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# Quantifying the isotopic composition of $NO_x$ emission sources: An analysis of collection methods

D. Fibiger (1) and M. Hastings (2)

(1) Department of Chemistry, Brown University, Providence, RI, United States (dorothy_fibiger@brown.edu), (2) Department of Geological Sciences and Environemental Change Initiative, Brown University, Providence, RI, United States

We analyze various collection methods for nitrogen oxides, $NO_x$ ($NO_2$ and NO), used to evaluate the nitrogen isotopic composition ($\delta^{15}$N). Atmospheric $NO_x$ is a major contributor to acid rain deposition upon its conversion to nitric acid; it also plays a significant role in determining air quality through the production of tropospheric ozone. $NO_x$ is released by both anthropogenic (fossil fuel combustion, biomass burning, aircraft emissions) and natural (lightning, biogenic production in soils) sources. Global concentrations of $NO_x$ are rising because of increased anthropogenic emissions, while natural source emissions also contribute significantly to the global $NO_x$ burden. The contributions of both natural and anthropogenic sources and their considerable variability in space and time make it difficult to attribute local $NO_x$ concentrations (and, thus, nitric acid) to a particular source. Several recent studies suggest that variability in the isotopic composition of nitric acid deposition is related to variability in the isotopic signatures of $NO_x$ emission sources. Nevertheless, the isotopic composition of most $NO_x$ sources has not been thoroughly constrained. Ultimately, the direct capture and quantification of the nitrogen isotopic signatures of $NO_x$ sources will allow for the tracing of $NO_x$ emissions sources and their impact on environmental quality. Moreover, this will provide a new means by which to verify emissions estimates and atmospheric models.

We present laboratory results of methods used for capturing $NO_x$ from air into solution. A variety of methods have been used in field studies, but no independent laboratory verification of the efficiencies of these methods has been performed. When analyzing isotopic composition, it is important that $NO_x$ be collected quantitatively or the possibility of fractionation must be constrained. We have found that collection efficiency can vary widely under different conditions in the laboratory and fractionation does not vary predictably with collection efficiency. For example, prior measurements frequently utilized triethanolamine solution for collecting $NO_x$, but the collection efficiency was found to drop quickly as the solution aged. The most promising method tested is a $NaOH/KMnO_4$ solution (Margeson and Knoll, *Anal. Chem.*, 1985) which can collect $NO_x$ quantitatively from the air. Laboratory tests of previously used methods, along with progress toward creating a suitable and verifiable field deployable collection method will be presented.