



Nitrogen and Triple Oxygen Isotopic Analyses of Atmospheric Particulate Nitrate over the Pacific Ocean

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Nitrate plays a significant role in the biogeochemical cycle. Atmospheric nitrate (NO_3^- and HNO_3) are produced by reaction precursor as NO_x (NO and NO_2) emitted by combustion, biomass burning, lightning, and soil emission, with atmospheric oxidants like ozone (O_3), hydroxyl radical (OH), peroxy radical and halogen oxides. Recently, industrial activity lead to increases in the concentrations of nitrogen species (NO_x and NH_y) throughout the environment. Because of the increase of the amount of atmospheric nitrogen deposition, the oceanic biogeochemical cycle are changed (Galloway et al., 2004; Kim et al., 2011). However, the sources and formation pathways of atmospheric nitrate are still uncertain over the Pacific Ocean because the long-term observation is limited.

Stable isotope analysis is useful tool to gain information of sources, sinks and formation pathways. The nitrogen stable isotopic composition ($\delta^{15}\text{N}$) of atmospheric particulate NO_3^- can be used to posses information of its nitrogen sources (Elliott et al., 2007). Triple oxygen isotopic compositions ($\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$) of atmospheric particulate NO_3^- can be used as tracer of the relative importance of mass-independent oxygen bearing species (e.g. O_3 , BrO ; $\Delta^{17}\text{O} \neq 0 \text{‰}$) and mass-dependent oxygen bearing species (e.g. OH radical; $\Delta^{17}\text{O} \approx 0 \text{‰}$) through the formation processes from NO_x to NO_3^- in the atmosphere (Michalski et al., 2003; Thiemens, 2006).

Here, we present the isotopic compositions of atmospheric particulate NO_3^- samples collected over the Pacific Ocean from 40°S to 68°N . We observed significantly low $\delta^{15}\text{N}$ values for atmospheric particulate NO_3^- on equatorial Pacific Ocean during both cruises. Although the data is limited, combination analysis of $\delta^{15}\text{N}$ and $\Delta^{17}\text{O}$ values for atmospheric particulate NO_3^- showed the possibility of the main nitrogen source of atmospheric particulate NO_3^- on equatorial Pacific Ocean is ammonia oxidation in troposphere. Furthermore, the $\Delta^{17}\text{O}$ values for atmospheric particulate NO_3^- originated from Central North Pacific Ocean showed the possibility of atmospheric particulate NO_3^- formation pathway is reaction NO_2 with halogen oxides. At the presentation, we discuss the sources and formation pathways of atmospheric particulate NO_3^- by using stable isotopic analyses for each region.

References

- J. N. Galloway, A. R. Townsend, J. Willem Erisman, M. Bekunda, Z. Cai, J. R. Freney, L. A. Martinelli, S. P. Seitzinger, M. A. Sutton, *Biogeochemistry*, 70, 153–226, 2004
- T. W. Kim, K. Lee, R. G. Najjar, H.D. Jeong, H. J. Jeong, *Science* 334, 505–509, 2011
- E. M. Elliott, C. Kendall, S. D. Wankel, D. A. Burns, E. W. Boyer, K. Harlin, D. J. Bain, and T. J. Butler, *Environ. Sci. Technol.*, 41, 7661-7667, 2007
- G. Michalski, Z. Scott, M. Kabling, and M. H. Thiemens, *Geophys. Res. Lett.*, 30, 1870, 2003
- M. H. Thiemens, *Annu. Rev. Earth Planet. Sc.*, 34, 217-262, 2006