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Origin of atmospheric lead around Omura Bay, Nagasaki, Japan

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Recently, air quality in Japan is suffering from pollution from other countries including China. Atmospheric heavy metals such as lead are revealed to originate from other countries (e.g., Mukai et al., 1999). North of the Kyushu Island, where Omura Bay is located, is an appropriate area to directly observe such cross-border air pollution. Aerosol particles were sampled with temporal high resolution from May 2011 to August 2012 on the eastern hill of Omura Bay. In order to assess the responsibility of the cross-border pollution for the anthropogenic metals in the air around this area, we measured the trace element concentration and Sr-Pb isotope ratios of 1M-HCl leachates and the residues of the aerosol samples. The residues can be considered as silicate mineral particles of natural origin, while the leachates represent soluble component of aerosol, such as mist, fume, and sea splay. The leachates contain 8-18 times the amount of Zn, Cd, and Pb that the silicates do. This suggests that these elements are of anthropogenic origin.

The $^{87}Sr/^{86}Sr$ of silicate components is high in winter and spring (0.712-0.714), and is lowest in Summer (0.706). These high and low ratios are typical of Asian dust and Japanese local sediment, respectively. The seasonal change is considered to reflect the difference of dominant wind direction between winter and summer. $^{87}Sr/^{86}Sr$ correlates with soluble Cd and Pb ($r>0.7$), but not with Zn ($r=0.28$). Anthropogenic Pb and Cd seem to increase their flux with the contribution of Asian dust to the mineral particles. However, the Pb isotope ratios of soluble component suggest that their origin is Japan in March and February ($^{206}Pb/^{207}Pb$: 1.16, $^{208}Pb/^{207}Pb$: 2.44), when much amount of Asian dust arrives, while the soluble Pb from September to December ($^{206}Pb/^{207}Pb$:1.13-1.15, $^{208}Pb/^{207}Pb$: 2.42-2.43), when the contribution of Asian dust to silicate is not significant, is suggested to have originated from Beijing, far-east Russia, or Central Asia. This indicates that the cross-border air pollution occurs not in spring when is the acme of Asian dust but in autumn and winter. Coal combustion for heating and wintry atmospheric pressure pattern may have acted synergistically. The reasons why the soluble Pb in spring has local origin, and why the highest Pb flux occurs in spring are presumed that 1) the significantly high altitude of the transportation path of Asian dust refused the influence of Chinese urban air, and 2) the large amount of dust particles had adsorbed and concentrated the lead on their surface from sea splays and Japanese air in crossing the Japan Sea and in settling long distance from the high altitude to Omura Bay.

In this study, we could not conclusively indicate the actual source of atmospheric lead. Lead isotope ratios are now widely used to estimate the origin of anthropogenic lead. However, citable $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ data of high precision are rare on the contrary to $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$, or $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$, perhaps because the study of atmospheric lead isotopes have been mainly conducted by using quadropole ICP-MS. Accumulation of $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ data of atmospheric lead sources is necessary for more accurate identification of the pollutant source.

Keywords: aerosol, anthropogenic lead, Sr-Pb isotope ratios, cross-border pollution