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Interpretation of the Chemical Transformation of Individual Asian Dust Particles Collected on the Western Coast of Korean Peninsula

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ABSTRACT

This paper is focused on the comprehensive and detailed interpretation for the chemical transformation of individual Asian dust (hereafter called "AD") particles during long-range transport from source regions to receptor area. A multi-stage particle sampler was operated at a ground-based site in Taean, Korea directly exposed to the outflow of air masses from China during AD period in April 2003. Both quantitative and qualitative analyses for size-classified individual particles were carried out by a microbeam X-ray fluorescence (XRF) method and a microbeam Particle Induced X-ray Emission (micro-PIXE), respectively. Among major characteristic elements, the elemental masses of soil derived components, sulfur, and chloride varied as a function of particle size showing the monomodal maximum with a steeply increasing at 3.3-4.7 μm particle size. Although the details on chemical composition of AD particle collected on a straight line from source area to our ground-based site are needed, a large amount of Cl coexisted in and/or on AD particles suggests that AD particles collected in the present study might be actively engaged in chemical transformation by sea-salt and other Cl containing pollutants emitted from the China's domestic sources. Through the statistical analyses it was possible to classify individual AD particles into six distinct groups. The internally mixed AD particles with Cl, which has various sources (e.g., sea-salt, coal combustion origin HCl, gaseous HCl derived from the adsorption of acids to sea-salt, and Cl containing man-made particles) were thoroughly fractionated by the elemental spectra drivened by the double detector system of micro-PIXE.

Key words: Asian dust particles, Chemical transformation, XRF, Micro-PIXE

1. INTRODUCTION

During long-range transport of air masses containing AD particles over China continent, Yellow Sea, and Korea Peninsula, AD particles can encounter gaseous or particulate matters and react with each other, leading to a chemical transformation (Hwang *et al*., 2008; Roth and Okada, 1998; Iwasaka *et al*., 1988). As suggested by Li and Okada (1999), the artificial fine particles emitted at the industrial areas in China may be incorporated into AD particles before getting out of China. The AD particles mixed with various anthropogenic pollutants can then be subject to long-distance transport to the receptor areas in East Asia as well as the Pacific Ocean.

During the past few decades, a good few researches have been carried out to the study of the processes of chemical alteration of AD particles like gas-to-particle transformation, coagulation, and growth processes of particles during long-rage transport (Zhang *et al*., 2001; Zhang and Iwasaka, 1999; Duce *et al*., 1980).

Coal-fired plants produce about 80 percent of China's national electricity output (China's Development Research Center of the State Council). Coal-fired power stations produce large quantities of waste ash and fly ash. These coal plants worsen air quality and create acid rain which then hurts soil quality and food safety. The harmful by-products of coal combustion of China should be a grave pollution problem in East Asia. When one assesses the chemical transformation of AD particles he must therefore consider these various by-products of coal combustion.

AD particles would also be expected to be affected by the local sources such as of coal-fired power station operating at receptor area. Even though sulfur dioxide (SO_2) can be removed and recovered in the flues of the power station, small amounts of SO_2 is still emitted from coal fired power station. This locally exhausted

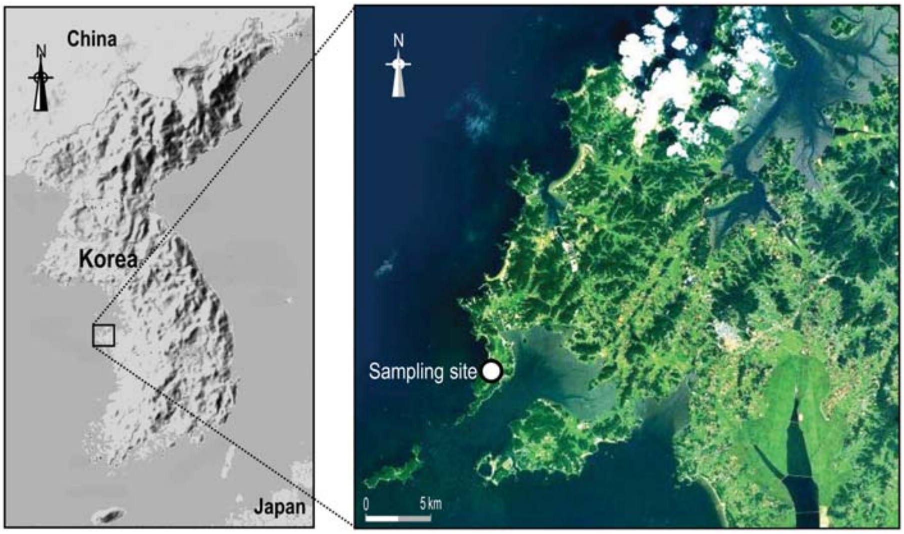

Fig. 1. Maps showing particle collection site on the western coast of Korean Peninsula.

SO_2 can cause additional change in chemical properties of AD particles.

A study of HCl emissions from the seven coal-fired power plants in Maryland indicates that HCl emissions in 1991 totaled between 4,500 and 6,700 tons per year (Griffin, 2010). Therefore, the actual impact of HCl emitted from coal-fired thermal power plants should also be carefully considered on the chemical transformation of AD particles. Hence, as mentioned above, in consideration of the HCl drived from coal combustion, a thorough assessment is crucial for the diagnosis of an aging process of AD particles.

The main objective of this study was assessing the aging processes of individual AD particles collected on the western coast of Korean Peninsula through both quantitative and qualitative X-ray microbeam techniques.

2. MATERIALS AND METHODS

2. 1 Collection of Size-segregated Ambient Particles

Aerosol sampling was performed at a ground-based site (N36.7; E126.2) with a height of 15 m above ground in Taean (see Fig. 1), Korea during AD period in April 2003. Because of its closeness to China, this sampling site marked as an empty circle in Fig. 1 can be directly exposed to the outflow of air masses from China during springtime (see Fig. 2). Thus, this site is one of the well suited areas to measure AD in Korean Peninsula.

As one of local emission sources, the Taean thermal power generation station is located in the nothernest site about 20 km far away from our ground based particle sampling site (see Fig. 2). This power generation plant is operated with the bituminous coal that was mined at Guizhou Province located in southwest China. More details on this power generation station was already described in early study (Ma *et al.*, 2010).

For the purpose of segregating particles as a function of their size, an Andersen impactor sampler (AV-100, Tokyo Dylec Co.) was operated with 28.3 liter per minute flow rate on April 12, 2003. The file up of particles caused a long sampling duration time obscures the chemical determination of particle-to-particle variations. Therefore, the duration of sampling was adjusted to 50 minutes and 40 minutes at stage 1 to 3 and stage 4 to 5, respectively. This short sampling duration time is also a great way to estimate the particle composition because the long collection process can cause condensation or evaporation of volatile compounds, or chemical reactions. The heavy particle build-up under each nozzle often prevents sharp cut-size characteristics. A short sampling duration time also parcially partially reduces particle bounce and blow-off that may otherwise occur.

During sampling period the range of wind speed was 3.1-4.5 m s^{-1}, and it was generally blowing from west. The temperature was around 6.8-7.4°C and average relative humidity in sampling period was 69%.

2. 2 Chemical Analysis of Individual AD Particles

2. 2. 1 Quantitative Analysis

In the present study, the elemental mass of individual AD particles was determined by an X-ray microprobe

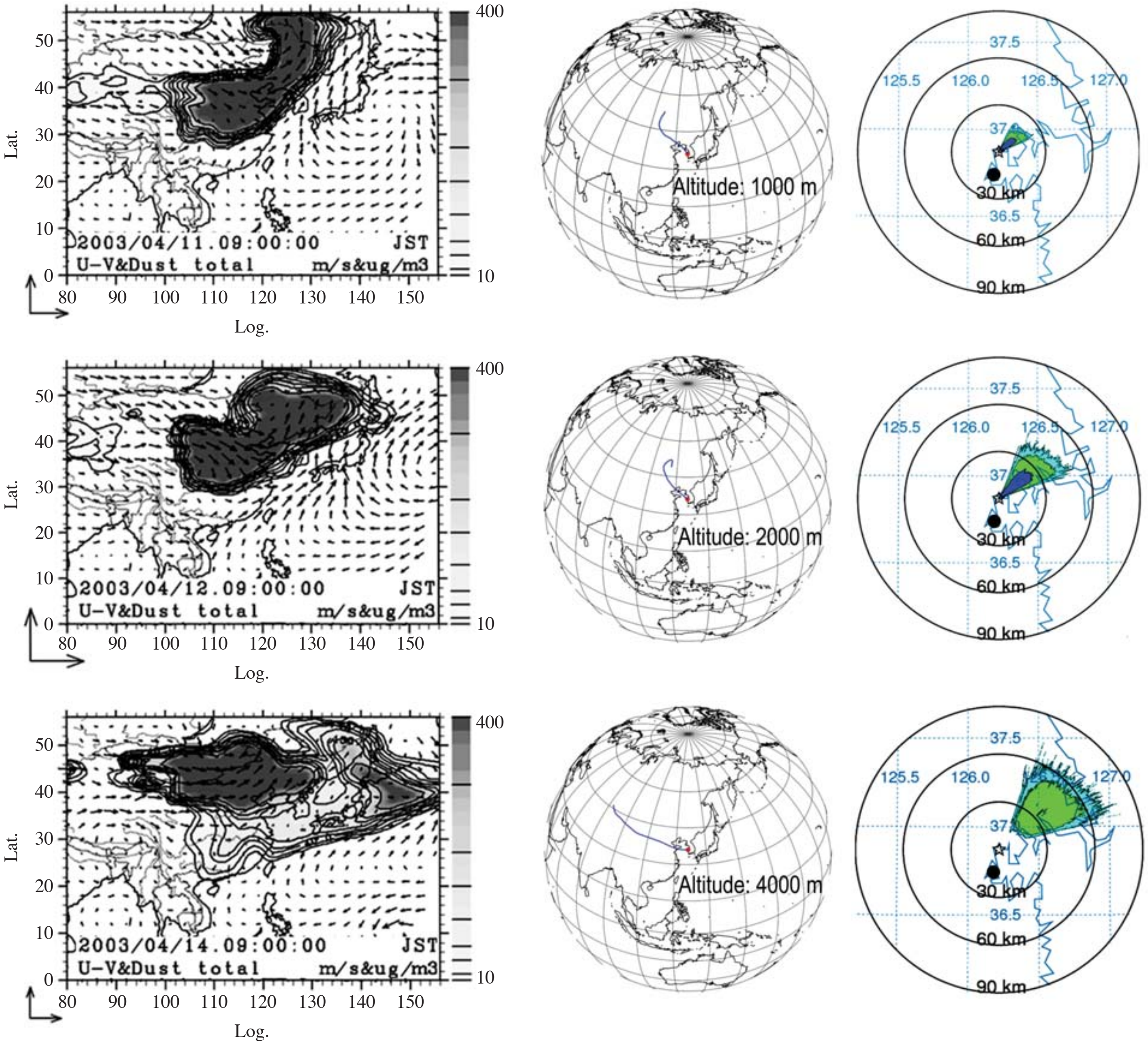

Fig. 2. Forecasted dust moving and concentration by the CFORS model (left), the METEX backward trajectories started at our sampling site (center), and the forward aerosol distributions at Taean thermal power generation station simulated by the NOAA ARL HYSPLIT model (right). A filled circle in HYSPLIT model results indicates the ground-based sampling site.

system equipped at the BL-37XU of Super Photon ring 8 GeV (SPring-8). By virtue of this micro-analytical technique based on XRF method, multiple elements were successfully analyzed at femtogram level sensitivity.

Details of the set-up of analytical facility and the calibration of the X-ray microprobe system to yield quantitative measures of elemental mass in individual AD particles have been described elsewhere (Hayakawa *et al.*, 2001). Through the instrumentality of this XRF analytical technique, a total of 150 individual AD particles at each stage of Andersen air sampler were analyzed.

2. 2. 2 Qualitative Analysis

Although the excellent sensitivity is one of better traits, the restriction of elemental composition ($Z > 12$) is the greatest flaw in the XRF method of present situation for proper understanding of the chemical transformation of AD particles by sea-salt and trace elements derived from local sources. However, micro-PIXE analysis that was installed at the division of Takasaki Ion Accelerator for Advanced Radiation Application (TIARA) in Japan Atomic Energy Research Institute (JAERI) is employing the double detector system (Sakai *et al.*, 2005) and this system is helpful in overcoming the problem of XRF method. As already introduced in earlier study (Ma, 2010), the double detector system of micro-PIXE that combines HP-Ge and Si (Li) X-ray detectors has a broad range (including Na) of target elements. This micro-PIXE still has problem that qualitative analysis of element is not yet facilitate.

The instrumental set-up and detection efficiency of the double X-ray detectors system for micro-PIXE analysis have been described in detail elsewhere (Sakai *et al.*, 2005).

3. RESULTS AND DISCUSSION

3. 1 Forecasted AD Moving

The Chemical weather FORecasting System (CFORS) developed by Research Institute for Applied Mechanics, Kyushu University (Uno *et al.*, 2003) and intensively used during the ACE (Asia Pacific Regional Aerosol Characterization Experiment)-Asia shows AD inflows into East Asia including our sampling site (Fig. 2). Detailed description of CFORS such as general concept of model system and tracer modules can be found elsewhere (Uno *et al.*, 2003).

The Meteorological Data Explorer (METEX) (Center for Global Environmental Research, 2010) backward trajectories (1,000, 2,000, and 4,000 m heights of air parcel) for 72 hours trajectory length started at our sampling site at 09 UTC, April 12, 2003 is also displayed at center of Fig. 2. According to these model results (i.e., AD inflows into East Asia and the movements of air parcel during field event), a dense AD storm on April 11, 2003 was coming from the desert and loess regions in eastern China and southern Mongolia through urban Beijing metropolis by the prevailing winds originated from northwest. And it was extended to the Korean Peninsula after passing through the Yellow Sea.

Since four plants (a total power generating capacity of about 2.7 GW), owned by Huaneng Power International, Datang International Power Generation Co Ltd, China Shenhua Energy and Beijing Jingneng Thermal Power Co Ltd, exist in urban Beijing (China's Development Research Center of the State Council), the AD particles passed over urban Beijing metropolis easily contacted with the pollutants originated from coal-fired power plants.

However, there remain a lot of uncertainties regarding the linkage between chemical alteration of AD particles and the emission sources of receptor region such as a large scale a coal-fired power plant, in particular under the stable meteological condition. Therefore, in the present study, the National Oceanic Atmospheric Administration (NOAA) HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) dispersion model (Rolph, 2003) was run to estimate the impact of locally emitted air pollutants from the Taean thermal power generation station on chemical transformation of AD particles. A detailed model description of HYSPLIT was given by Rolph (2003). The right side of Fig. 2 displays the simulated forward aerosol dispersion (composited aerosol layer from the surface to 500 m) started from Taean thermal power generation station. The results of this HYSPLIT dispersion model indicate that the aerosols emitted from a coal-fired power plant bounded for the northeast part

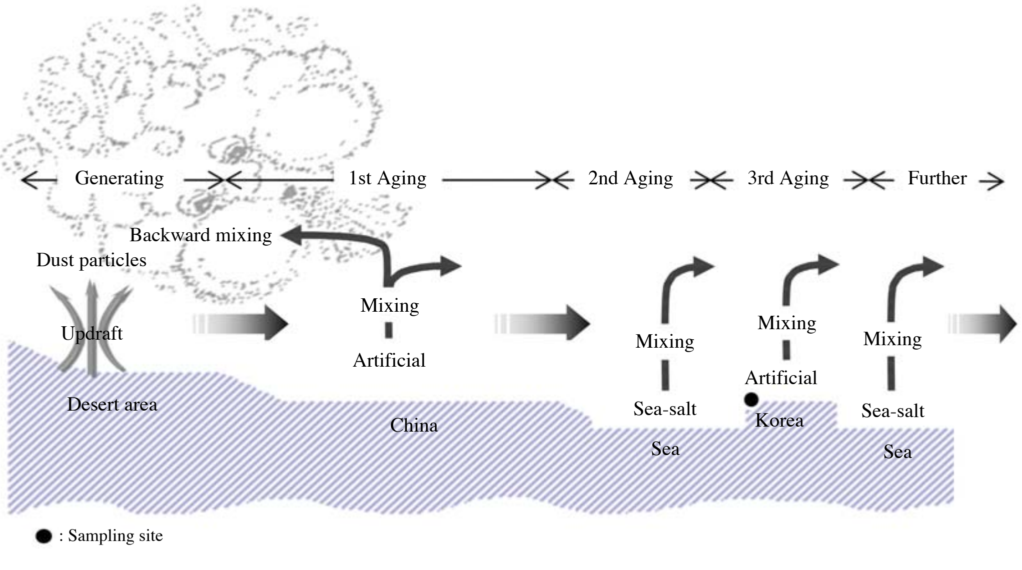

Fig. 3. An illustrative explanation of the transformation processes of the AD particles during long range transport.

of the Korean Peninsula. Thus it suggests that the pollutants emitted from the Taean thermal power generation station did not directly affect the aging processes of AD particles.

Fig. 3 shows an illustrative explanation of the transformation processes of the AD particles between source area and our sampling site during long-rage transport. After finishing the first aging process, AD particles are often condemned to get together with sea-salts (i.e., second aging process) then experienced the third aging process. Hwang *et al.* (2008) reported that AD particles were mixed with sea-salts entrained over the Yellow Sea, as well as air pollutants from the eastern China coastal areas.

3. 2 Size Reliance of Elemental Mass of Individual AD Particles

As mentioned earlier, a total of 150 individual particles chosen indiscriminately for each four-stage from stage 2 to 5 (4.7, 3.3, 2.1, 1.1 μm aerodynamic size, respectively) were irradiated by XRF microbeam. The summary of elemental masses for six characteristic elements (Al, Si, S, Cl, V, and Cr) in individual particles was visually summarized by box plots in Fig. 4.

The long box and whisker, in particular for Al, Si, and S, displayed in Fig. 4 indicates that the individual particles collected during our intensive measurement have a complex chemical nature derived from various mechanisms. As might be expected, as the representative soil components, the masses of Al and Si showed relatively high levels with a variation in wide range, while minor trace elements (V and Cr) had negligible mass within a small variation range relative to the other elements. Elsewhere, in comparison to mineral compositions, Cl showed more or less uniform mass distribution with a high mass through whole particle size. As a rather distinct phenomenon, elemental masses of individual particles show particle size dependence marking a high level at stage-3 for Al, Si, Cl, and V. On the other hand, the masses of S and Cr were observed to increase in the fine particle fraction (1.1-2.0 μm).

For the purpose of thorough estimating of the change in chemical quality of AD particles, the ratios of four kinds of elemental masses to Si mass for size-resolved individual particles were determined (Fig. 5). The numerical levels on upper adjacent refer to the medians. The elemental mass ratio of desert sand is the referential average value of desert sands collected at four different desert regions (Yinchuan, Wuwei, Dulan, and Yanchi) in China (Ma *et al.*, 2008). This box plot, comparing four size fractions and desert sand for the output of mass ratio of element to Si, shows that ratios have a significant fluctuation with respect to particle size. As compared with the median value of dust sand, with the exception of V in particle size range from 3.3 to 4.7 μm, there was a tendency towards overwhelmingly higher ratios in size-resolved ambient particles. It could therefore be suggested that AD particles collected in the present study might be actively

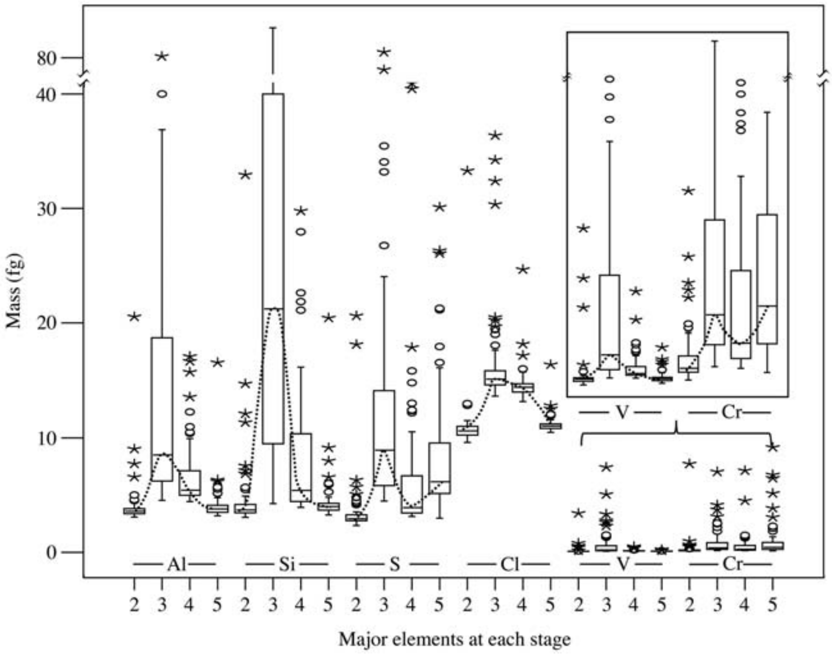

Fig. 4. Box plot showing size dependence of elemental mass of individual AD particles.

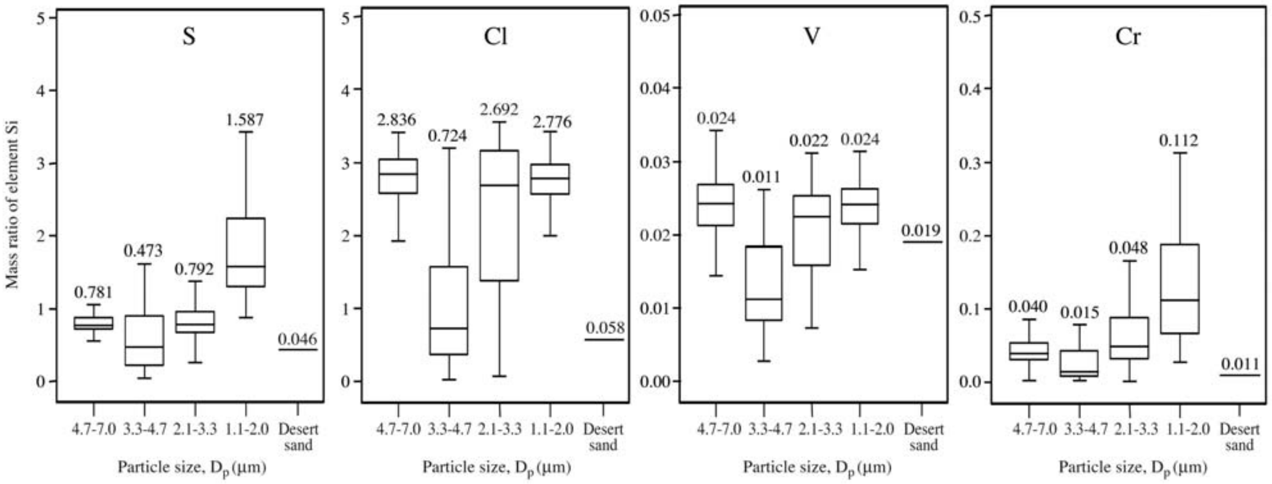

Fig. 5. Ratios of four kinds of elemental masses to Si mass for size-resolved individual AD particles. Dp μm means the 50% cut-off size of Andersen sampler.

engaged in chemical transformation. The fine particles collected on stage-5 (1.1-2.0 μm) has the highest ratio; the particles deposited on stage-3 (3.3-4.7 μm) have the least value of ratio. As described earlier, high mass ratios of S and Cr are likely responsible for the unusually high levels in fine fraction of particle. The increase of S mass ratio in fine mode particle can be explained by the inflow of artificial S containing fine particles into our sampling site. It is well known that SO_2 can be emitted from the fossil fuel-fired generating facilities like coal-fired power plants. The reason for the high elevating of Cr mass ratio in fine fraction of particle might be that the harmful substances like mercury and chromium can emitted via coal power station flues (Griffin, 2010) and they can also inflow into receptor area during AD event.

It is noteworthy that the ratio of Cl to Si is quite high (from 0.724 to 2.836) compared to other elements. This Cl enrichment can be apparently explained by the fact that AD particles aged by gaseous and particulate Cl emitted from the China's domestic sources were additionally reacted with sea-salt. Although various polluters such as automobiles and municipal incinerators can also be considered as the sources of ambient Cl, one cannot rule out the possibility that Cl was driven from coal consumption (e.g., coal-fired power plant). About this Cl enrichment of individual particles, details are discussed at the final part of this manuscript.

3. 3 Classification of Individual AD Particles

3. 3. 1 By Cluster Analysis

In order to classify individual AD particles, the elemental masses measured by XRF microbeam were subjected to cluster analysis. Most particles could be clustered with highly dissimilar dendrogram which summarizes the process of clustering. It could therefore be suggested that elemental compositions were chemically heterogeneous throughout the whole individual AD particles.

As a result, individual AD particles can be categorized into six clusters. Cluster I (Si > Cl > Al > K > S > P) with a small portion of total particle population was found to be high mass for soil components and Cl. Cluster II (Cl > Si > Al > S > P) and Cluster III (Cl > Si > Ca > Al > S) with also a small account rate of total particle population were showing Cl and mineral compositions-rich. Cluster IV (Cl > S > Si > Al) with a poor particle number had high mass for Cl and S with coexisting of crustal elements. Cluster V (Cl ≫ Si > Al > S > P) could be categorized as the particles mainly containing Cl with slight soil components. Cluster VI (Cl > Al > Si > S) showing absolute share of particles seemed to express the modification of AD particles with marine derived Cl, and/or artificial Cl especially drived coal-fired power plant, and S.

3. 3. 2 By Micro-PIXE Spectra

As discussed above, Cl-rich particles had an ascendancy over other types of AD particles and it would mainly be induced by the particle-to-particle homogeneous reaction between AD paricles and sea-salt particles. On the other hand, the gaseous HCl generated by the reaction between sea-salt and acids (e.g., heterogeneous reactions of sea salt with HNO_3 and H_2SO_4) might have relevance to this enrichment of Cl (Roth and Okada, 1998). It can also be considered that large amounts of other chloride-containing fine particles

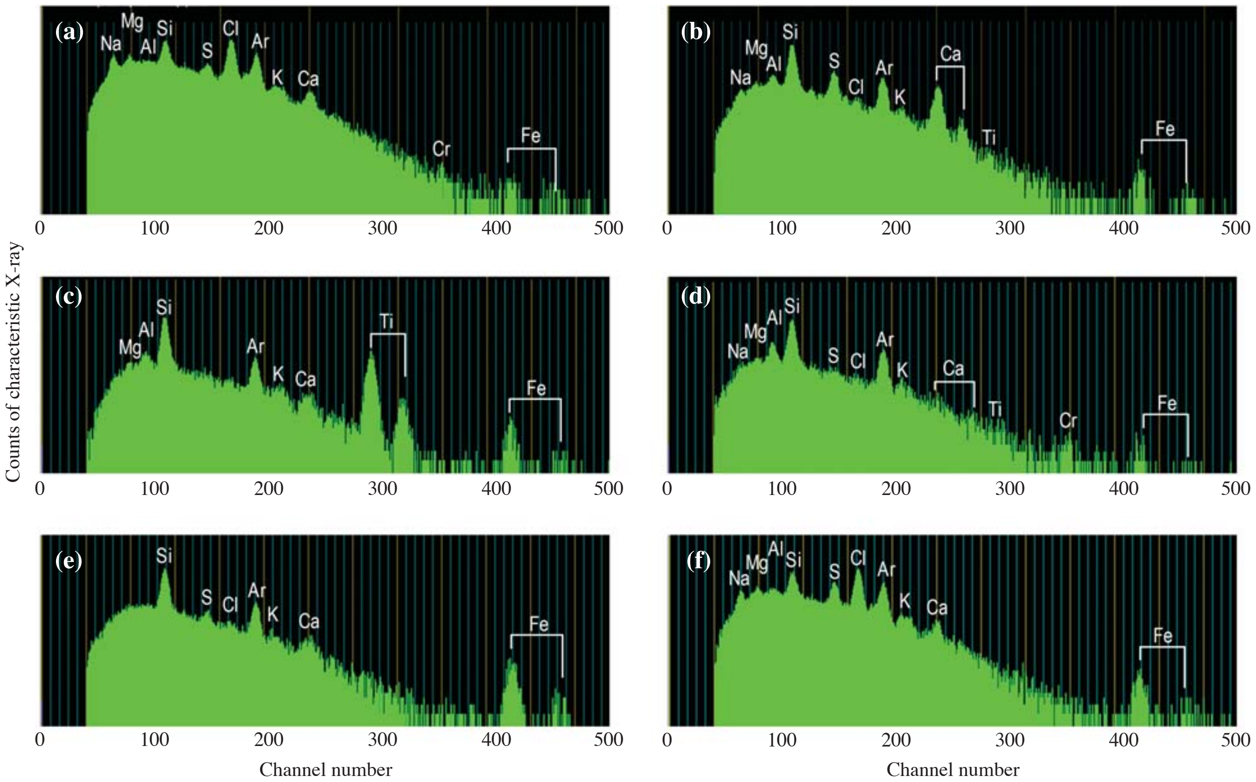

Fig. 6. Micro-PIXE spectra of individual AD particles experienced different aging processes during long-range transport.

(e.g., NH_4Cl or the vaporized gaseous HCl from fine mode NH_4Cl druing the state of increased temperature) were mixed with AD particles and had a significant impact on the present results. Elsewhere, under the state of coal consuming (e.g., power plant) in China, the impact of HCl originated from coal combustion should also be actually considered.

Therefore, in dealing with Cl, one has to distinguish among sea-salt, HCl emitted from coal combustion at power plant, gaseous HCl derived from the adsorption of acids to sea-salt, and other Cl containing man-made particles.

The elemental spectra driven by the double detector system of micro-PIXE are useful for fractionating AD particles into internally mixed AD particle with Cl. Fig. 6 is displaying micro-PIXE spectra for several distinctive AD particles experienced different aging processes during a long-range transport to our sampling site.

Particles (a) and (f) were remarkable Cl-rich AD particles that might be traced back to below two aging processes. (1) artificially generated $HCl_{(g)}$ was adsorbed on an AD particle before getting out of China and then experienced a further aging process with sea-salt. As described earlier, the man-made $HCl_{(g)}$ was emitted mostly from coal combustion at power plants. It also diversely generated from the large-scale production of chemical reagent and vinyl chloride for PVC plastic and other numerous smaller-scale applications including household cleaning, production of food additives, descaling, and leather processing. Even though, HCl is fairly short-lived in the atmosphere (one to five days) (Griffin, 2010), the HCl generated in China and not sufficiently reacted with ammonia or alkaline cations such as calcium or potassium before outgoing China might have a chance to mixing with an aged AD particle. (2) enriched Cl by both $HCl_{(g)}$ generated by [$2NaCl+H_2SO_4$ (coal-fired power plant origin) ($Na_2SO_4 + 2HCl\uparrow$)] and sea-salt above the Yellow Sea. Especially, particle (a) was containing a small peak of Cr probably emitted from coal-fired power plant. Although Cr compounds are used in paints, metal polish, and the tanning of leather, Cr is often found in coal-fired flyashes (Brigden *et al*., 2002; Llorens *et al*., 2001). Also several earlier studies (Ma *et al*., 2010; He *et al*. 1998) indicated that the high level of Cr concentration might be a chemical peculiarity of the ash produced from combustion of Chinese coal material.

Particle (b) cloud be classified as an AD particle mixed with sea-salt without further aging process. Meanwhile, particle (c) cloud be categorized as a non-aged AD particle with Ti and Si-rich. Particle (d) could be classed as a Si and Al rich AD particle with anthropogenic Cr and disregardably small peaks of sea-salt. Particle (e) had the appearance of the aging processes with acidic materials such as the artificially emitted $HCl_{(g)}$ and SO_2.

4. CONCLUSIONS

In this study, we made an attempt to provide a thorough discussion on the aging processes of individual AD particles during long-range transboundary. A comparison of the chemical properties of desert sands at four different desert areas in China reported earlier (Ma *et al.*, 2008) and those of AD particles collected at our Taean site suggests that AD particles collected in the present study might be actively engaged in chemical modification. Especially, a large amount of Cl coexisted in and/or on AD particles suggests that the chemical transformation of AD particles vigorously occurred by sea-salt and other Cl containing pollutants emitted from the China's domestic sources.

Despite the shortcomings of two different analytical methods, a combinational approach of the micro-XRF technique in SPring-8 with the micro-PIXE in TIARA provided insight into the chemical nature of individual AD particles, especially distinguishment of Cl sources from sea-salt, power plant, and other man made. Although the numerical limitation of particles that analyzed and discussed in the present study is apprehensive, from the results of our single AD particle analysis it was possible to thoroughly fractionate the internally mixed individual AD particles into several types depending on their aging processes.

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