

Sources of anthropogenic Pb in wet depositions collected in remote and urban areas in Japan

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Pb isotope ratios of wet deposition (snow and rain) collected from remote and urban areas in Japan were precisely determined to identify sources of anthropogenic aerosols. Samples were collected from 1 urban area (Niihama City, Ehime Prefecture), 3 suburb areas, and 5 remote mountain areas. Remote areas were more than 30 km from Niihama, whereas suburb areas were less than 30 km from Niihama. Snow was collected using a Ziploc^R freezer bag (Asahi Kasei) as soon as possible (within 24 h) after each snowfall event without melting and compaction. Rain was collected in a low-density polyethylene (LDPE) bottle using PE funnel and a PFA tube. Melted snow and rain were filtered through a membrane filter (pore size 0.45 μm). Solid phase extraction was conducted using a column of NOBIAS chelate PA-1 (Hitachi High-Technology) and an auto solid phase extractor (SPE-100, Hiranuma) to remove Tl. After sample introduction, 3 M nitric acid was passed through the column and Pb was collected. The eluent was evaporated and redissolved in 2% nitric acid to make a concentration of 1 μg/L of Pb. A double-focusing MC-ICP-MS (NEPTUNE, Thermo Fisher Sci.) with a desolvating nebulizer (Aridus IITM, Teledyne CETAC Technologies) was used for Pb isotope measurement. NIST SRM 981 Pb and SRM 997 Tl solutions were used as a Pb standard and a doping Tl standard for mass calibration.

Pb concentrations of rain were from 0.0020 to 0.408 μg/L, whereas those of snow were from 0.159 to 1.94 μg/L. The ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁶Pb and ²⁰⁸Pb/²⁰⁶Pb values of rain were 17.9766 18.2028, 0.085801 0.86759 and 2.06412 2.12250, respectively. The ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁶Pb and ²⁰⁸Pb/²⁰⁶Pb values of snow were 17.5897 18.1572, 0.85829 0.88399 and 2.09905 2.13902. The RSD of ²⁰⁶Pb/²⁰⁴Pb for rain was 0.009% at a concentration of 2 ng/L. According to the comparison of Pb isotope ratios for reference materials and atmospheric aerosols in China, Korea and Japan (Fig. 1), the sources of Pb of wet

depositions in remote areas were mainly long-range transport from the continent, whereas those in urban areas were mainly fly ash from waste incineration.

