1.149 Influence of urbanization on the sources of atmospheric nitrate; Evidence from the triple oxygen isotopes of nitrate in dry and wet deposition.

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Abstract:

Atmospheric nitrate deposition resulting from anthropogenic activities negatively affects human and environmental health. Identifying deposited nitrate that is produced locally versus that originating from long-distance transport would help inform efforts to mitigate such impacts. However, distinguishing the relative transport distances of atmospheric nitrate in urban areas remains a major challenge since it may be produced locally and/or come from upwind regions. To address this uncertainty, we assessed spatiotemporal variation in monthly weighted-average triple oxygen isotopic compositions of wet and dry nitrate deposition during one year at urban and rural sites along the western coast of the northern Japanese island of Hokkaido, downwind of the East Asian continent. Triple oxygen isotopic compositions of nitrate in wet deposition at the urban site mirrored those of wet and dry deposition at the rural site, ranging between +23 and +31 permil with higher values during winter and lower values in summer, which suggests greater relative importance of oxidation of nitrogen dioxide by ozone during winter and hydroxyl radicals during summer. In contrast, triple oxygen isotopic compositions of nitrate in dry deposition at the urban site were lower (range from +19 to +25 permil) and displayed less distinct seasonal variation. These results suggest that, relative to nitrate in wet and dry deposition in rural environments and wet deposition in urban environments, nitrate in

dry deposition in urban environments forms from relatively greater oxidation of nitrogen monoxide by peroxy radicals and/or oxidation of nitrogen dioxide by hydroxyl radical. Given greater concentrations of peroxy radicals and hydroxyl radical in cities, these results imply that dry nitrate deposition results from local NOx emissions more so than wet deposition, which is transported longer distances. These results illustrate the value of stable isotope data for distinguishing the transport distances and reaction pathways of atmospheric nitrate pollution.