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Quantifying nitrate dynamics in hydrosphere using the triple oxygen isotopes as tracers

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In most studies that have been conducted to date, the gross uptake rate of nitrate has been estimated by incubation experiments using ^{15}N tracer techniques. In this conventional approach, ^{15}N labeled NO_3^- is added into bottles or mesocosms that simulate in situ conditions, which leads to the production of particulate organic-^{15}N ($PO^{15}N$) through assimilation over a known incubation period of several hours to several days. This $PO^{15}N$ is then gathered and quantified by mass spectrometry. However, these experimental procedures are generally costly, complicated, and time consuming, especially those that employ mesocosms.

Furthermore, the ^{15}N tracer method has several problems with determining accurate nitrate uptake rates. For example, the usual ^{15}N tracer method does not include assimilated nitrogen released to dissolved organic-^{15}N ($DO^{15}N$) during incubation within the estimated uptake rates, which results in the rates being underestimated. Additionally, incubation itself could also result in the production of artifacts by changing the physical/chemical environments. Finally, it is difficult to simulate nitrate uptake by periphyton or microbes on the lake floor through typical incubation in bottles.

The gross nitrate uptake rate estimated using the triple oxygen isotopic compositions of NO_3^- overcomes the aforementioned problems inherent in the conventional ^{15}N-labelled tracer methods. Accordingly, the triple oxygen isotope method can be an alternative to standard techniques for the determination of accurate gross nitrate uptake rates in hydrosphere that contain detectable quantities of atmospheric nitrate within the total nitrate.

In this study, we applied this the triple oxygen isotope method in the water column of Lake Mashu, Japan, which is a crater lake with a depth of 211 m. To quantify the geochemical dynamics of nitrate in the oligotrophic environment, the stable isotopic compositions of nitrate, including the ^{17}O anomalies, were determined twice in one year (June and August 2007) in the lake.

While the total inventory of nitrate in the lake water decreased from 4.2 to 2.1 Mmol (Mmol = 10^6 mol) during the period between the observations, the average triple oxygen isotopic compositions of nitrate were uniform at +2.5 permil, which corresponded to an average mixing ratio of atmospheric nitrate to total nitrate of 9.7 +- 0.8%. Using the total mass of the atmospheric nitrate deposited onto the entire catchment area of the lake during a period of 2 months (0.047 Mmol), we estimated that 0.52 +- 0.34 Mmol of the remineralized nitrate was fed into the water column through nitrification, while 2.6 +- 0.4 Mmol of nitrate was simultaneously removed from the water column by assimilation. The lake water dissolved nitrate was characterized by rapid removal through assimilation during summer until it was almost completely removed from the euphotic layer, as well as continuous feeding into the lake through nitrification and deposition, regardless of the seasons, which corresponds to a gross annual flux of 3.2 +- 0.3 Mmol yr^{-1} and 0.35 +- 0.2 Mmol yr^{-1}, respectively. These results indicate that atmospheric nitrate deposited into the lake will be assimilated quickly having a mean residence time of 1.2 +- 0.1 years. Besides, more than 90% of the assimilated nitrate will be remineralized to nitrate and assimilated again via active nitrogen cycling in the lake.

Keywords: nitrate, atmospheric deposition, assimilation rate, nitrification rate, nitrogen cycle, triple oxygen isotopes