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Microbeam Analysis of Yellow Sand Dust Particles

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Abstract. A microbeam system was applied to analysis of yellow sand dust particles. These particles from the Asian continent sometimes cause turbid conditions in Japan, especially in spring. These particles are sometimes deformed by mixing with anthropogenic aerosols. For better understanding of the deformation mechanism, analysis of single particles is indispensable. For this purpose, we developed a microbeam analysis system with a spatial resolution better than 1 μm. The system is composed of two X-ray Si(Li) detectors for PIXE, an annular ion-implanted Si detector for RBS, an ion-implanted Si detector for off-axis STIM and a Si-PIN photodiode for direct STIM analyses. The combination of PIXE, RBS and off-axis STIM methods enabled simultaneous analysis for hydrogen to metal elements and revealed the chemical composition of these particles. Yellow sand dust particles were impacted on a thin polycarbonate film and analyzed by the microbeam analysis system. After quantification of these particles, the chemical composition of each particle was obtained. Silicon, calcium and light elements are the main elements of the particles, originating mainly from soil dust from Asian continent. These particles contain sulfur and heavier elements and are deformed by mixing with anthropogenic aerosols. Sodium, magnesium and chlorine elements which come from marine aerosols are also included in these particles and may be adsorbed during transport from the Asian continent. Single particle analysis of yellow sand dust will lead to a better understanding of their deformation process during transport.

Keywords: Microbeam analysis, PIXE, RBS, STIM, Yellow sand

INTRODUCTION

Yellow sand dust particles from the Asian continent sometimes cause turbid conditions in Japan, especially in spring, which is known as a Kosa event. The particles are carried from the Yellow River basin and deserts, and are considered as a natural phenomenon. The particles are principally composed of minerals and their diameters are around 4 μm. Recently, it is regarded as an environmental problem due to forest reduction, soil degradation and desertification rather than a natural seasonal phenomenon. Furthermore, these particles are sometimes deformed by mixing with anthropogenic aerosols during transport and affects agricultural production, climate, living condition and human health. Therefore, physical and chemical changes during transport as well as mechanism of formation, growth and transport have to be known. For better understanding of these mechanisms, analysis of single particles is indispensable. For this purpose, we developed a microbeam analysis system with a spatial resolution better than 1 μm. The combination of PIXE, RBS and off-axis STIM methods enabled simultaneous analysis for hydrogen to metal elements and revealed the chemical composition of these particles. In this study, we applied the system to analyze yellow sand dust particles and investigated their properties.

SAMPLING

Aerosol particles were collected at the campus of Nagasaki University (32.78°N, 129.87°E; 20m) on 9,10 and 19, May 2005. Nagasaki University located at Kyushu Island in western part of Japan, where yellow sand dust particles largely affects the daily life. Recently, fine particles whose sizes are in the range of 0.3-1.0 μm in diameter occasionally cause significant turbid condition over the Nagasaki area. Since yellow sand dust particles are mostly larger than 1.0 μm, these fine particles are mainly anthropogenic aerosols and not the components of yellow sand dust particles [1,2]. To collect true yellow sand dust particles, sampling was carried out when concentration of coarse particles increases and that of fine particles shows usual atmospheric condition by using an optical particle counter (OPC). On 9 and 10, May, coarse particles showed higher concentration, indicated Kosa events and fine particle showed as usual condition. On 19, May, concentrations of fine and coarse particles were low. Sampling was carried out using a single stage impactor developed in our laboratory [3]. Aerosol particles were impacted on a thin polycarbonate film [4] at flow rates of 1 *l*/min (face velocity: 530cm/sec). The effective 50 % cut-off diameter is estimated to be ~2 μm and sufficient for collection of yellow sand dust particles. Sampling times were 6 hours. In total 3 samples were collected.

ANALYSIS SYSTEM

Analysis was carried out using the microbeam analysis system at Tohoku University. Technical details of the microbeam and analysis system were presented in previous papers [5, 6] and further development was carried out for the efficient analysis of single aerosol particles. For multimodal analysis, two X-ray detectors for PIXE analysis and three charged particle detectors for RBS and STIM were mounted simultaneously.

Two X-ray detectors were set in vacuum at 115 degree with respect to the beam axis. The first one has large sensitive area (60mm^2, LS60148; Princeton Gamma-Tech) and is suitable for trace elemental analysis. To reduce pile-up events or deformation of the spectrum by recoil protons, a Mylar filter (300 μm) was attached to the front of the detector. Maximum solid angle is ~ 0.13 sr. The second detector (LS10138; Princeton Gamma-Tech) with a high-energy resolution (~ 136eV), a thin Be entrance window (7.5μm) and small solid angle of ~0.02 sr was used to detect X-rays lower than 5 keV. The system enables to detect X-rays ranging from 1 to 30 keV with good energy resolution and detection efficiency.

An ion-implanted Si annular detector (ANPD300-19-100RM; Canberra) was used for RBS measurement. The annular detector is very efficient, improving solid angle (~ 0.17 sr) without deteriorating angular spread and without interfering with the Si(Li) detectors. Mean scattering angle was 170 degree. Another annular Si surface barrier (TC-019-150-100; Ortec) detector was used in the previous studies, which caused parasitic field contamination and deteriorated beam performance [7]. The new annular detector improved beam performance and solid angle.

Hydrogen was analyzed using off-axis STIM. In the previous study, the Si-PIN photodiode was used for both off-axis STIM and Direct STIM measurements [6]. Superior energy resolution of the Si-PIN photodiode was efficient for direct STIM measurement [8]. However, the small size of the detector occasionally restricted detection efficiency. In order to solve the problem, an ion-implanted Si detector (BU-012-050-100) with a larger sensitive area of 50 mm^2 was newly installed. Detection efficiency of the system is now three times higher than that of the previous system. The scattering angle was 28 degrees.

RESULTS

Simultaneous PIXE / RBS / off-axis STIM analysis employed for scanning areas of 25 ×25 μm^2 and 100×100 μm^2. Yellow sand dust particles were uniformly distributed, thus direct STIM measurements for defining the analysis area were not needed. Energy of the proton beam was 3 MeV and beam spot size was 1×1 μm^2 with a beam current of 50 ~ 100 pA. Total accumulated charge was around 0.2 μC.

Quantitative PIXE analysis was performed using the GeoPIXEII software [9]. After generating the elemental maps, individual particles were selected from these maps and then PIXE, RBS and off-axis STIM spectra were extracted. Elemental concentrations of these particles for elements heavier than Na were deduced from fitting of the extracted PIXE spectra. Carbon and oxygen were quantified by analyzing the extracted RBS spectra. Concentration of carbon and oxygen were derived from peak yields which were calibrated by measuring peak yields from Mylar films of known thickness. Quantitative analysis of hydrogen was carried out by analyzing the extracted off-axis STIM spectra. For the quantitative analysis of hydrogen, intensities of hydrogen were calibrated by measuring hydrogen yields from Mylar films of different thickness. The relation between hydrogen peak yield and hydrogen quantity was linear and was used to calibrate the quantitative analysis of hydrogen.

After quantification of these particles, the chemical composition of each particle was obtained.

More than 100 particles were analyzed and the elements Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn and Br were quantified. FIGURE 1 shows typical elemental maps of yellow sand dust particles collected on 11 May, 2005, when high concentration of coarse particles was observed. The scanning area was 100 × 100 μm^2 and total accumulated charge was 0.21 μC. Distribution of aluminum equals that of silicon. Chlorine, potassium, calcium, iron, oxygen, carbon and hydrogen also show similar distributions. Aluminum, silicon, calcium, iron and light elements are major components. Number of oxygen atoms of these particles is around eight times higher than that of mineral elements. These particles also contained carbon and hydrogen. It is apparent that these particles mainly existed as oxide or carbonate and were originated mainly from soil dust. These particles might adsorb organic carbon during transportation. In contrast, the particles collected on 19 May, when atmosphere was as usual, sodium-rich and silicon-rich groups, which correspond to marine and soil aerosols, were observed. Half of the particles are estimated to be marine aerosols.

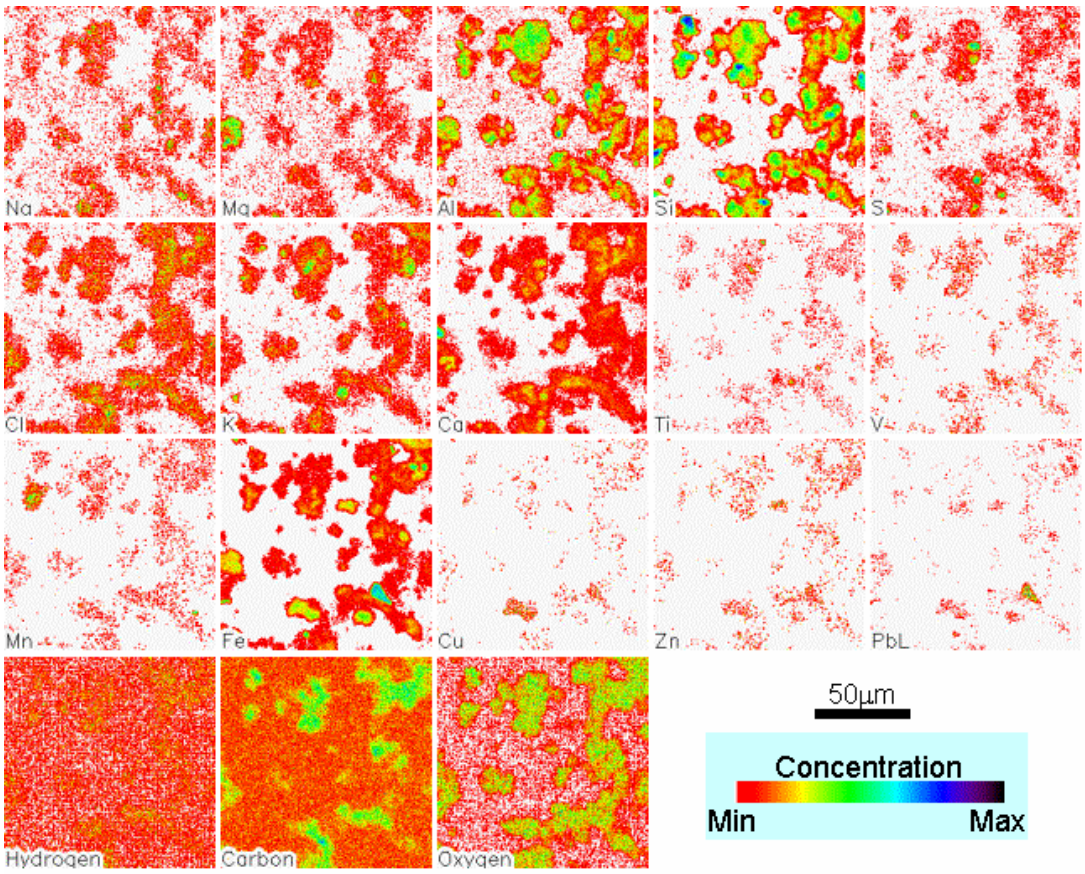

FIGURE 1. Elemental Distribution images of Yellow Sand Dust Particles collected on 11 May, 2005

FIGURE 2 shows the ternary diagram for calcium, silicon and aluminum. In this diagram, marine particles, in which sodium and chlorine are the main components, were excluded. The distribution of the particles which were collected on 10 and 11 May were very similar, except for that of calcium-rich components. On the other hand, the particles collected on 19 May show lower calcium concentration. These facts correspond to the difference of the soil components of Asian continent and Japan Island. Correlation function between calcium and chlorine is shown in FIGURE 3 for these particles. As calcium increases, chlorine also increases for the particles collected on 11 May. Backward trajectory analysis using NOAA HYSPLIT [10] indicated that the particles which came on 11 May took almost the same route from Chinese coast to Nagasaki with a low velocity as the ones came on 10 May. Amount of sodium on 11 May is also larger than that of 10 May. It implies that the particles came on 11 May adsorbed

marine aerosols or deformed by mixing with marine aerosols more than those that came on 10 May. These particles collected on 10 and 11 May contained more sulfur and heavier elements, such as manganese and zinc, than those collected on 19 May. These particles were transported from Asian continent over the industrial area to Japan and might adsorb these elements by mixing with anthropogenic aerosols.

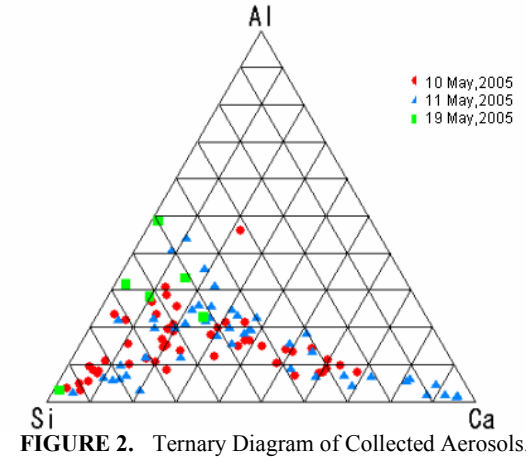

FIGURE 2. Ternary Diagram of Collected Aerosols.

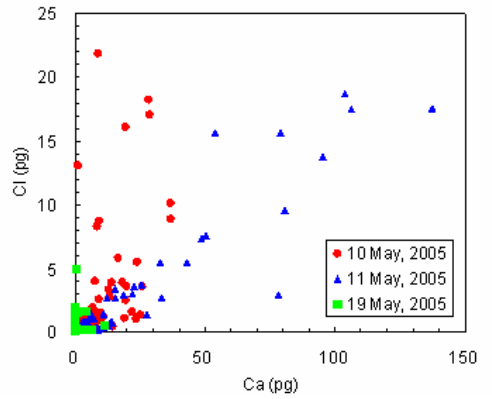

FIGURE 3. Correlation between Calcium and Chlorine.

CONCLUSIONS

The microbeam system was applied for analysis of yellow sand dust particles. Simultaneous PIXE, RBS and STIM analysis enabled to measure the elements from hydrogen to metal elements and revealed the chemical composition of these particles. The major elements of yellow sand dust particles are silicon, calcium and light elements and exist as oxide or carbonate. The ternary diagram of aluminum, silicon and calcium shows the difference of the origin of the soil components of Asian continent and Japan Island. Correlation of calcium and chlorine is different for each Kosa event depending on the difference of transport to Japan Island. Yellow sand dust particles contain more sulfur and heavier elements such as manganese and zinc. These particles were transported from Asian continent over the industrial area to Japan and might adsorb these elements by mixing with anthropogenic aerosols. These results could be obtained by analyzing single aerosol particles and were not obtained by the bulk analysis which averages over many particles. Single particle analysis of yellow sand dust will lead to a better understanding of their deformation process during transport.

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