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Isotopomer signatures of N_2O emitted from an arable loess soil under different process conditions - a soil microcosm study.

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Soils represent the major source of the atmospheric greenhouse gas nitrous oxide (N_2O) and there is a need to better constrain the total global flux and the relative contribution of the microbial source processes. The aim of our study was to evaluate isotopomer analysis of N_2O (intramolecular distribution of ^{15}N) as well as conventional nitrogen and oxygen isotope ratios (i) as a tool to identify N_2O production processes in soils and (ii) to constrain the isotopic fingerprint of soil-derived N_2O.

We conducted three microcosm experiments with arable loess soil fertilized with $^{15}NO_3^-$-labeled or non-labeled ammonium nitrate. In experiment 1, soils were incubated at varying moisture (55, 75 and 85% water-filled pores pace) in order to establish different levels of nitrification and denitrification. Isotopomeric characterization of emitted N_2O was conducted by mass spectrometric analysis of $\delta^{18}O$, average $\delta^{15}N$ ($\delta^{15}N^{bulk}$) and ^{15}N site preference (SP = difference in $\delta^{15}N$ between the central and peripheral N positions of the asymmetric N_2O molecule). Total rates and N_2O emission of denitrification and nitrification were determined by ^{15}N-analysis of headspace gases and soil extracts of the $^{15}NO_3^-$ treatment. N_2O emission and denitrification increased with moisture whereas gross nitrification was almost constant. In the dry treatment, more than half of the N_2O flux was derived from nitrification, whereas denitrification was the dominant N_2O source in the intermediate and wet treatments. Moisture conditions were clearly reflected by the isotopic signatures since highly significant differences were observed for average $\delta^{15}N^{bulk}$, SP and $\delta^{18}O$. Experiment means of the intermediate and wet treatments gave negative $\delta^{15}N^{bulk}$ (-18.0 and -34.8 permil,

respectively) and positive SP (8.6 and 15.3 permil, respectively), which we explained by the fractionation during N_2O production and partial reduction to N_2. In the dry treatment, mean SP was relatively low (1.9 permil) which suggests that nitrification produced N_2O with low or negative SP. The clear influence of process condition on isotopomer signatures suggests that the isotopomer approach might be suitable for identifying N_2O source processes. However, more research is needed to determine the impact from process intensity and microbial community structure. Isotopomer signatures were within the range reported from previous soil studies which supports the assumption that SP of soil derived N_2O is lower than SP of tropospheric N_2O. Experiments 2 and 3 were conducted to study the NO_3^--to-N_2O step and the N_2O-to-N_2 step of denitrification separately. SP of N_2O produced during anaerobic incubation of NO_3^- amended loess soil in the presence of 10 kPa acetylene (experiment 2) ranged between 20 and 35 permil. Anaerobic incubation of NO_3^- depleted loess soil supplemented with N_2O (experiment 3) resulted in increasing SP in the residual N_2O while N_2O was reduced to N_2. From experiments 2 and 3 we conclude that positive SP of N_2O emitted in experiment 1 is the result of both partial processes, i.e. production and reduction of N_2O.