

## Quantifying gross $N_2O$ flux and production using ${}^{15}N_2O$ pool dilution technique and direct gas-flow core method

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Soils are not only a major source but also a potential sink for atmospheric nitrous oxide  $(N_2O)$ , a potent greenhouse gas and the most important substance for stratospheric ozone depletion. Net N<sub>2</sub>O flux at the soil-atmosphere interface is the balance of simultaneously occurring gross  $N_2O$  production and consumption.  $N_2O$  is consumed via reduction to  $N_2$ , i. e. the terminal product of the denitrification process, which is difficult to measure against the high atmosphere background. The enigmatic lack of measurements on gross  $N_2O$  flux or  $N_2$  production still impedes our understanding of the controls on soil N<sub>2</sub>O emissions and the closure of the global nitrogen cycle. Here, we combined the <sup>15</sup>N<sub>2</sub>O pool dilution technique and direct gas-flow core method to disentangle 1) gross N<sub>2</sub>O fluxes at the soil-atmosphere interface, and 2) gross  $N_2O$  production and consumption in the soil. The  $^{15}N_2O$  pool dilution method entails adding <sup>15</sup>N<sub>2</sub>O to the chamber headspace, measuring <sup>14</sup>N<sub>2</sub>O and <sup>15</sup>N<sub>2</sub>O concentrations and applying a model to simultaneously solve for gross N<sub>2</sub>O flux and consumption rate at the soil-atmosphere interface. The direct gas-flow core method substitutes the soil air and chamber headspace with helium to a nearly  $N_2$ -free atmosphere in order to directly measure both N<sub>2</sub>O and N<sub>2</sub>fluxes; N<sub>2</sub> flux is the gross N<sub>2</sub>O consumption and its sum with  $N_2O$  flux is the gross  $N_2O$  production in the soil. Soil samples were taken from grassland, cropland, beech and pine forest soils, representing a broad range of land uses and soil types. Additionally, we compared measurements from intact soil cores (reflecting inherent soil bulk density and porosity) and sieved soils (eliminating heterogeneity in porosity). Gross N<sub>2</sub>O production rate in the soil was highest in the silty grassland soil (41.04 $\pm$ 4.6  $\mu$ g N kg<sup>-1</sup>  $h^{-1}$ ) and lowest in the sandy pine forest soil (1.84±1.82 µg N kg<sup>-1</sup> h<sup>-1</sup>). The intact soil cores and sieved soils showed similar trends. Gross N<sub>2</sub>O production rates in the soil exceeded gross N<sub>2</sub>O fluxes at the soil-atmosphere interface by at least an order of magnitude, suggesting that most of the N<sub>2</sub>O produced is possibly directly consumed and diffused as  $N_2$ . The gross  $N_2O$  consumption rate at the soil-atmosphere interface only accounted for 7% of  $N_2$ production in the soil, suggesting that  $N_2O$  in the soil air that is diffusing to the atmosphere is seldom consumed. Gross N<sub>2</sub>O fluxes at the soil-atmosphere interface, gross N<sub>2</sub>O production in the soil and N<sub>2</sub> production were all significantly correlated with soil water content,  $NH_4^+$ , dissolved organic C, microbial biomass C and N (p < 0.05). The fraction of gross  $N_2O$  consumption at the soil-atmosphere interface to  $N_2$  production in the soil increased with decreasing soil water content (p=0.055). Overall, this study shows that gross soil N<sub>2</sub>O reduction to N<sub>2</sub> within microbial cells and/or soil microsites is of paramount importance for the regulation and avoidance of soil N<sub>2</sub>O losses and that gross  $N_2O$  consumption at the soil-atmosphere interface is contributing to only a small part of  $N_2$ production.