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Composition and Characteristics of Ionic Components of Aerosols Collected at Gosan Site in Jeju Island, Korea

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Abstract

The total of 1,454 aerosol samples were collected by high volume tape sampler at the Gosan Site in Jeju Island from 1992 to 1999, and the major water-soluble ionic components were chemically analyzed. The mean concentrations of nss-SO_4^{2-}, NH_4^+, and NO_3^- showed high values, which were 6.73, 1.45, and 1.45 $\mu g/m^3$, respectively, while Ca^{2+} and K^+ concentrations were low with the values of 0.49 and 0.42 $\mu g/m^3$. The concentrations of most components increased in spring but decreased in summer, especially with the remarkable increase of Ca^{2+} and NO_3^- concentrations in spring. The seasonal comparison of nss-SO_4^{2-} concentrations showed higher values with the order of spring > fall > winter > summer, but spring > winter > fall > summer for NO_3^-. Meanwhile, the concentration levels of Na^+ and Cl^- increased more in winter season. According to the investigation of wind direction effect, the concentrations of most aerosol ionic components showed higher values consistently at the westerly and northerly wind conditions. Based on the factor analysis, the atmospheric aerosols in the Gosan Site are considered to be largely affected by marine sources, followed by anthropogenic and soil sources.

Key words : Gosan Site, Aerosol, Ionic components, Factor analysis, Wind influence

1. INTRODUCTION

Major sources of atmospheric aerosols include sea salt, crustal dust, gas-to-particle conversion materials such as sulfate and nitrate, and others. The chemical compositions of aerosols are important factors controlling the earth's radiation budget and geochemical cycles of chemical species through wet and dry deposition process (Kido *et al.*, 2001). The atmospheric aerosols affect not only the radiative balance of the earth directly by the scattering or absorbing light, but also

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the albedo and the life-time of clouds indirectly by acting as condensing nuclei, so that they are increasingly responsible to the global weather changes (Huebert and Bates, 1998).

Due to a recent rapid industrial development, the large amount of pollutants has been produced in the East Asia (Arndt *et al.*, 1998). Sulfur emission in East Asia is projected to exceed its total emission of North American and Europe in the year 2010 without future control of emissions (He *et al.*, 2003; Guttikunda *et al.*, 2003). Especially the energy consumption in China has increased rapidly with the growth of economic activities in recent decades. Over the past two decades, China's SO_x emission has grown by more than a factor of 3. This trend is expected to continue with SO_x and NO_x emissions, and projected to gradually increase two to three times between 1990 and 2020. These gaseous as well as aerosol components are transported long distances off the continent (Chen *et al.*, 1997). The westerly wind prevails over the region of East Asia during fall to spring seasons, and aerosol particles and gaseous pollutants originating from the Asian continent are frequently transported to the north and even the central Pacific (Kido *et al.*, 2001). The Korean peninsula is situated downwind from the Asian continent, and is often affected by air pollutants derived from the continent under strong westerly wind conditions.

Recently, a few active studies have been performed to investigate the characteristics of aerosols. Focused on intensive investigation of atmospheric aerosols at the east Asia and northwest Pacific areas, the ACE (Aerosol Characterization Experiment) project has been performing by IGAC (International Global Atmospheric Chemistry) (http://geo.arc. nasa.gov/sgg/ACE-Asia/, http://saga.pmel.noaa.gov/aceasia/). Song and Carmichael (1999) reported that the constituents of atmospheric aerosol particles in East Asia were dominated by continental aerosol from the desert/loess areas, sea-salt aerosol from the ocean, and fossil-fuel combustion derived pollutants (SO_x and NO_x).

The Jeju Island is a vantage place to monitor air pollutants transported in the northeast Asia area. This island is considered as one of the most unpolluted areas in Korea with no large industrial sources and low population density, and located at the middle between China and Japan, separated about 100 km away to the south from the Korean peninsula. Therefore, it is suitable for monitoring the long-range transported air pollutants from the continent of northeast Asia (Sakai *et al.*, 2000; Kim *et al.*, 2000, 1998a, b; Song and Carmichael, 1999; Carmichael *et al.*, 1997; Akimoto and Narita, 1994). The Gosan Site is located at the far western point of the Jeju island, and especially it is the major measurement site for the ACE-Asia project. This study has been focused on the seasonal variation of aerosol compositions and the characteristics of atmospheric aerosols collected at the Gosan Site from March 1992 to December 1999.

2. EXPERIMENTAL

2. 1 Collection of aerosol samples

The Gosan observation station is located on the 72

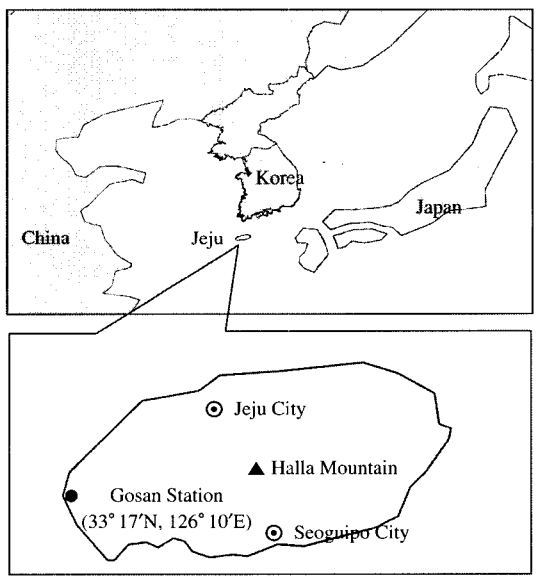

Fig. 1. Location of Gosan Site at Jeju Island, Korea.

meter-high hill, the western edge of Jeju Island (33° 17′N, 126° 10′E) as shown in Fig. 1. The air sampler for aerosol sampling was installed in a trailer. The aerosol samples have been collected using high volume tape sampler (Kimoto Electric Co., Model 195A), which is an automatic sampling system with roll type teflon filters (PTFE filter, 100 mm × 10 m roll tape). The total 1,454 samples were collected from March 1992 to December 1999. Most of them were collected at 24 hour basis, but some at 6 or 12 hour unit.

2. 2 Analysis of aerosol samples

For the extraction of water-soluble ionic components, the PTFE roll filters containing the collected aerosol samples were cut along a circle (75 mm diameter). They were immersed into 0.3 mL of ethanol and 50 mL of ultrapure water, and then sonicated for 30 minutes, and shaken for an hour. The insoluble particles were removed by the membrane filter (0.45 μm), and the filtrates were stored in 4°C refrigerator for the chemical analysis. Na^+, K^+, Ca^{2+}, and Mg^{2+} were analyzed by atomic absorption spectrophotometer (GBC, Avanta-P), but NH_4^+ was analyzed using UV-Visible spectrophotometer (Kontron, UVIKON860) by indophenol method. SO_4^{2-}, NO_3^-, and Cl^- anions were analyzed by ion chromatograph (Dionex, DX-100 and DX-500), and the analytical conditions were as follows; sample volume = 25 μL, eluent = 2.4 mM Na_2CO_3 /2.25 mM $NaHCO_3$, column = IonPac AG4A-SC, IonPac AS4A-SC.

3. RESULTS AND DISCUSSION

3. 1 Analytical results of aerosols

The correlation between the sums of cation and anion equivalent concentrations for the analytical data has been investigated to identify the ion-balance, as shown in Fig. 2. From the results, the correlation coefficient (r) was 0.941, showing relatively good linearity. Also, the imbalance has been investigated to compare the differences between the sums of cation and anion equivalent concentrations, and the % imbalance as well as data quality control parameters are represented in

Table 1. Data quality control parameters and % imbalance.

Regression result		% imbalance	
Number of points	1454	~10% imbalance	747
Slope	0.957	11%~20% imbalance	374
Intercept	0.007	21%~50% imbalance	288
Correlation coefficient (r)	0.941	>50% imbalance	45

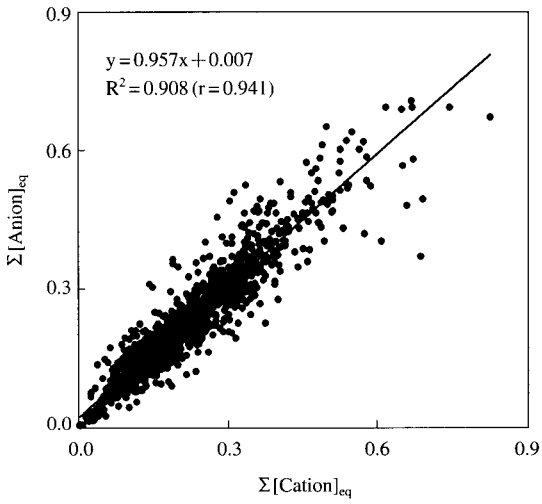

Fig. 2. Scattergram between the sums of cation equivalent concentrations ($\Sigma[Cation]_{eq}$) and anion equivalent concentrations ($\Sigma[Anion]_{eq}$).

Table 2. Comparison of aerosol compositions between the Gosan Site and other domestic and foreign areas.

	Gosan[1]	Kang-wha[3]	Kago-shima[2]	Tsu-shima[3]	Seoul[4]	Nanjing[3]
NH_4^+	1.45	2.03	2.28	1.73	4.93	3.13
Na^+	1.71	1.40	1.99	2.18	1.04	1.60
K^+	0.42	1.50	0.36	0.35	0.22	4.15
Ca^{2+}	0.49	1.08	0.87	0.60	1.80	10.33
Mg^{2+}	0.27	0.34	0.41	0.27	0.18	0.81
nss-SO_4^{2-}	6.73	6.51	8.98	6.91	12.79	20.57
NO_3^-	1.45	3.99	1.65	1.98	4.10	24.85
Cl^-	1.78	2.03	2.19	2.14	4.03	3.23

[1] from March 1992 to December 1999
[2] spring 1993 (Carmichael *et al.*, 1997)
[3] from June 1992 to May 1993 (Carmichael *et al.*, 1997)
[4] February, 1992 (Wakamatsu *et al.*, 1996)

Table 1. It has shown that 96.9% among the total samples was below 50% imbalance and only 3.1% was above 50% imbalance, showing quite good ionic balances (Ayers and Manton, 1991). The concentrations of water-soluble ionic components at the Gosan Site are compared with other domestic and foreign areas in Table 2. From the result, the concentrations of most components at Gosan Site were almost same level as Kangwha, Kagoshima, and Tsushima, which were remote sites in Korea and Japan, but lower level than Seoul and Nanjing, which were typical urban sites in Korea and China.

3. 2 Seasonal and monthly variations of concentrations

The seasonal concentrations of water-soluble ionic components were compared in Table 3. The concentrations of cations were in the order of $NH_4^+ > Na^+ > K^+ > Ca^{2+} > Mg^{2+}$ in summer, fall and winter seasons, but $NH_4^+ > Ca^{2+} > K^+ > Na^+ > Mg^{2+}$ in spring. Among the cations, NH_4^+ had a seasonal average of $1.18 \sim 1.59\ \mu g/m^3$ which was relatively higher than those of other components. It has shown a little bit high concentration values in spring, even though its deviation is not so large compared with the case of other cations. It has been known that the NH_4^+ components are mostly induced from the excrements of animals, manure, and corrosion of plants, especially about 80% of all NH_4^+ from the excrements of animals (Howells, 1995). Besides, the consumed amount of manure as well as the biological activity and temperature have been known to be the most influencing factors for determining the amount of NH_4^+ generation (Pio *et al.*, 1996; Carmichael *et al.*, 1996). It can be considered in this study that NH_4^+ concentrations has been increased in spring, summer and fall seasons due to the factors mentioned above. The mean concentration of Ca^{2+} was $0.67\ \mu g/m^3$ in spring season, which was about $1.3 \sim 4.2$ times larger than those in other seasons.

In Jeju Island, the wind usually flows from the northwest in spring, the southeast in summer, the northeast in fall, and the north in winter (Carmichael *et al.*, 1997, 1996). The fact that Ca^{2+} concentration has been increased largely in spring and winter seasons can be related to the seasonal wind directions. The atmospheric aerosols in Jeju area are considered to be influenced by the mainland China. The Asian Dust storm from China in spring season is cross over into Jeju area by the northwest wind so that the concentration of Ca^{2+}, a main component of soil, certainly increases in the atmospheric aerosols (Park *et al.*, 2001; Murayama *et al.*, 2001; Kotamarthi and Carmichael, 1993; Okada *et al.*, 1990, 1987; Iwasaka *et al.*, 1988).

The concentrations of water soluble anions were in the order of $SO_4^{2-} > NO_3^- > Cl^-$ in spring and summer, but $SO_4^{2-} > Cl^- > NO_3^-$ in fall and winter seasons. Especially, SO_4^{2-} concentration was $6.04 \sim 7.93\ \mu g/m^3$ which was the highest among the other components. In seasonal comparison, it was $7.93\ \mu g/m^3$ in spring and $6.04\ \mu g/m^3$ in summer. It is a general trend that SO_4^{2-} concentration increases highly in winter season by

Table 3. Seasonal comparison of arithmetic mean concentration ($\mu g/m^3$).

Component	Spring		Summer		Fall		Winter	
	Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.
NH_4^+	1.59	1.28	1.42	1.33	1.44	1.02	1.18	0.70
Na^+	1.61	1.30	1.22	1.09	1.81	1.27	2.42	1.70
K^+	0.48	0.43	0.23	0.26	0.45	0.38	0.42	0.34
Ca^{2+}	0.67	0.89	0.16	0.20	0.42	0.41	0.51	0.50
Mg^{2+}	0.27	0.23	0.17	0.16	0.27	0.19	0.34	0.23
nss-SO_4^{2-}	7.52	4.83	5.73	4.32	6.55	3.86	6.14	3.74
NO_3^-	1.83	1.58	0.94	0.92	1.26	1.07	1.33	1.07
Cl^-	1.65	2.28	0.91	1.26	1.66	2.03	3.12	3.16

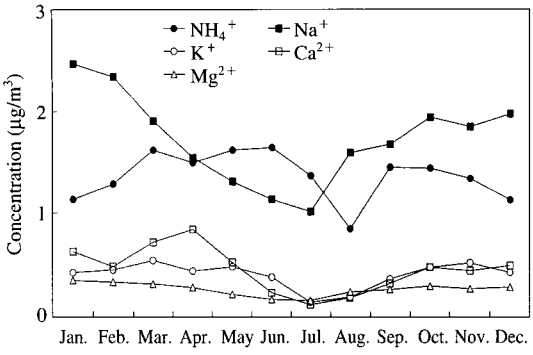

Fig. 3. Monthly variations of cation concentrations of atmospheric aerosols in the Gosan Site.

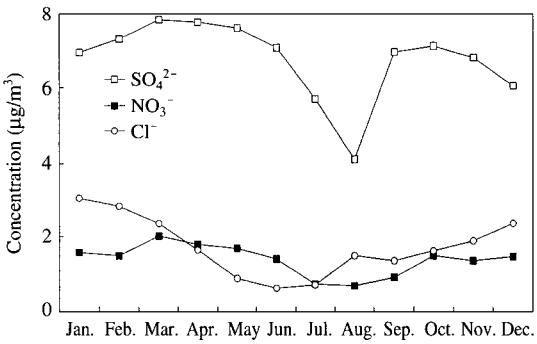

Fig. 4. Monthly variations of anion concentrations of atmospheric aerosols in the Gosan Site.

usage of fossil fuels in polluted urban areas. However, such trend was observed differently in Gosan area, in which the aerosols have contained SO_4^{2-} more in spring than in winter. The explanation can be assumed such that SO_4^{2-} in aerosol in Jeju area might be transferred from other areas, especially some polluted aerosols in spring season could be transferred through a long distance to Jeju area. The NO_3^- concentration has shown the highest in spring season with 1.83 μg/m^3, even higher than that in winter. It can also be explained with the same reason as in the case of SO_4^{2-} (Park *et al.*, 2002; Zhang *et al.*, 2000; Zhang and Iwasaka, 1999).

The monthly variations and comparisons of the concentrations of major ionic components are shown in Fig. 3 and 4. The SO_4^{2-}, NO_3^-, and NH_4^+ originated mostly from anthropogenic sources had high concentrations in March and April. The Ca^{2+} originated mainly from soil source showed high concentrations in April and May with a similar pattern as SO_4^{2-}, NO_3^- and NH_4^+, so that these components might be assumed to have the same emission origin. On the other hand, the Na^+, Cl^-, and Mg^{2+} originated from marine source had the highest concentrations in winter, especially in December and January.

3. 3 Correlation between aerosol components

The correlation coefficients between the water soluble ionic components have been obtained in order to investigate their correlations, and the results are listed in Table 4. The marine components Na^+-Cl^- and Na^+-Mg^{2+} had high correlations, showing the correlation coefficients as 0.876 and 0.758, respectively. Such high correlations between those three components can be reasoned that the aerosols in Jeju area are influenced largely by the ocean, and especially because of the location of the Gosan Site close to a seashore. The components with next high correlations were NH_4^+ and SO_4^{2-}, and its coefficient was 0.841. It has been

Table 4. Correlation coefficients between water-soluble ionic components of atmospheric aerosols.

	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}	SO_4^{2-}	NO_3^-	Cl^-
NH_4^+	1	−0.163	0.459	0.059	−0.088	0.841	0.182	−0.238
Na^+	−0.163	1	0.209	0.298	0.758	0.045	0.294	0.876
K^+	0.459	0.209	1	0.443	0.258	0.645	0.438	0.064
Ca^{2+}	0.059	0.298	0.443	1	0.403	0.327	0.554	0.266
Mg^{2+}	−0.088	0.758	0.258	0.403	1	0.138	0.325	0.689
SO_4^{2-}	0.841	0.045	0.645	0.327	0.138	1	0.334	−0.141
NO_3^-	0.182	0.294	0.438	0.554	0.325	0.334	1	0.166
Cl^-	−0.238	0.876	0.064	0.266	0.689	−0.141	0.166	1

reported that NH_3 reacts quickly with H_2SO_4 in air to produce a salt affecting the albedo of clouds as well as a climate (Zhuang and Huebert, 1996). It was found in this study that the aerosol collected at Gosan area had contained $(NH_4)_2SO_4$ as a major component with the same trends in other areas (Kido *et al.*, 2001; Ma *et al.*, 2001; Ro *et al.*, 2001; Sutton *et al.*, 1998; Querol *et al.*, 1998). The correlation coefficients of K^+-SO_4^{2-} and Ca^{2+}-NO_3^- also showed quite high values, 0.645 and 0.554 respectively. Especially, the relatively high correlation of Ca^{2+}-NO_3^- was somewhat different result from that of other areas, and its behavior appeared mostly in spring season.

3. 4 Concentration variations by wind direction

The concentration variations of major aerosol components by wind directions have been investigated and the results are shown in Figs. 5-7. The wind direction used in this study is at 850 hPa, a pressure at altitude of about 1,500 meter. As in Fig. 5, the concentrations of SO_4^{2-} have high values at the northwesterly wind of around 210° ~ 30°, but low values at the southeasterly wind of around 30° ~ 210°. The concentrations of NO_3^- and K^+ have high values at the northwesterly wind conditions as a similar trend as SO_4^{2-}, as shown in Fig. 6. This implies that those anthropogenic aerosol components, emitted from the continent, may be intro-

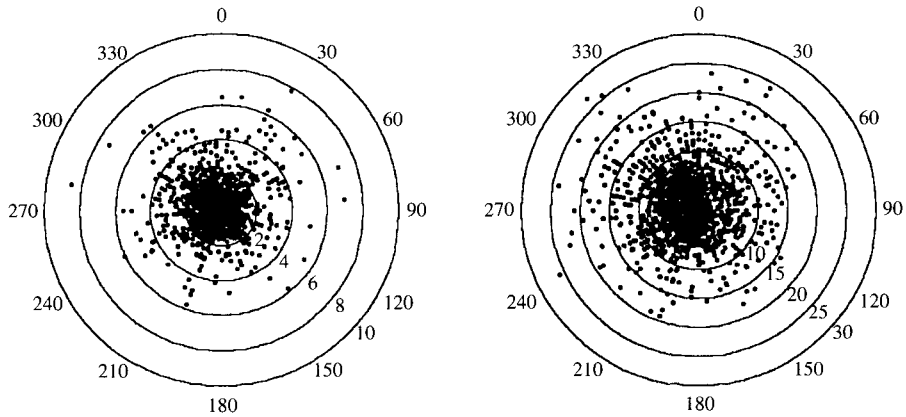

Fig. 5. Variations of NH_4^+ and SO_4^{2-} concentrations (μg/m³) of atmospheric aerosols by wind direction.

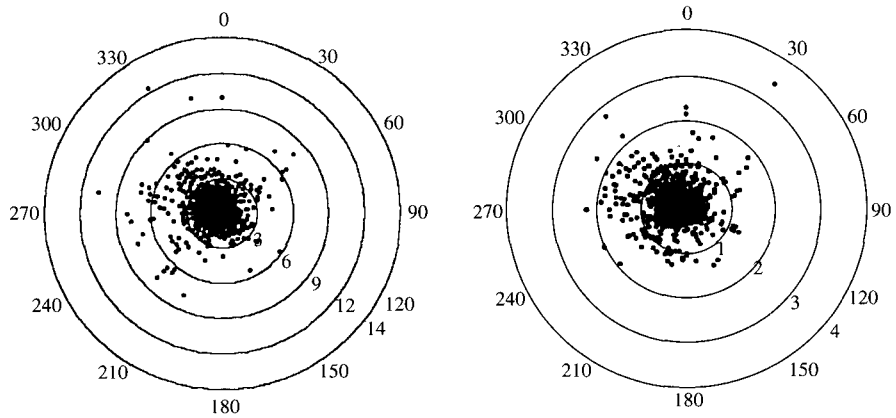

Fig. 6. Variations of NO_3^- and K^+ concentrations (μg/m³) of atmospheric aerosols by wind direction.

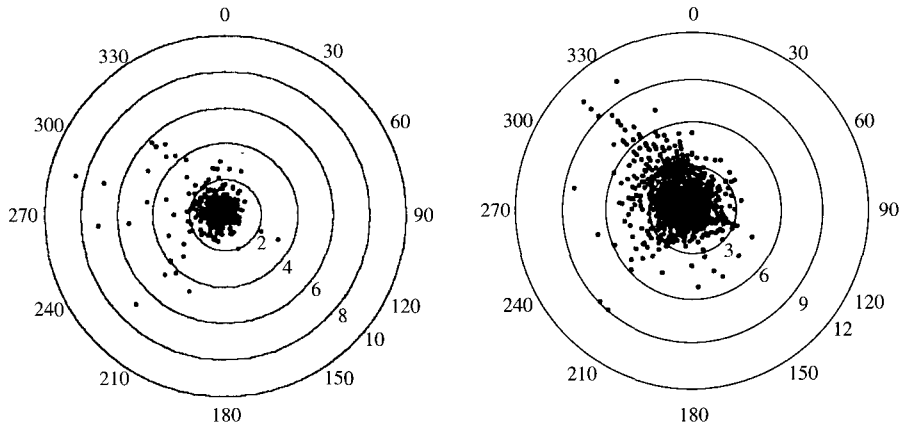

Fig. 7. Variation of Ca^{2+} and Na^{+} concentrations ($\mu g/m^3$) of atmospheric aerosols by wind direction.

duced into Jeju area by long-range transport. The concentrations of $Ca^{2+}$and Na^{+}, as shown in Fig. 6, have also high values at the wind direction of around 210° ~330°, especially Ca^{2+} component shows apparently high increase of its concentration at the westerly wind of around 270°. On the basis of these results, it is assumed that the air pollutants emitted from the continent might be derived into Jeju area, and affect the composition of atmospheric aerosols.

3. 5 Factor analysis

The factor analysis has been performed to investigate the characteristics and origin of aerosols in Gosan area, by a varimax factor matrix using a statistical SPSS program (Lee *et al.*, 2000; Seto *et al.*, 2000; Olsen *et al.*, 1990; Crawley *et al.*, 1986), and the results are listed in Table 5. For this analysis, three factors were chosen to consider an eigenvalue, and these cumulative factor loadings showed 82.6% explanation. The first factor showing 39.5% explanation could be considered as sea-salt effect mostly due to Na^{+}, Cl^{-} and Mg^{2+}, which indicates the Gosan aerosols are mainly influenced by ocean. The second factor with largely NH_4^{+}, nss-SO_4^{2-}, and K^{+} loadings showed 31.6% explanation, and these have typical anthropogenic origin. Therefore the aerosols in Gosan area can be considered to have an influence by anthropogenic sources as well as sea-salts. The third factor having 11.5% variance showed high loadings in Ca^{2+} and NO_3^{-}. Ca^{2+} is one of the main components emitted from soil sources, but NO_3^{-} is a typical anthropogenic component. The fact that these two components having different source origins each other are included within the same factor group indicates that they might be transferred into atmosphere of Jeju with a same pathway through a long-range transport, rather than emitted merely in Jeju area. As a result of this factor analysis and the concentration variation by wind direction previously mentioned, there may exist a strong

Table 5. Results of factor analysis for aerosol components.

Component	Factor 1	Factor 2	Factor 3
NH_4^{+}	−0.137	0.938	−0.031
Na^{+}	0.944	−0.017	0.154
K^{+}	0.162	0.658	0.476
Ca^{2+}	0.205	0.095	0.852
Mg^{2+}	0.840	0.057	0.273
nss-SO_4^{2-}	−0.069	0.933	0.227
NO_3^{-}	0.142	0.179	0.824
Cl^{-}	0.929	−0.157	0.064
Eigenvalue	3.2	2.5	0.9
Variance %	39.5	31.6	11.5
Cumulative %	39.5	71.1	82.6
Source	Sea salt	Anthropogenic	Soil, Anthropogenic

possibility that these components have been flown into the atmospheric aerosols in Jeju area after they had been emitted in the continent of China.

4. CONCLUSIONS

The compositions and characteristics of the aerosols have been investigated in this study, and the results are obtained. From the concentration comparison of aerosol components, the seasonal deviation of NH_4^+ concentrations was small, but Ca^{2+} concentration in spring was 1.3~4.2 times higher than other seasons, probably due to the Asian Dust effect. The SO_4^{2-} and NO_3^- concentrations were relatively high in spring, but low in summer. By the monthly variations, the concentrations of SO_4^{2-}, NO_3^- and NH_4^+ components were high in March and April, Ca^{2+} was high in April and May, and Na^+, Cl^- and Mg^{2+} were high in December and January of mid winter. Na^+/Cl^-, NH_4^+/SO_4^{2-}, Na^+/Mg^{2+}, K^+/SO_4^{2-}, and Ca^{2+}/NO_3^- showed high correlations with the correlation coefficients of 0.554~0.876. The concentration variations by wind directions showed that nss-SO_4^{2-} and NO_3^- had high concentrations at the westerly to northerly wind, but low at the southeasterly wind condition. The concentrations of Ca^{2+}, K^+, and Na^+ were also high at the westerly wind, and especially Ca^{2+} concentration shows apparently high increase. It implies that the air pollutants emitted from the China continent could be overflown into Jeju area by long-range transport. The factor analysis shows that the atmospheric aerosols in Gosan area have been influenced largely by anthropogenic sources as well as sea-salts. The fact that Ca^{2+} and NO_3^- components, having different source origins each other, are loaded in the same factor group explains that they might be transferred into atmosphere of Jeju with a same pathway through a long-range transport.

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