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심포지움 3) Wet Removal of Asian Dust Particles Confirmed by Individual Cloud and Rain Drops Studies

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1. INTRODUCTION

Asian dust (AD) storm is a serious and growing environmental problem in East Asia as well as the Pacific Basin. The AD finally dissipates when the particles are removed from the atmosphere by dry and wet removal processes. Until now, numerous studies aided by the field and model simulations have been carried out to determine the characteristics of AD particles. However, there were only a few studies which focused on the assessment of the wet scavenging of AD storm particles. The results of such studies are expected to provide a better knowledge concerning their removal characteristics of AD storm particles. In order to fully understand the wet scavenging properties of AD particles and those wet deposition amounts, the chemical characteristics of the cloud and rain samples collected during the AD storm event have to be specified. In the present study, both individual cloud droplets and individual raindrops were collected the AD storm period. Moreover, their chemical compositions were determined by PIXE and microprobe analytical techniques.

2. METHODS

2.1 Sampling of individual rain and cloud drops

Yellow rainfall episode occurred in the Kyoto region of Japan on Apr. 7, 2001. This yellow coloration of rain was induced by soil particles originatingfrom the desert and loess areas in China. For the sampling of individual raindrops as a function of their size, we designed the sampling and handling apparatuses. Sampling apparatus consist of a dewar vacuum flask filled with liquid nitrogen and 7-stage stainless steel sieves. More details about sampling and handling processes of raindrops were described elsewhere (Ma *et al*, 2004a). The sampling of individual cloud droplets was carried out on the summit of Mt. Taiko (683 m elevation) in west coast of Japan during dense AD storm event on 22 Mar. 2002. For the purpose of sampling of individual cloud droplets, the collodion film replication method, which was introduced from Ma *et al* (2004b), was applied.

2.2 Analysis of trace elements in residues in individual cloud and rain drops

For the quantification analysis of the ultra trace elements in the individual solid particles retained in cloud droplets, the XRFmicrobeam system equipped at Super Photon ring 8 GeV (SPring-8) BL-37XU was applied. This method has been successfully used to carry out the reconstruction of elemental map and the quantification analysis for multiple elements with ~femto gram level sensitivity. The absolute quantities of elements contained in both the soluble and insoluble fractions of rain drops were determined by PIXE analytical method. More detailed information concerning analytical procedures and experimental set-ups used for PIXE and XRF microbeam system were described elsewhere (Ma *et al*, 2004a; Ma *et al*, 2004b).

3. RESULTS AND DISCUSSION

Fig. 1 shows the four kinds of XRF elemental maps of dry residuals in individual cloud droplets.

Large amount of cloud droplets containing Ca are present on this mapping area (lower left panel). In addition, as a one of substantial fractions of the droplet residues, map of Zn was drown at right upper of Fig. 1.

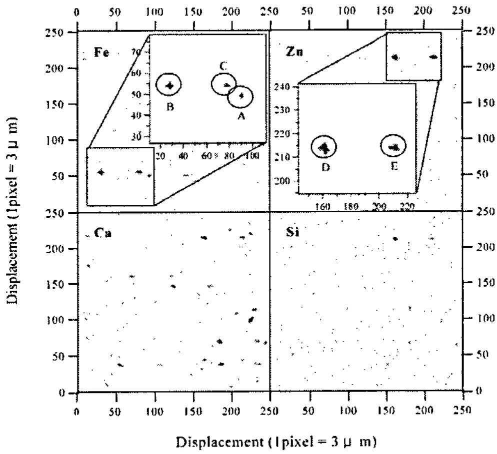

Fig. 1. XRF elemental maps of residuals in individual cloud droplets.

Fig. 2 describes the plotting of E.F.s of elements in dry residual particles in cloud droplets ($(Z/Si)_{cloud}/(Z/Si)_{desert\ sand}$) vs. those in precipitation clouds ($(Z/Si)_{precipitation\ cloud}/(Z/Si)_{desert\ sand}$). Analysis and plotting of E.F.s not only indicate the good correlationship between elemental masses in residualparticles of cloud base droplets and those of precipitation cloud, but also classify elements into soil origin and non-soil origin. A pair of E.F.s with high correlation coefficient suggests that both precipitation cloud and cloud base droplets collected at our sampling sites were affected by long-range transport of Asian dust storm particles.

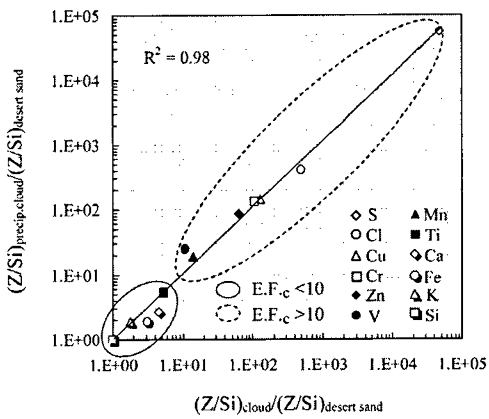

Fig. 2. Plotting of crustal E.F.s of elements in individual cloud droplets vs. those of element in size-classified precipitation cloud. The composition of the desert sand was given by PIXE analysis of sand collected at Yinchuan desert area in China.

An example of the elemental map for Si, S, Cl, K, Ca, and Fe contained in a raindrop is drawn in Fig. 3. Each elemental map was drawn on the 128×128 pixels by scanning of about 1μm micro beam on the sample surface. These visualized elemental maps of six element types in several particle clusters enable us to estimate the chemical mixing state of raindrop residual particles. In addition, it is also presumed that the chemical transformation of dust particles is made by wet scavenging processes.

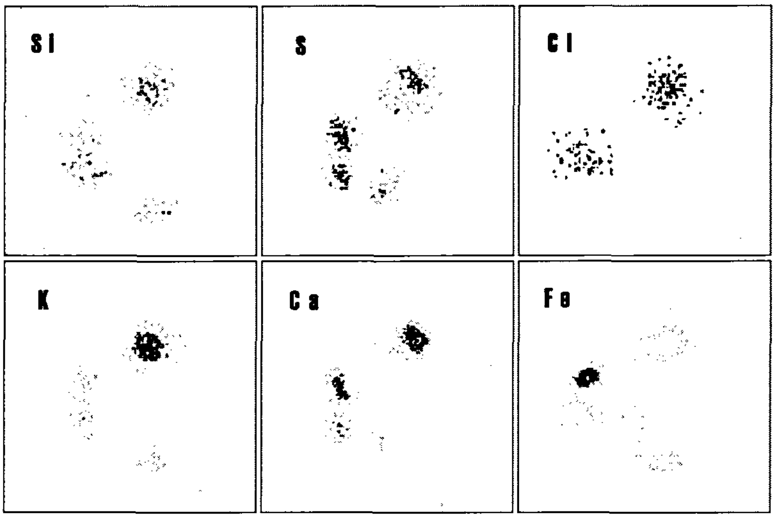

Fig. 3. Elemental maps (scanning area: 760×760 μm^2) on a whole raindrop (0.5 mm diameter) fallen in a yellow rainfall event.

Table 1 summarizes the result of factor analysis showing the sources identified from 17-element in solid particles retained in size-segregated raindrops. Individual solid particles were successfully divided into four factors (94% cumulative), three factors (98% cumulative), two factors (85% cumulative), three factors (96% cumulative), and four factors (96% cumulative) in the raindrop size $Rd_{<0.5mm}$, $Rd_{0.5mm}$, $Rd_{0.7mm}$, $Rd_{1.0mm}$, and $Rd_{>1.7mm}$, respectively.

Table 1. Statistical summary of mass for 17 elements in individual AD storm particles retained in size-segregated raindrops. Rd means the mesh size.

Raindrop size	Factor 1		Factor 2		Factor 3		Factor 4		Cumulative %
	Identification	% of Variance	Identification	% of Variance	Identification	% of Variance	Identification	% of Variance	
[a]$Rd_{<0.5mm}$	Crustal>Marine	48.49	Crustal	28.32	Crustal>Marine>Sulfur	10.64	[b]Artificial>Crustal	6.79	94.24
$Rd_{0.5mm}$	Crustal>Sulfur>Marin	61.37	Crustal	28.45	Sulfur>Marine>Crustal	7.93	-	-	97.74
$Rd_{0.7mm}$	Marine>[c]Crustal	52.92	[d]Crustal>Marine	32.28	-	-	-	-	85.20
$Rd_{1.0mm}$	Crustal>Sulfur	63.48	Crustal	17.84	Crustal>Sulfur>Marine	14.97	-	-	96.30
$Rd_{>1.7mm}$	Crustal>Sulfur	46.06	[e]Crustal	32.80	Crustal>Sulfur>Marine	9.94	[f]Crustal	6.86	95.66

Fig. 4 illustrates the scavenging of AD particles by rainout and washout mechanisms and their transformation within the droplet. It is known that fresh mineral particles are mostly hydrophobic. However, if the ambient water droplets spread on them as a horizontal film, their surface can be said to be perfectly wettable (hydrophilic). This enables the transfer of mineral particles into cloud droplets via nucleation scavenging. If mineral particles do not take up any water (i.e. they cannot be dissolved in cloud droplet by itself), they can be scavenged by droplet (i.e., through collision).

Furthermore, it should be addressed that AD particles can also be scavenged by falling raindrops. This washout process generally comprises Brownian diffusion, interception, and inertial impaction.

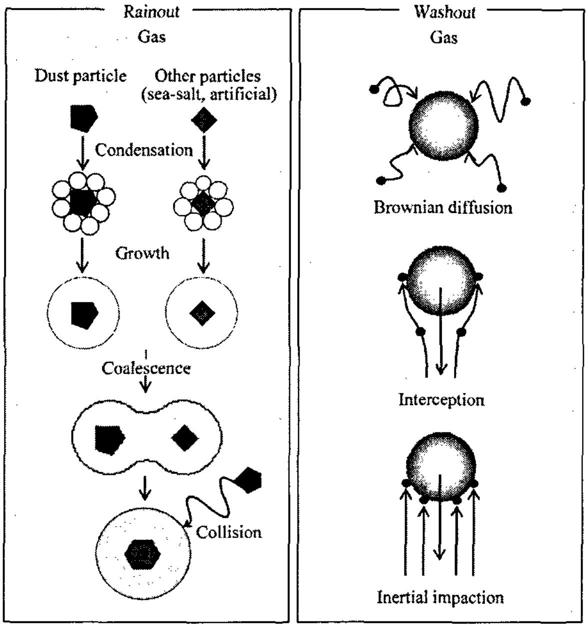

Fig. 4. An illustration depicting the scavenging mechanism of AD particles by rainout and washout mechanisms.

REFERENCES

Ma C.-J., S. Tohno, M. Kasahara, and S. Hayakawa (2004a) The nature of individual solid particles retained in size-resolved raindropsfallen in Asian dust storm event during ACE-Asia, Japan. Atmospheric Environment, 38, 2951-2964.

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