

Using Isotopic Fingerprints to Trace Nitrous Oxide in the Atmosphere

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Nitrous oxide (N₂O) is a potent greenhouse gas and the strongest ozone-destroying substance emitted this century. Reliable predictions of future emissions, requires knowledge of the responsible N₂O source processes. Isotopic composition of N₂O is a tracer to distinguish between different emission pathways, as well as constraining the stratospheric N₂O sink. The four most abundant N₂O isotopic species are: ¹⁴N¹⁴N¹⁶O (99%), ¹⁴N¹⁵N¹⁶O (α , 0.4%), ¹⁵N¹⁴N¹⁶O (β , 0.4%) and ¹⁴N¹⁴N¹⁸O (0.2%). Due to its asymmetric molecular structure, N₂O (α) and N₂O (β) differ only in the position of the ¹⁵N atom, and the difference in their abundance – known as site preference (SP) – is a particularly powerful indicator for different N₂O production mechanisms.

Here we illustrate the potential of laser spectroscopy for real-time, high-precision analysis of isotopic composition in ambient N₂O. Furthermore, we present applications, in agricultural as well as suburban environments, illustrating the advantage and necessity of real-time data of trace gas isotope ratios. In an extensive campaign above a managed grassland, nitrifier-denitrification and denitrification were identified as prevalent sources of N₂O and variations in isotopic composition were attributed to alterations in the extent to which N₂O was reduced to N₂ [1]. In an ongoing project, we validate the real-time N₂O isotope data against a process-based biogeochemical soil model (DNDC) with an isotope sub-module (SIMONE), which is based on published isotope effects [2]. At a suburban site, the isotopic composition of atmospheric N₂O was monitored over 18 months to determine the source isotopic composition, which varied significantly compared to chemical and meteorological parameters. FLEXPART-COSMO transport modelling in combination with modified EDGAR inventory emissions was able to capture variability in N₂O mole fraction well, but simulations of isotopic composition showed little agreement with observations, indicating that the range of literature values of isotopic source signatures significantly underestimates the true variability [3].

In summary, we are convinced that real-time analysis of N₂O isotopic composition is an efficient approach to disentangle N₂O source / sink processes in agricultural as well as suburban / industrial environments. Combination of point measurements with modelling approaches provides spatial resolution and enables validation of emission inventories.

References

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