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粗大粒子、微細粒子中の鉛化学種およびその形成過程に関して
Clarification of lead (Pb) species and its formation mechanisms in coarse and fine aerosol particles using X-ray absorpt

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There are very few studies on chemical speciation and atmospheric chemistry of trace elements. Among the trace elements in the aerosol particles, lead(Pb) has long been measured due to the toxicity of this element. The measurement of Pb isotope ratios in aerosol particles has also been employed as a powerful tracer for air-mass transportation because Pb isotope ratios differ significantly depending on the emission area/source. However the speciation of Pb has not been clarified, although their solubility is important to estimate the health effects for human/animal body. Furthermore, Pb in the aerosol particle is the dominant source of Pb, which is used as oceanic circulation tracer, in the surface seawater. Thus, the speciation of Pb in the aerosol particle is one of the important issue. In this study, we attempted to determine the Pb species in aerosol samples using X-ray absorption fine structure (XAFS) analysis.

Size-fractionated aerosol particles were collected by a high-volume aerosol sampler with cascade impactor at Higashi-Hiroshima. Lead LIII-edge (absorption edge: 13.04 keV) X-ray absorption near-edge structure (XANES) spectra were recorded on SPring-8 on BL01B1 and at KEK PF-AR on NW10A to identify the Pb species. The analyzed sample-sets of size-fractionated aerosol particles are follows: Spring (Asian dust event), summer (two sample sets), fall, and winter (transboundary pollution event).

Lead species in size-fractionated aerosol particles are different between fine and coarse aerosol particles. In the fine aerosol particles, the dominant Pb species were two or three components, $PbSO_4$, $Pb(NO_3)_2$ and PbC_2O_4, in all seasons. That is, the seasonal variation of Pb species in the fine aerosol was not found. It is because the Pb species in the fine aerosol particles are formed by uniform chemical reaction with H_2SO_4, HNO_3 and oxalic acid in droplet through a year. Pb species in the coarse aerosol particles, except for on Asian dust event, were $2PbCO_3$-$Pb(OH)_2$, $Pb(NO_3)_2$ and PbC_2O_4. These $2PbCO_3$-$Pb(OH)_2$ and partial PbC_2O_4 were derived from road dust, and $Pb(NO_3)_2$ in coarse aerosol particles was formed by chemical reactions with HNO_3 on the surface of the particle. In Asian dust event, the coarse aerosol particles showed PbSiO3 as major Pb species, although other species, $2PbCO_3$-$Pb(OH)_2$ and PbC_2O_4, were also obtained. Thus we could found the clear seasonal variation in the coarse aerosol particles. These results will be able to estimate the accurate estimation of Pb solubility to the surface seawater.