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Temporal variations of atmospheric lead isotope ($^{207}Pb/^{206}Pb$, $^{208}Pb/^{206}Pb$) as a source function for deep-water in the East/Japan Sea

SEONGHU CHOI, MAN-SIK CHOI AND DASOM YANG
Chungnam National University
Presenting Author: tmi_01389@naver.com

The period of deep-water formation can be constrained by confirming the temporal variation of stable Pb isotopes ($^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$) in the atmosphere, as dissolved Pb in seawater originates from atmospheric Pb. For this, we analyzed Pb concentrations and isotopes in aerosol samples collected in Ulleung island from October 2003 to October 2008, and collected atmospheric Pb data from the surrounding areas of the East/Japan Sea to examine changes in atmospheric Pb characteristics over about 30 years.

Atmospheric Pb concentrations showed seasonal variations, with higher values observed in winter (23.7 ng/m^3) and spring (22.8 ng/m^3) compared to summer (12.4 ng/m^3) and autumn (13.7 ng/m^3), while EF values were lower in spring (56.6) than in other seasons (103-126). However, there were no significant seasonal differences in Pb isotope ratios.

Annual variations in atmospheric Pb characteristics were observed, particularly in Pb isotopes ($^{207}Pb/^{206}Pb$, $^{208}Pb/^{206}Pb$; mean ± SD), from 2003 (0.880 ± 0.010, 2.144 ± 0.019) to 2008 (0.864 ± 0.007, 2.107 ± 0.015). The dominant air mass flow throughout the sampling period was from northern China, as suggested by the back-trajectories of the air masses and cluster analysis (HYSPLIT, NOAA). This suggests that changes in Pb isotope ratios in the source area have led to changes in atmospheric Pb isotopes in the study area.

The data on Pb isotope ratios, combined with data from around the East/Japan Sea, confirm a smooth variation over a period of about 30 years. The Pb isotope ratios increased until the early 1990s and decreased from the early 2000s to the late 2010s. This annual variation reflects the steady changes in atmospheric Pb sources in China. This can occur for two reasons: 1) atmospheric Pb emitted from leaded gasoline in the past was deposited and subsequently re-suspended and mixed with current coal Pb, or 2) the proportion of imported coal used in China increased, resulting in a mixture of Pb with different values. In addition, changes in atmospheric Pb isotope ratios over time can be used to determine when deep water formed.