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Closed-loop 15N measurement of N2O and its isotopomers for real-time greenhouse gas tracing

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Quantifying sources of nitrous oxide is essential to improve understanding of the global N cycle and to develop climate-smart agriculture, as N_2O has a global warming potential 300 times higher than CO_2. The isotopic signature and the intramolecular distribution (site preference) of ^{15}N are powerful tools to trace N_2O, but the application of these methods is limited as conventional methods cannot provide continuous and *in situ* data. Here we present a method for closed-loop, real time monitoring of the N_2O flux, the isotopic signature and the intramolecular distribution of ^{15}N by using off-axis integrated cavity output spectroscopy (ICOS, Los Gatos Research). The developed method was applied to a fertilizer inhibitor experiment, in which N_2O emissions were measured on undisturbed soil cores for three weeks. The treatments consisted of enriched urea-N (100 kg urea-N/ha), the same fertilizer combined with the nitrification inhibitor nitrapyrin (375 g/100 kg urea), and control cores. Monitoring the isotopic signature makes it possible to distinguish emissions from soil and fertilizer. Characterization of site preference could additionally provide a tool to identify different microbial processes leading to N_2O emissions. Furthermore, the closed-loop approach enables direct measurement on site and does not require removal of CO_2 and H_2O. Results showed that 75% of total N_2O emissions (total=11 346 μg N_2O-N/m^2) in the fertilized cores originated from fertilizer, while only 55% of total emissions (total=2 450 μg N_2ON/m^2) stemmed from fertilizer for the cores treated with nitrapyrin. In the controls, N_2O derived from soil was only 40% of the size of the corresponding pool from the fertilized cores, pointing towards a priming effect on the microbial community from the fertilizer and demonstrating the bias that could be introduced by relying on non-treated cores to estimate soil emission rates, rather than using the isotopic signature. The site preference increased linearly over time for the cores with fertilizer and those with nitrapyrin, but the increase was stronger for the fertilized cores: during the first 10 days of the experiment, theses cores showed a more negative site preference than the cores with inhibitor, while during the last 10 days, the site preference for the fertilized cores was more positive than that of the inhibitor. This change indicates that the site preference of ^{15}N can be used to distinguish the processes of nitrification and denitrification, the former having been supressed by nitrapyrin in the cores treated with the inhibitor. Low enrichment levels (5% atomic excess in this study) sufficed in order to separate emissions from soil and fertilizer, making the proposed closed-loop approach a cost-effective and practical tool to obtain a continuous, *in situ* characterization of N_2O sources.