Comparison of Chemical Compositions of Size-segregated Atmospheric Aerosols between Asian Dust and Non-Asian Dust Periods at Background Area of Korea

Won-Hyung Kim, Jung-Min Song, Hee-Jung Ko,[†] Jin Seog Kim,[†] Joung Hae Lee, and Chang-Hee Kang[*]

Department of Chemistry, Jeju National University, Jeju 690-756, Korea. [*]E-mail: changhee@jejunu.ac.kr
[†]Division of Metrology for Quality of Life, Korea Research Institute of Standards and Science, Daejeon 305-340, Korea
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The size-segregated atmospheric aerosols have been collected at 1100 m site of Mt. Halla in Jeju, a background area in Korea, using 8-stage cascade impact air sampler during Asian dust and non-Asian dust storm periods. Their ionic and elemental species were analyzed, in order to examine the pollution characteristics and composition change between Asian dust and non-Asian dust periods. The major ionic species such as nss-SO_4^{2-}, NH_4^+, and K^+ were predominantly distributed in the fine particles (below 2.1 μm diameter), and besides the NO_3^- was distributed more in coarse particle fraction than fine particle. On the other hand, the typical soil and marine species *i.e.*, nss-Ca^{2+}, Na^+, Cl^-, and Mg^{2+}, were mostly existed in the coarse particles (over 2.1 μm diameter). As well in the elemental analysis of aerosols, the major soil-originated Al, Fe, Ca, and others showed prominently high concentrations in the coarse particle fraction, whereas the anthropogenic S and Pb were relatively high in the fine particle fraction. From the comparison of aerosol compositions between Asian dust and non-Asian dust periods, the concentrations of the soil-originated species such as nss-Ca^{2+}, Al, Ca, Fe, Ti, Mn, Ba, Sr have increased as 2.7-4.2 times during the Asian dust periods. Meanwhile the concentrations of nss-SO_4^{2-} and NO_3^- have increased as 1.4 and 2.0 times, and on the contrary NH_4^+ concentrations have a little bit decreased during the Asian dust periods. Especially the concentrations of both soil-originated ionic and elemental species increased noticeably in the coarse particle mode during the dust storm periods.

Key Words : Size-segregated atmospheric aerosol, Chemical composition, Asian dust, Background area, Jeju Island

Introduction

Atmospheric aerosol is a mixture of primary particles, generated from both anthropogenic and natural sources, and secondary particles produced by gas-to-particle conversion processes in the atmosphere. The compositions of the aerosol particles could be widely varied in accordance with the emission type by human activities and industrial sources, as well as inflow pathways. The aerosols originated from different sources generally have different particle size ranges and chemical compositions. The fine particles are usually produced from anthropogenic sources such as combustion processes and by gas-to-particle conversion, on the other hand the coarse particles are mainly emitted from natural processes such as soil dust and sea salt. Therefore the observations on the particle size distribution and chemical composition of aerosols would be valuable for understanding their physical and chemical characteristics, emission sources, and formation mechanism, as well as for establishing control strategies.[1] The coarse size particles, bigger than the size of 2.5 μm, usually contain the imperfect combustion materials of fossil fuels, and the metal elements produced from mining industries, as well as the soil and marine species. On the other hand, the fine size particles below the size of 2.5 μm mostly contain the anthropogenic species emitted from fossil fuel combustions, automobile exhausts, manufacturing chemicals, and the secondary aerosols convert-

ed from gaseous air pollutants. The chemical species in fine particles such as H^+, SO_4^{2-}, NH_4^+, OC (organic carbon), and EC (elemental carbon) are very important for the air quality and human health.[2] Especially the high concentrations of OC and SO_4^{2-} may cause a serious respiratory disease.[3]

The Asian dust storm carries a large amount of fine and dry soil particles to northeastern Asian countries. Asian dust storms originated from the desert regions in China and Mongolia provide a mechanism for the injection of surface materials into the lower and middle troposphere by the prevailing monsoon winds with intense frontal activity.[4,5] This dusts are then transported thousands of kilometers downwind by westerly over the Pacific Ocean, and occasionally reached North America.[6-9] The long-range transport of soil dusts from natural and anthropogenic sources has been the important subject of intense investigation, because it may cause a considerable impact on climate, air-quality and biogeochemical processes on a global scale.[10] Recently the Asian dust storm has become a serious problem due to the desertification expanded in China and the industrial pollutants absorbed in the dust particles. Especially because these particles sometimes include various harmful secondary air pollutants, it is needed to continuously monitor and investigate the physical and chemical characteristics of the atmospheric aerosols.

Korean peninsula is located at the northeastern edge of Asia continent so that it is seriously affected by various air

pollutants as well as Asian dust storms through the prevailing westerly winds.[11] In order to examine the effect of the long-range transport of air pollutants as well as Asian dust storms, it is more desirable to monitor them at a background area to be able to minimize the influences by locally emitted air pollutants. Jeju Island is a good place for studying influences of the trans-boundary air pollutants moved from Asian continent, because it is a clean air area located at more than about 100 km south far from Korean peninsula, 500 km from China and 250 km from Japan.[12-17]

The 1100 m altitude site of Mt. Halla in Jeju Island (*i.e.*, Mt. Halla-1100 Site) is a background area, so that the long-range transport of air pollutants moving through atmospheric boundary layer can be effectively monitored.[18-22]

In this study, the size-segregated atmospheric aerosols have been collected at the Mt. Halla-1100 Site and analyzed for their compositions, in order to examine the chemical change between Asian dust and non-Asian dust storm particles. In addition, the pollution characteristics of atmospheric aerosols flown into Jeju Island through the atmospheric boundary layer have been also investigated.

Experimental

Sampling of Atmospheric Aerosols. Air sampler was installed at the Mt. Halla-1100 site, locating at 1100 m altitude of Mt. Halla (33°21'N, 126°27'E) in Jeju Island. The collection of the size-segregated aerosol samples have been made using 8-stage cascade impact air sampler (Thermo Anderson, model 20-800, USA) with membrane filters (mixed ester of cellulose membrane, 80 mm diameter/1 μm pore size). The particle size of aerosols had been fractionated into 8-stage (9.0 up, 5.8, 4.7, 3.3, 2.1, 1.1, 0.65, and 0.43 μm aerodynamic diameters), using the above cascade impact sampler with the flow rate of 28.3 L/min during the whole study periods. The collection of aerosol samples was carried out between March and October of 2006; 6 times in spring (1st; March 10-12, 2nd; April 1-10, 3rd; April 10-17, 4th; April 18-23, 5th; April 23-26, 6th; April 30-May 2), once in summer (7th; June 6-16), and once in fall (8th; October 16-25). During the sampling periods, the Asian dust events had actually occurred five times; March 11-12, April 8-9, 19-20, 23-25, and May 1. Therefore the aerosol filter samples were divided into two groups, *i.e.*, the Asian samples (1st, 2nd, 4th, 5th, 6th) and the non-Asian samples (3rd, 7th, 8th).

Analysis of Ionic Components. The ionic species have been analyzed simultaneously by ion chromatography, using the extracted solution of water-soluble fractions from the aerosol filters with ultrapure water. For the analysis of NH_4^+, Na^+, K^+, Ca^{2+}, and Mg^{2+} cations, the Metrohm Modula IC with Metrohm Metrosep Cation 1-2-6 column was applied. Its instrumental conditions were as follows: flow rate = 1.0 mL/min, injection volume = 20 μL, eluent = 4.0 mM tartaric acid/1.0 mM pyridine-2,6-dicarboxylic acid. For the analysis of SO_4^{2-}, NO_3^- and Cl^- anions, the same IC system with Metrohm Metrosep A-SUPP-4 column was used under the following instrumental conditions: flow rate = 1.0 mL/min,

injection volume = 100 μL, eluent = 1.8 mM $NaHCO_3$/1.7 mM Na_2CO_3, and suppressor solution = 0.1% H_2SO_4. The instrumental detection limit (IDL, μg/L) of the IC analysis was recorded on the whole as follows; NH_4^+ = 2.4, Na^+ = 1.8, K^+ = 7.8, Ca^{2+} = 4.8, Mg^{2+} = 8.8, SO_4^{2-} = 9.6, NO_3^- = 10.5, and Cl^- = 4.8.

Analysis of Elemental Components. The elemental species were analyzed by the US EPA method, *i.e.*, "Compendium of Methods for the Determination of Inorganic Compounds in Ambient Air".[23] The aerosol filters were digested with the acid solution (5.55% HNO_3/16.75% HCl) by a microwave digestion system (CEM, Model MARS-5, USA). The filter samples were placed in PFA vessels containing 10 mL acid solutions, and then heated up to around 180 °C for 10 minutes in microwave system. The extracted solutions were filtrated with a syringe filter (Whatman, PVDF, 25 mm/0.45 μm) and then diluted to 25 mL volume with ultrapure water. All elemental species were analyzed by ICP-OES system (Thermo Jarrell Ash, Model IRIS-DUO, USA) equipped with an ultrasonic nebulizer (CETAC Tech., Model U-5000AT, USA). The instrumental conditions for the ICP-OES analysis were as follows: RF power = 1,150W/40.68 MHz, Ar flow rate (coolant = 16.0 L/min, auxiliary = 1.5 L/min, carrier = 0.5 L/min), and simultaneous or sequential mode. The instrumental detection limits for each individual element were approximately in the range of 0.3-4.2 μg/L.

Results and Discussion

Concentrations and Size Distribution of Aerosol Components. The average mass concentration of atmospheric aerosols measured eight times during spring, summer and fall seasons was 91.8 μg/m³. The average mass concentrations of the aerosols for the Asian dust periods and non-Asian dust periods were found to be 180.9 and 47.4 μg/m³, respectively. The average concentrations for the ionic and elemental species of size-segregated aerosols are presented in Table 1. In the table, the non-sea salt (nss) concentrations of sulfate and calcium had been obtained by the equations of $[nss\text{-}SO_4^{2-}] = [SO_4^{2-}] - [Na^+] \times 0.251$ and $[nss\text{-}Ca^{2+}] = [Ca^{2+}] - [Na^+] \times 0.04$ respectively, for eliminating the influence of marine sources on those aerosol compositions.[24-26] The classification for the fine and coarse particle modes was conducted due to the base of 2.1 μm aerodynamic diameter instead of 2.5 μm, because of the nominal cut-size limitation on the cascade impact sampler used in this study.

The size distribution of the aerosol components in Table 1 shows that the percentage contribution of nss-SO_4^{2-} in fine particle mode below 2.1 μm is 78%. The ammonium ions are also distributed largely within this fine particle mode, representing 64% of the total concentration. Generally, the SO_4^{2-} species exists in the atmosphere as a secondary pollutant which has been oxidized from gaseous SO_2, and tends to be distributed mostly in a submicron mode with the diameter of below 1 μm at urban or inland regions.[27] The ammonium ion is also a typical secondary air pollutant, originated mainly from anthropogenic sources by gas-to-

Table 1. Concentrations (ng/m^3) and size distribution of size-segregated aerosol components

Species	Particle size (µm)								
	0.4-0.7	0.7-1.1	1.1-2.1	2.1-3.3	3.3-4.7	4.7-5.8	5.8-9.0	9.0-	Total
NH_4^+	145.4	185.0	122.2	55.5	51.5	49.1	51.0	51.8	711.4
$nss\text{-}SO_4^{2-}$	772.5	978.1	540.4	180.4	138.4	72.5	110.6	144.7	2,937.5
NO_3^-	71.1	116.3	181.9	259.0	325.7	141.6	257.8	265.2	1,618.5
K^+	22.0	28.8	34.5	30.0	32.3	34.7	33.4	29.8	245.5
$nss\text{-}Ca^{2+}$	20.9	35.6	66.6	110.2	132.7	87.8	128.6	90.5	673.1
Na^+	70.0	76.4	97.8	134.8	160.5	122.9	144.0	163.1	969.4
Mg^{2+}	5.5	12.1	16.0	22.7	24.6	15.3	21.9	22.1	140.1
Cl^-	5.8	8.2	16.5	36.8	52.6	26.7	43.8	68.3	258.8
Al	62.5	48.3	295.9	529.9	643.8	393.8	646.1	683.9	3,304.2
Fe	19.3	38.4	212.1	395.4	427.4	269.6	513.4	471.0	2,346.6
Ca	9.4	19.3	156.3	324.2	456.5	276.9	532.5	632.4	2,407.5
Na	139.1	146.4	181.9	220.1	245.8	189.0	226.7	252.4	1,601.6
K	51.3	88.9	136.3	163.9	198.0	114.1	191.3	198.4	1,142.2
Mg	8.9	14.1	78.7	142.8	175.8	99.4	181.9	200.9	902.5
Ti	0.9	0.8	7.8	16.6	20.0	13.3	23.1	23.7	106.2
Mn	1.1	2.4	5.7	7.1	8.4	4.8	8.6	9.0	47.0
Ba	0.2	0.5	2.9	4.5	5.5	3.2	5.9	5.1	27.8
Sr	0.2	0.3	1.2	2.0	2.3	1.3	2.3	2.6	12.1
Zn	13.6	20.5	31.0	21.3	25.7	3.5	8.1	10.3	134.1
V	0.5	0.7	3.2	5.2	6.2	3.6	6.2	6.6	32.1
Cr	1.5	1.9	1.6	2.9	2.7	1.7	10.2	10.7	33.4
Pb	4.6	10.5	9.1	6.7	2.5	1.1	1.3	8.9	44.6
Cu	0.8	1.4	1.1	1.5	1.0	0.7	4.3	1.3	12.2
Ni	2.9	2.5	4.0	1.4	2.4	1.9	1.9	4.4	21.3
Co	0.2	0.3	0.8	1.4	1.5	1.1	1.6	1.7	8.6
Mo	0.2	0.1	0.2	0.2	0.2	0.1	0.2	0.2	1.3
Cd	0.2	0.3	0.3	0.3	0.3	0.2	0.2	0.2	1.9

particle conversion processes in the atmosphere. On the other hand, 77% of the NO_3^- species were distributed in coarse particle mode of over 2.1 µm. Usually the fine nitrate ions are originated from some anthropogenic pollution sources, and exist as NH_4NO_3 salt produced from a reaction between HNO_3 and NH_3. However the nitrate ions in coarse particle mode have been known to exist as $NaNO_3$ after a reaction with sea-salt components.[27] Therefore the distribution ratios (F/C) of fine to coarse particle mode for NH_4^+, $nss\text{-}SO_4^{2-}$, and NO_3^- were 1.7, 3.5, and 0.3, respectively. Meanwhile the percentage of soil-originated $nss\text{-}Ca^{2+}$ is 82% of the total concentrations in the particles of over 2.1 µm diameter, showing the predominant distribution in a coarse mode. This result implies that the soil-originated $CaCO_3$ may react with gaseous HNO_3 to produce calcium nitrate salt and exist in a coarse particle mode.

The marine-originated Na^+, Mg^{2+}, and Cl^- were also distributed mostly in coarse particle mode likewise as the $nss\text{-}Ca^{2+}$ species. From the analytical results of the elemental species, the soil-originated Al, Fe, and Ca showed prominently 88%, 89%, and 92% in the coarse particles, respectively. On the other hand, Pb and Zn distributed with almost even proportions in fine and coarse modes. When the nominal cut-size limitation of aerosol particles was set to the

diameter of 4.7 µm diameter, the percentage of Pb and Zn would be 75% and 84% respectively, explaining the existence of anthropogenic Pb and Zn mainly in comparatively fine particles below 4.7 µm diameter.

Based on the investigation for the size distributions of aerosol components, the anthropogenic aerosol species such as $nss\text{-}SO_4^{2-}$, NH_4^+, Pb, and Zn are mainly distributed in the fine particle mode, except in case of NO_3^-. On the other hand, the soil-originated species ($nss\text{-}Ca^{2+}$, Al, Fe, Ca, Ba, Ti, *etc.*) and the marine-originated species (Na^+, Mg^{2+}, Cl^-) are mostly distributed in the coarse particle mode.
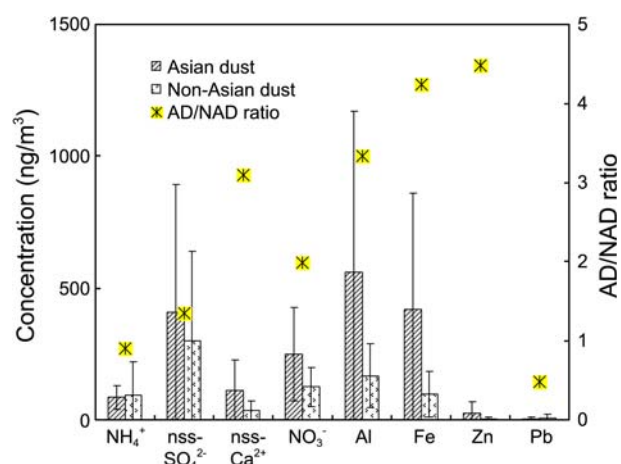
Composition Characteristics of Asian Dust and Non-Asian Dust Particles. The compositions of size-segregated aerosol particles have been compared for Asian dust and non-Asian dust periods in order to examine their chemical characteristics. It can be verified from Table 2 that the water-soluble ionic concentrations are in the order of $nss\text{-}SO_4^{2-} > NO_3^- > Na^+ > nss\text{-}Ca^{2+} > NH_4^+ > Cl^- > K^+ > Mg^{2+}$ for the Asian dust period, and $nss\text{-}SO_4^{2-} > NO_3^- > NH_4^+ > Na^+ > nss\text{-}Ca^{2+} > Mg^{2+} > K^+ > Cl^-$ for the non-Asian dust period. Figure 1 shows the concentrations of some major aerosol species for the Asian dust and non-Asian dust periods with their ratios. The average $nss\text{-}SO_4^{2-}$ concentrations of the total aerosol for the Asian dust period and non-Asian dust

Table 2. Concentrations (ng/m³) of the ionic and elemental species for Asian dust (AD) and non-Asian dust (NAD) periods and their ratios

Species	AD	NAD	AD/NAD
NH_4^+	683.7	757.7	0.9
$nss\text{-}SO_4^{2-}$	3254.5	2409.4	1.4
NO_3^-	1988.1	1002.6	2.0
K^+	328.4	107.4	3.1
$nss\text{-}Ca^{2+}$	901.9	291.6	3.1
Na^+	1366.6	307.5	4.4
Mg^{2+}	150.1	123.5	1.2
Cl^-	377.6	60.6	6.2
Al	4480.8	1343.1	3.3
Fe	3275.2	792.4	4.1
Ca	3287.3	935.0	3.5
Na	2137.0	709.3	3.0
K	1520.6	511.5	3.0
Mg	1248.0	326.6	3.8
Ti	146.8	38.8	3.8
Mn	63.8	19.0	3.4
Ba	37.7	11.1	3.4
Sr	15.8	5.8	2.7
Zn	187.1	39.3	4.8
V	45.8	8.3	5.5
Cr	43.6	14.6	3.0
Pb	32.6	64.2	0.5
Cu	16.6	3.9	4.2
Ni	27.0	10.0	2.7
Co	11.7	3.1	3.8
Mo	1.0	1.6	0.6
Cd	2.2	1.4	1.6

Figure 1. Concentrations for Asian Dust (AD) and Non-Asian Dust (NAD) periods and the concentration ratios of AD/NAD of the major aerosol components.
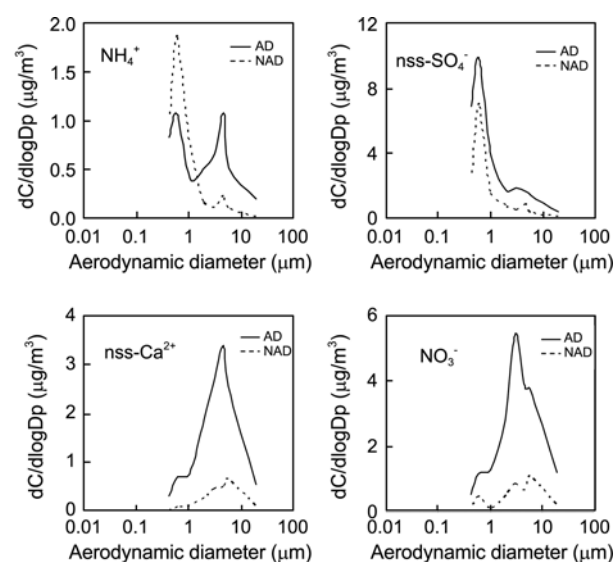
Figure 2. Concentration variations with size distributions of the ionic components for Asian Dust (AD) and Non-Asian Dust (NAD) periods.

period were 3254.5 ng/m³ and 2409.4 ng/m³, respectively, and the concentration ratio (AD/NAD) was 1.4. The NO_3^- concentrations for AD and NAD periods were 1988.1 ng/m³ and 1002.6 ng/m³, respectively, and its AD/NAD ratio was 2.0. Especially, the $nss\text{-}Ca^{2+}$ concentrations during AD and NAD periods were 901.9 ng/m³ and 291.6 ng/m³, respectively, and its AD/NAD ratio was 3.1, suggesting an evidence of the noticeable increase by the Asian dust storms. On the other hand, NH_4^+ concentrations during AD and NAD periods were 683.7 ng/m³ and 757.7 ng/m³, respectively, and its AD/NAD ratio was 0.9, showing a little bit decrease of the concentrations during Asian dust periods.[19,28]

Figure 2 shows the concentration variations of major ionic species by the Asian dust storms. In aerosol sampling by cascade impact sampler, the particle size of aerosols usually is represented as an aerodynamic diameter instead of a real Stokes diameter, and dC/dlogDp indicates the derivative of the concentrations according to the particle size diameter within each stage of the cascade impactor.[29] As shown in Figure 2, it is evident that the soil-originated $nss\text{-}Ca^{2+}$ mostly exist in a coarse mode and the concentrations increase noticeably during the Asian dust periods. Likewise, the concentrations of anthropogenic NH_4^+ and NO_3^- were increased remarkably in coarse particle mode during the Asian dust

periods. On the order hand, the $nss\text{-}SO_4^{2-}$ concentrations increased only in fine particle modes during the Asian dust periods, showing a somewhat different pattern from the above three species.

The concentrations of elemental species in Table 2 have been shown as the order of Al > Ca > Fe > Na > K > Mg > Zn > Ti > Mn > V > Cr > Ba > Pb > Ni > Cu > Sr > Co > Cd > Mo for the Asian dust periods. Meanwhile, the order during the non-Asian dust periods was Al > Ca > Fe > Na > K > Mg > Pb > Zn > Ti > Mn > Cr > Ba > Ni > V > Sr > Cu > Co > Mo > Cd. The concentrations of the main soil-originated Al, Ca, and Fe were 4480.8, 3287.3, and 3275.2 ng/m³ during the Asian dust periods, and 1343.1, 935.0, and 792.4 ng/m³ during the non-Asian dust periods, so that AD/NAD ratios resulted to 3.3, 3.5, and 4.1, respectively, as indicated in Table 2 and Figure 1. It can also be seen in Figure 3 that the concentrations of Al, Fe and Zn are much
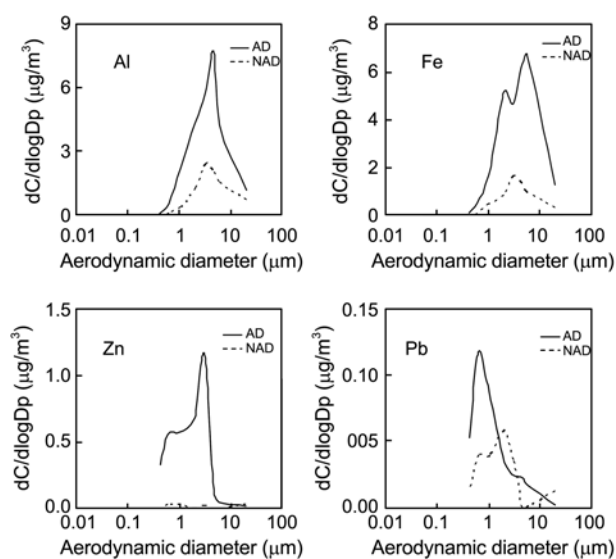
Figure 3. Concentration variations with size distributions of major elemental components for Asian Dust (AD) and Non-Asian Dust (NAD) periods.
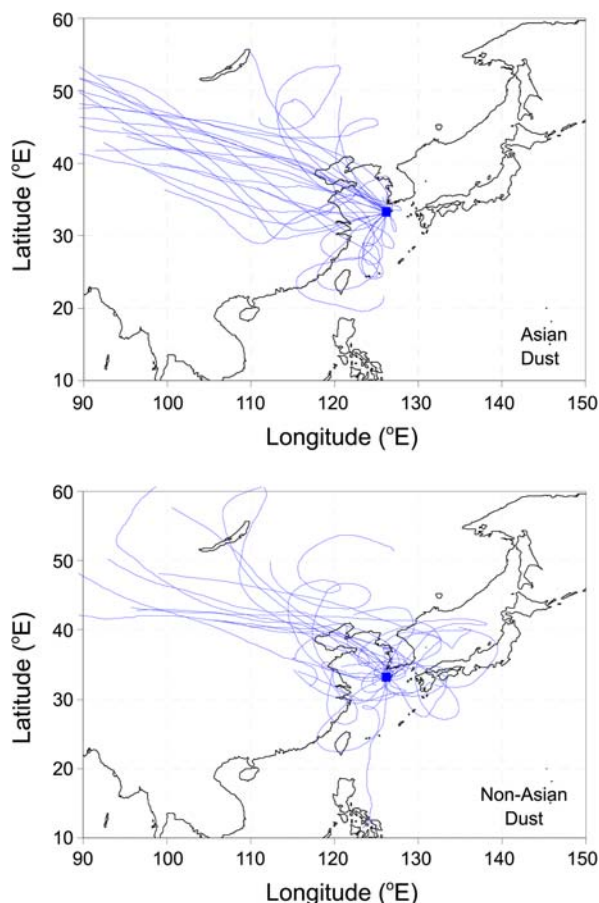

Figure 4. Inflowing pathways of air mass by 5-day backward trajectories during the Asian Dust (upper) and Non-Asian Dust (lower) periods.

high in coarse particle modes for the Asian dust periods. It was found that other soil-originated metal elements were highly increased in coarse particle modes during the Asian

dust periods. The AD/NAD ratios of Zn, Ti, Mn, Ba and Sr were 4.8, 3.8, 3.4, 3.4, and 2.7, respectively, assuring a definite increase during the Asian dust periods, even though their concentration values were not so high. On the other hand, anthropogenic lead was distributed mainly in fine particle mode, of which the concentrations showed relatively a little decrease compared to the above soil species during the Asian dust periods. Similarly, the concentrations of Mo and Cd elements did not show a big difference during Asian dust and non-Asian dust periods.

Inflow Pathway of Air Mass. The pathway of the air mass which had been moved into Jeju Island during the sampling periods has been investigated by the backward trajectory analysis. The backward trajectory was ascertained using the HYSPLIT4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) model of National Oceanic and Atmospheric Administration (NOAA).[30] The information about the wind directions and speeds for the northern hemisphere was provided by FNL (FiNaL run at NCEP). Accordingly, the grid size was 190.5 km × 190.5 km, the 14 vertical layers had been composed from the ground to 850 hPa. The starting height of the trajectories was 1500 m and the total run time was 120 h.[31-33] The backward trajectories showed that the air masses had moved mainly through the central and northeast regions of China and Mongolia into Jeju Island during the Asian dust periods (Figure 4). On the other hand, during the non-Asian dust periods, the air masses had moved by various pathways through the north Pacific and/or the East Sea as well as Siberia and East China.

During the 5th and 6th Asian dust periods, the soil-originated components (Ca^{2+}, K^{+}, Al, Fe, Ca, Ti, Ba, *etc.*) showed prominently high concentrations, and it could be reasoned by the air mass inflow mainly transported from Mongolia and China. On the other hand, the concentrations of those components were relatively low during the non-Asian dust periods, and this could be assured by relatively high influx frequencies of the air masses through the East Sea and the north Pacific. The concentration of $nss\text{-}SO_4^{2-}$ was also relatively high during the Asian dust periods, and NO_3^{-} concentrations had a similar pattern with those of the soil components. This could be reasoned by the same backward trajectory explanation, of which most of the air masses had been moved from China into Jeju Island during the Asian dust periods.

Conclusion

In this study, the collection of size-segregated atmospheric aerosol samples had been made at 1100 m site of Mt. Halla in Jeju Island and analyzed their compositions. Based on the analytical results, the size distribution of aerosol components and the composition changes between Asian dust and non-Asian dust periods were examined. From the investigation for the size distributions of aerosol components, the anthropogenic aerosol species such as $nss\text{-}SO_4^{2-}$, NH_4^{+}, and Pb were mainly distributed in a fine particle mode, but NO_3^{-} was existed relatively more in a coarse particle mode. On the

other hand, the soil-originated species such as nss-Ca^{2+}, Al, Fe, Ca, Ba, Ti, *etc*. and the marine-originated species such as Na^+, Mg^{2+}, and Cl^- were mostly distributed in the coarse particle mode. During the Asian dust periods, the concentrations of the soil-originated species such as nss-Ca^{2+}, Al, Ca, Fe, Ti, Mn, Ba, Sr have increased as 2.7-4.2 times as compared to those during the non-Asian dust periods. Meanwhile the concentrations of nss-SO_4^{2-} and NO_3^- have increased as 1.4 and 2.0 times, and on the contrary NH_4^+ concentrations have a little bit decreased during the Asian dust periods. Especially it was evident that most soil components were distributed in coarse particles and showed high concentrations during the Asian dust periods. The backward trajectory analysis has also confirmed that those components are moved mostly from the deserts and loess plateaus in China into Jeju Island.

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