Time series and source identification of trace elements in atmospheric aerosols measured at Amami-Oshima in the spring of 2001 and 2003 under the APEX field campaign program

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Abstract

An intensive field program was performed to measure atmospheric aerosols at Amami-Oshima, southwest Japan, in April 2001 (E2) and March-April 2003 (E3), under the APEX project. Chemical analysis of the collected aerosols with three size distributions was made for elemental and organic

carbons (EC/OC) by improved method, water soluble ions by ion chromatography, and trace elements by PIXE. This report summarizes the major findings from an analysis of about 20 trace elements focusing on their relationship with soil dusts and anthropogenic aerosols.

The two types of enhancement of atmospheric aerosols were observed in the E2 and E3 experiments. One is high non-sea salt sulfate (nss-SO_4^{2-}) in fine particles ($PM_{2.5}$; 2.5 μm<d), which is an indicator produced from the pollutants emitted from urban and/or industrial area, and the other is high soil dusts which were found when sand storms ("Kosa" event) were observed over Japan Islands. According to the backward trajectory analysis, the air masses rich in high atmospheric aerosols in both types were transported from north China, compared to the air masses from Korean peninsular and Japan islands. It clearly indicates that polluted aerosols mixed with mineral dusts generated inland northwest China or Mongolia were transported to Amami.

(1) Averaged mass concentration in fine and/or coarse particles in E2 and E3 showed that trace elements were divided to two groups. One is the major group with the averaged mass concentration more than 100 ng m^{-3} in fine or coarse (2.5μm<d<10μm) particles, including S, Si, Al, Fe, Na, Mg, K, Ca, Cl. The other is the minor group with the averaged mass concentration less than 30 ng m^{-3} in both fine and coarse particles, including Ti, Mn, Sr, Zn, Pb, Br, Co, Hg, V, Cr, Ni, Cu.

(2) The trace elements higher in the concentration of coarse particles than in that of fine particles were Si, Al, Fe, Na, Mg, Ca, Cl, Ti, Mn, Sr, Co, Hg, Cr, Br, and those higher in fine particles were S, K, Zn, Pb, V, Ni, and Cu.

(3) In the major coarse particles, the dominant source of Si, Al, Fe, Mg, K, Ca, Ti, Mn, Sr, Co, V is estimated to be soil dusts transported from inland China, because of strong positive correlation ($r>0.80$) between every two elements. In contrast, Na, Cl, and Br were derived from sea-salt particles.

(4) In the major fine particles, the dominant source of S, K, Mn, Zn, Pb, and Cu is estimated to be anthropogenic pollutants in urban/industrial area of north China, because of strong positive correlation ($r>0.81$) between every two elements.

(5) The strongest positive correlation in fine particles was found between K and Mn ($r=0.98$), and between Zn and Pb ($r=0.97$). Thus, the dominant source of K and Mn was soil dusts in coarse particles, and anthropogenic pollutants in fine particles, while Zn and Pb showed the only one dominant source of pollutants.

(6) According to the regression analysis, the concentration ratio of Si to Al (Si/Al) was 2.50 ($R^2=0.99$) in coarse particles, almost equal to the value of 2.56 measured in atmospheric aerosols at Nagoya during 1976-1978. In contrast, Al/Fe in coarse particles was 1.35 ($R^2=0.95$), much different from the value of about 0.9 measured in Kyusyu in a period of sand storms in 1988 and 1989.

(7) The mass concentration ratio among Si, Al, and Fe was almost equal in all the E2 and E3 periods. It indicates that even the enhancement of atmospheric aerosols by sand storms did not affect the

concentration ratio of these three major trace elements, while a little bit increase in the concentration of Fe was observed. Soil dusts blown up by the strong vertical motion in inland China could always exist in spring over Amami.

(8) The trace elements in atmospheric aerosols at Amami were also discussed by comparing with those in the averaged composition in upper continental crust, and in atmospheric aerosols measured in Beijing, China, between March 2001 and August 2003.