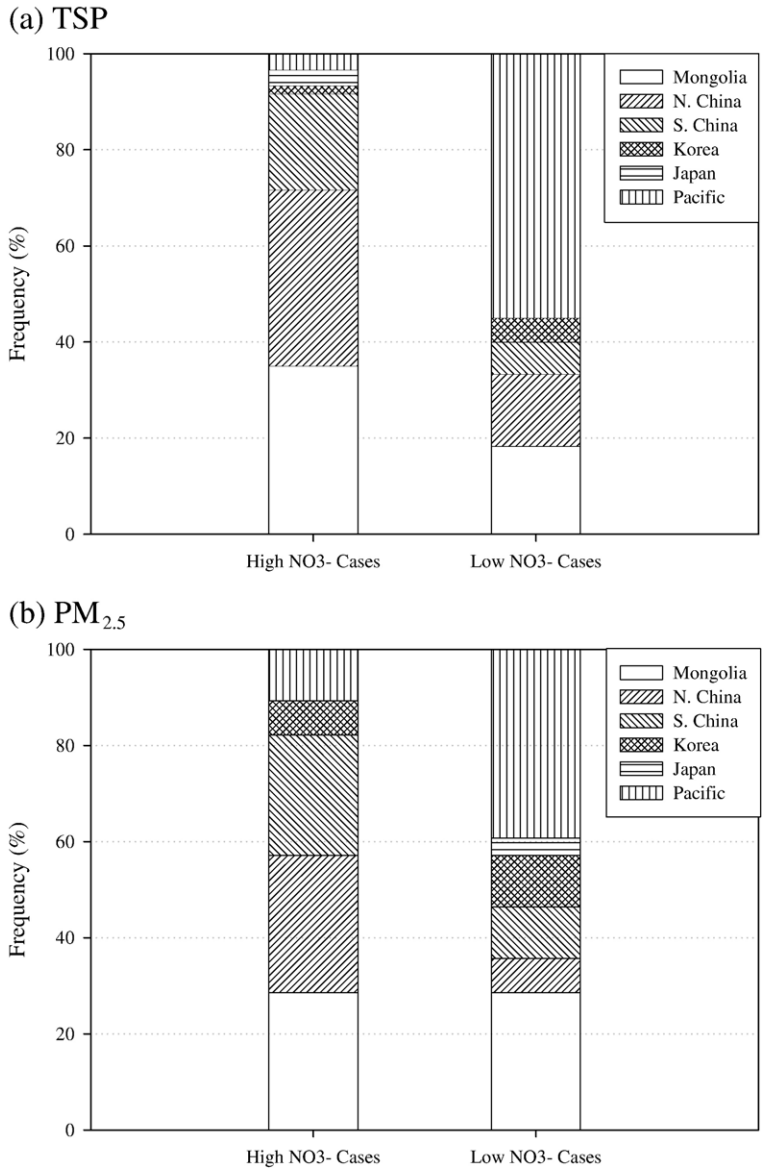

Fig. 3. Contributions of air parcel trajectories in each sector on the high and low nitrate cases.

Zhuang et al., 1999). However, as it is mentioned in 3.2.1, nitrate in TSP was associated mainly with crustal species rather than sea-salts in this study. Therefore, the nitrate in TSP seems to be formed mostly by the reaction between crustal species and nitric acid, and the nitrate in $PM_{2.5}$ seems to be formed mostly by the reaction between NH_4^+ and nitric acid at Gosan area.

4. Summary

In this study, the characteristics of nitrate concentrations and the factors determine nitrate concentrations in TSP and $PM_{2.5}$ were discussed based on the inorganic ions and elemental data in TSP and $PM_{2.5}$ measured at Gosan, Jeju Island, Korea between March 1998 and February 2003.

For both TSP and $PM_{2.5}$, sodium and chloride were highly correlated to each other and anthropogenic species, such as NH_4^+ nss-SO_4^{2-}, and nss-K^+ were also highly correlated among themselves. However, nitrate in TSP was highly correlated with nss-Ca^{2+} and nss-Mg^{2+} while nitrate in $PM_{2.5}$ was highly correlated with NH_4^+. For the high nitrate, similar relationships were found. The meteorological parameters did not show

close relationship with the nitrate concentration in both TSP and $PM_{2.5}$.

Contrary to the several measurement studies which have suggested that nitrate in coarse particles was often associated with crustal species but also associated as well as sea-salts, nitrate in TSP was associated mainly with crustal species (43% of total high nitrate cases) and rarely associated with sea-salts (7% of total high nitrate cases). Considering Gosan is located on the coastal area, it needs further study.

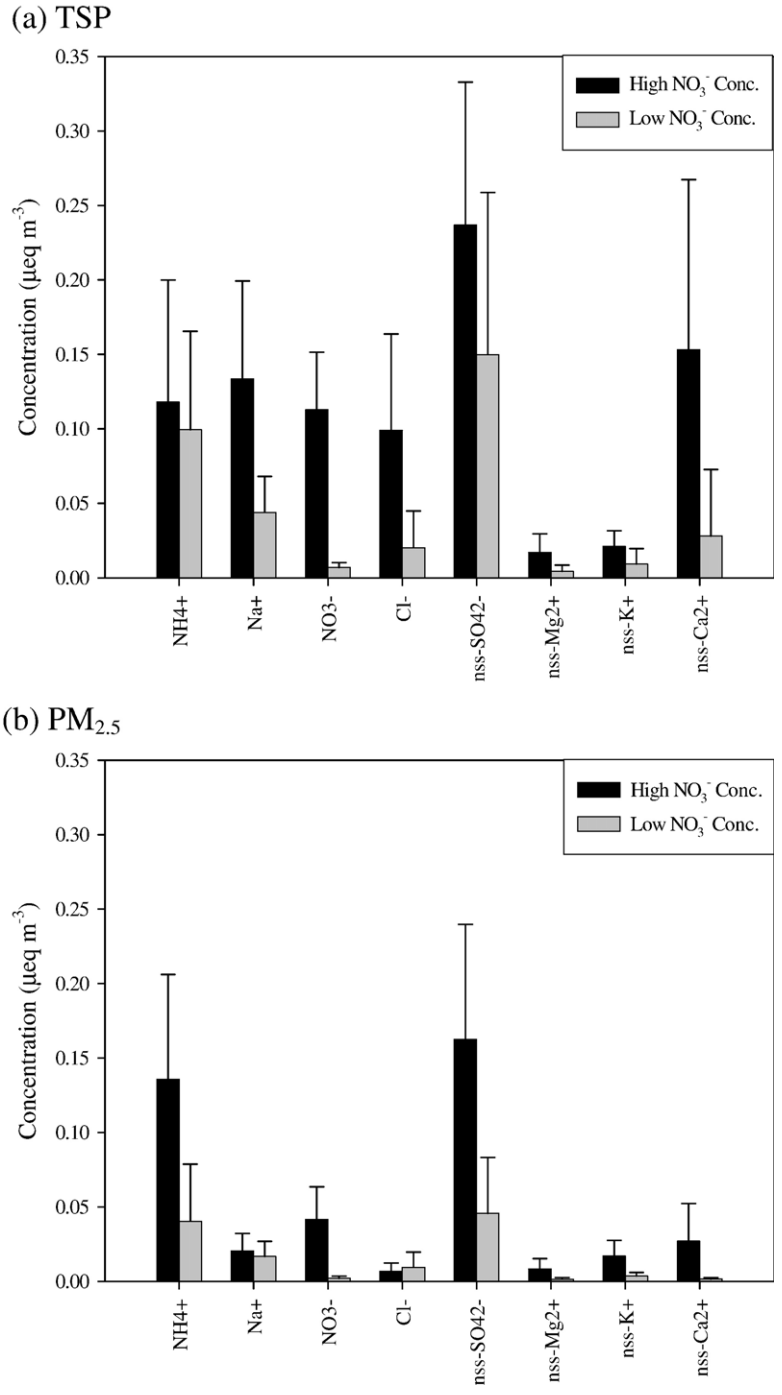

Fig. 4. Comparison of the mean ion concentrations for the high and low nitrate concentration cases when the air parcels moved from Mongolia.

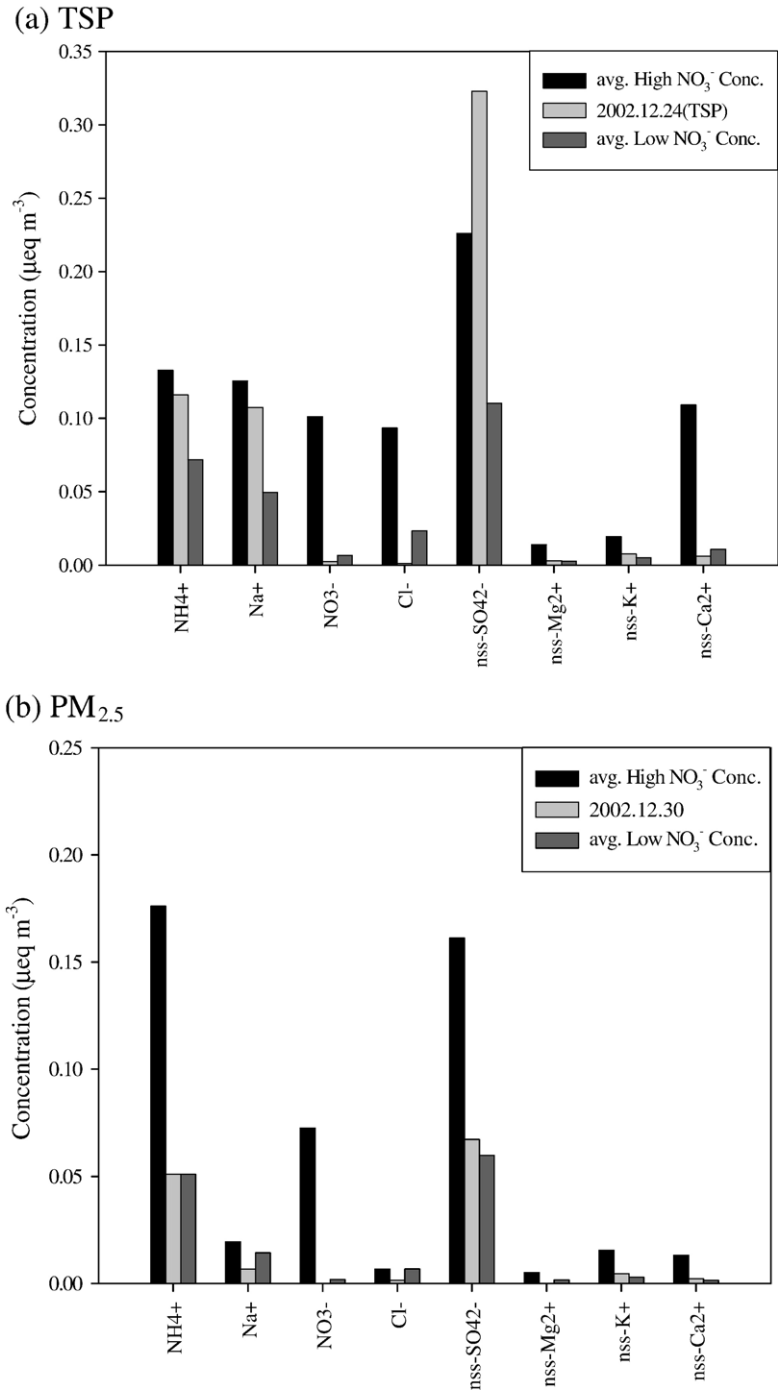

Fig. 5. Comparison of the mean ion concentrations for high and low nitrate concentration cases when the air parcels moved from Southern China.

In the high nitrate cases in TSP, the concentrations of crustal species such as nss-Ca^{2+} and nss-Mg^{2+} were high, and the air parcels moved mostly from Mongolia. In the high nitrate cases in $PM_{2.5}$, the concentrations of anthropogenic species, such as NH_4^+ were high, and the air parcels moves mostly from southern China. However, the nitrate concentrations in TSP were low when the concentration of nss-Ca^{2+} was low even though the air parcel moved from Mongolia. In contrast, the nitrate concentrations in $PM_{2.5}$ were low if NH_4^+ was low even when the air parcel stagnated over southern China.

It has been widely known that reactive nitrogen oxides including nitric acid in the gas phase tend to react with alkaline coarse particles such as sea-salt or dust particles in the atmosphere. Also, it has been well understood that atmospheric ammonia and nitric acid can condense and form fine particles of ammonium nitrate if atmospheric chemical and meteorological conditions are favorable. However, in Northeast Asia, detailed transport and transformation mechanisms have not been clearly understood. Especially, understanding on the case of the interactions between naturally emitted dust particles and anthropogenic chemical species has been limited. That was the main reason behind the intensive studies such as PEM-West A and B and ACE-Asia). Still these were rather short-term intensive studies and a quantitative analysis result based on the long-term measurements presented in this study is important to further understand the transport and transformation characteristics of anthropogenic air pollutants in the region.

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