Pilot study measuring N_2O mole fraction, $\delta^{15}N^{bulk}$-N_2O, $\delta^{15}N^{\alpha}$-N_2O, and $\delta^{15}N^{\beta}$-N_2O using Picarro G-5101i instrument reveals analytical challenges

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Nitrous oxide (N_2O) continues to increase in the atmosphere and ranks the 3rd in long-lived greenhouse gas in radiative forcing (behind CH_4 and CO_2). It is also the dominant ozone-depleting substance emitted in the 21st century. While there are many challenges to understanding N_2O in the free troposphere, the NOAA global flask network offers insights. For example, variations in the N_2O mole fraction measured at NOAA sites are thought to be influenced in part by heightened microbial production from fertilized agricultural systems. Detrended monthly data display large interannual variability, which at some sites challenges the concept of a "mean" seasonal cycle. But soil microorganism processes are spatiotemporally heterogeneous, limiting our ability to constrain the anthropogenic influence on N_2O production at a global scale. The intramolecular position of ^{15}N (β position $^{15}N^{14}N^{16}O$ versus α position $^{14}N^{15}N^{16}O$) in addition to $\delta^{15}N^{bulk}$-N_2O can aid in our understanding of both the biological controls and stratospheric influence of tropospheric N_2O.

Laser based mid-infrared instruments are now available that can measure position specific isotopes of N_2O. The INSTAAR Stable Isotope Lab is testing the capability of the Picarro G-5101i instrument to measure potential small changes in position specific N_2O isotopes at a subset of the NOAA flask network sites. Simultaneous and continuous measurement of N_2O mole fraction, $\delta^{15}N^{bulk}$-N_2O, $\delta^{15}N^{\alpha}$-N_2O, and $\delta^{15}N^{\beta}$-N_2O is performed using the Picarro G5101-*i* wavelength-scanned cavity ring-down spectrometer. The work builds upon that of Steiker (2014) and now incorporates an automated flask manifold along with a water trap and scrubbers for both carbon monoxide and carbon dioxide to eliminate spectroscopic influence. Preliminary results suggest that instrument stability is critical for making measurements that are precise enough to constrain interannual variability at the NOAA baseline sides, specifically to resolve site preference differences. It is still unclear whether or not tropospheric $\delta^{15}N^{bulk}$ or SP exhibits a season cycle, but if the technique can be perfected, monitoring these isotopomers over a longer decadal time scale could provide answers to questions concerning large scale shifts in source emissions, or help to quantify the stratospheric influence of seasonal N_2O variability, which is essential in order to refine the global N_2O budget.

References

Steiker, Amy E. Global Measurement of Nitrous Oxide Isotopomers using Caving Ring-down Spectroscopy, Masters thesis, Department of Ecology and Evolutionary Biology, University of Colorado, Boulder. 2014.