

## Identification of Sources of Lead in the Atmosphere by its Speciation and Isotopic Composition

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### 1. Introduction

Recently, chemical reactions of major elements occurred in the atmosphere have been clarified, whereas those of trace metals have not. In particular, formation processes of Pb species, which is concerned to cause a health hazard, are still unclear. The identification of species and formation process of Pb is important to evaluate the human hazards. In addition, Pb species is expected to be used as a transboundary pollution tracer, because Pb species are different depending on each emission area. In this study, Pb species in size-fractionated aerosol sample were determined by XAFS spectroscopy together with the Pb isotope ratios to identify the formation mechanisms of Pb species in aerosol.

### 2. Sampling and Analysis Methods

Size-fractionated aerosol samples were collected by a high-volume cascade impactor in Higashi-Hiroshima. Sampling period was from 9th Oct. 2012 to 23th Oct. 2012 (2 weeks). Candidates of Pb source in atmosphere, fly ash of municipal solid incinerator (MSWI), heavy oil combustion, road dust and resuspension particles on the roof, were also collected. Weathered Hiroshima-granite, which is crustal material of the sampling area, was also collected as a possible natural Pb source. Lead species were determined by X-ray absorption fine structure (XAFS) spectroscopy. Heavy metals concentrations were measured by ICP-MS. Lead isotope ratios were determined by MC-ICP-MS with Tl doping technique after appropriate treatments.

### 3. Results and Discussion

Lead species were different between coarse and fine aerosol particles. Lead species in coarse aerosol particles were  $\text{PbC}_2\text{O}_4$ ,  $2\text{PbCO}_3\text{-Pb(OH)}_2$ , and  $\text{Pb(NO}_3)_2$ . Lead sources of coarse aerosol particles can be road dust because main Pb components in road dust were  $\text{PbC}_2\text{O}_4$  and  $2\text{PbCO}_3\text{-Pb(OH)}_2$ . This result was also suggested by EFs of Cu and Sb which are good indicators of road dust. Lead nitrate in coarse aerosol, which was not contained in road dust, might be formed by chemical reaction of natural Pb with gas-phase  $\text{HNO}_3$  in the atmosphere. In contrast, Pb species of fine aerosol particles were  $\text{PbC}_2\text{O}_4$ ,  $\text{PbSO}_4$ , and  $\text{Pb(NO}_3)_2$ . Major Pb sources in fine aerosol particles are fly ash of MSWI and heavy oil combustion based on the determination of Pb species in these materials examined in this study. This result was also supported by size-distributions of Cd, Ni and V.

In our presentation, the results of Pb isotopic composition will be discussed together with Pb species in aerosol samples.

Keywords: Aerosol, Lead species, Lead isotope, XAFS spectroscopy, MC-ICP-MS