Determination on the Triple Oxygen Isotopic Composition of Nitrous Oxide Emitted from Soils

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Nitrous oxide (N_2O) is a long-lived trace gas playing important roles in the atmosphere, such as stratospheric ozone depletion and global warming. The production of N_2O is closely related to multiple microbial processes in terrestrial ecosystems, in which N_2O emissions from both agricultural and natural soils represent 50 - 70% of total emission. The stable isotopic compositions of N_2O ($\delta^{15}N$ and $\delta^{18}O$) have been widely used as tracers to identify the production pathways of N_2O to establish strategies reducing N_2O emission from anthropogenic sources traditionally. However, possible progress of the microbial isotope fractionation processes in the heterogenous soil ecosystems, often made it difficult to identify the production pathways of N_2O. Based on the stability during the microbial processes, we proposed to use the $\Delta^{17}O$ of N_2O as an additional, more robust tracer to clarify the potential microbial production pathways of N_2O emitted from soil environments.

N_2O produced via nitrification pathway has oxygen atom originates from atmospheric oxygen molecules (O_2: $\Delta^{17}O = -0.4$ ‰) while the oxygen atom of N_2O produced via denitrification pathway originates from nitrate (NO_3^-: $\Delta^{17}O > 0$ ‰) or water (H_2O: $\Delta^{17}O = 0$ ‰). Therefore, it is expected that the N_2O production pathways could be elucidated from the $\Delta^{17}O$ value. In this study, we quantified the $\Delta^{17}O$ of N_2O emitted from soils, the forest areas in Gifu and Aichi prefectures and the tea plantations in Mie prefecture, to determine the contribution (mixing ratio) of the nitrification and denitrification. In the forest area of Gifu prefecture, we collected soil N_2O accumulated in snow. In Aichi and Mie prefectures, N_2O emitted from soils were collected using an open-flow chamber. In addition, in order to confirm the effect of atmospheric NO_3^-($\Delta^{17}O = 26.3 \pm 3$ ‰) on the N_2O production in soil ecosystems during the raining events, we also performed chamber experiments both during and after the natural raining events.

The $\Delta^{17}O$ values of N_2O emitted from the forested soils, both in Gifu and Aichi prefecture, were -0.4 ‰, implying that oxygen atom in N_2O originates from atmospheric O_2 via microbial nitrification. On the other hand, the $\Delta^{17}O$ value of N_2O emitted from tea plantation field was -0.2‰, implying that the oxygen atom in N_2O derives from O_2, H_2O and soil NO_3^-($\Delta^{17}O = +0.5$ ‰) via both nitrification and denitrification. The $\Delta^{17}O$ value of N_2O emitted from forested soils in campus (Aichi prefecture) during the natural raining days reached up to +5 ‰, implying that all the oxygen atoms in N_2O were derived from NO_3^-, and was probably derived directly from part of the atmospheric NO_3^-via denitrification in the surface soils. We concluded that the main microbial processes to produce N_2O within the soil ecosystems have been changed from nitrification to denitrification during the raining events.

Keywords: Nitrous Oxide, Triple Oxygen Isotopic Composition, Nitrification, Denitrification, Soil

