6.106 Quantifying isotopic signatures of atmospheric NOx emissions.

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Abstract:

Atmospheric nitrogen oxides (NO_x = NO + NO_2) play key roles in atmospheric chemistry, air quality, and radiative forcing, and contribute to nitric acid deposition. Given the variety of sources of NO_x (both natural and anthropogenic), their variability in space and time, and the relatively short lifetime of NO_x, it is difficult to directly link variability in NO_x concentrations or nitric acid deposition with emissions sources. NO_x isotopic signatures offer a potentially valuable tool to trace source impacts on atmospheric chemistry and regional acid deposition. However, previous work on NO_x isotopic signatures suggests large ranges in values, even from the same emission source, as well as overlapping ranges amongst different sources. These prior measurements have utilized a variety of methods for collection of NO_x, and recent tests reveal inconsistencies in efficiency of collection, as well as issues related to changes in conditions such as humidity, temperature, and NO_x fluxes. Our recently developed method accurately quantifies the nitrogen isotopic composition ($\delta^{15}N$) of NO_x (NO_x = NO + NO_2) after capturing the NO_x in solution as nitrate. The method has been thoroughly laboratory and field tested, and efficiently collects NO and NO_2 under a variety of conditions. This new method is used to quantify the isotopic composition of NO_x associated with different emission sources, including vehicles, microbial processes in agricultural soils, and biomass burning. The NO_x collection system is optimized to allow for short collections in high concentration plumes, as well as under sporadic pulse-oriented fluxes such those associated with soil emissions. Using a consistent method, we test whether it is possible to distinguish the isotopic ranges associated with different emission sources, and compare and contrast laboratory-based and field-based collections.