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Site selective on-site measurements of ambient N2O isotopomers by laser spectroscopy

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The intramolecular distribution of ^{15}N in N_2O can be used to obtain important information on the geochemical cycle of N_2O because many biological and chemical processes have distinct isotopic signatures. N_2O is a linear, non-symmetric molecule (N–N–O), with one nitrogen atom at the centre (α site) and one at the end (β site). Therefore, one can distinguish between two structural isomers containing one heavy isotope of nitrogen, namely $^{14}N^{15}N^{16}O$ and $^{15}N^{14}N^{16}O$, referred to as $^{15}N^{\alpha}$ and $^{15}N^{\beta}$, respectively.

Until recently, N_2O isotopomer studies at ambient concentrations were strongly limited by the lack of suitable analytical techniques. The standard methodology, laboratory-based isotope-ratio mass-spectrometry (IRMS), requires flask sampling and thus significantly constrains the ability to characterize the variations in N_2O isotopic species at time-scales relevant to environmental processes. Furthermore, isotopomers such as $^{14}N^{15}N^{16}O$ and $^{15}N^{14}N^{16}O$ have the same mass, making their quantification by IRMS a challenging and costly task, currently feasible by only very few laboratories.

We have developed a field-deployable quantum cascade laser absorption spectrometer (QCLAS) with a precision of 0.2 ‰ (120 s Allan minimum) for the site-specific isotopic ratios $\delta^{15}N^{\alpha}$ and $\delta^{15}N^{\beta}$ at 90 ppm N_2O [1]. This instrument allows the continuous determination of N_2O in enriched samples with ppm N_2O concentrations from combustion engines and microbial processes, such as waste water treatment.

To enable high precision N_2O isotopomer analysis at ambient concentrations, we designed a liquid nitrogen-free preconcentration unit. This automated unit achieves a 200 fold preconcentration in 20 minutes, quantitative N_2O recovery of $> 99\%$ and no measurable isotopic fractionation or interference from other atmospheric constituents [2].

The coupling of preconcentration and QCLAS allows characterizing the site-specific isotopic signature of N_2O source and sinking processes in various ecosystems under changing environmental conditions. N_2O isotopic signatures may then be employed to quantitatively apportion N_2O production on a local, regional or even global scale.

[1] H. Waechter, J. Mohn, B. Tuzson, L. Emmenegger, M.W. Sigrist, "Determination of N_2O isotopomers with quantum cascade laser based absorption spectroscopy", Opt. Express 16, pp. 9239-9244 (2008).

[2] J. Mohn, C. Guggenheim, B. Tuzson, M. K. Vollmer and L. Emmenegger, A liquid nitrogen-free preconcentration unit for measurements of ambient N_2O isotopomers by QCLAS, Atmos. Meas. Tech. Discuss. 2, pp. 3099-3127 (2010).