Tracing Nitrogen Cycles in Natural Waters using the Triple Oxygen Isotopic Compositions as Tracers

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The ^{17}O anomaly ($D^{17}O$) in nitrate can be used to quantify atmospheric nitrate inputs into terrestrial and aquatic nitrate reservoirs, while the remineralized nitrate produced through nitrification reactions shows usual mass-dependent isotopic signatures ($D^{17}O$), atmospheric nitrate exhibit ^{17}O anomaly around $D^{17}O$ = +25 per mil. To verify this hypothesis, we determined the $D^{17}O$ values of nitrate in Lake Mashu in June and August in 2007 as the part of the project of GEMS/Water. Lake Mashu is a famous subalpine, oligotrophic lake as one of the most transparent lake in the world. It is suitable experimental field to quantify atmospheric nitrate input, as there is no river flowing either into it or out of it.

To determine the $D^{17}O$ in nitrate, we developed a rapid, sensitive, and automated analytical system to determine the $d^{15}N$, $d^{18}O$, and $D^{17}O$ values of nitrate in nanomolar quantities by continuous-flow isotope ratio mass spectrometry without any cumbersome and time-consuming pretreatments. Our method was based on the isotopic analysis of nitrous oxide quantitatively converted from nitrate based on the simple reactions using spongy cadmium and sodium azide in an acetic acid buffer (McIlvin and Altabet, 2005). To place the isotopic analyses on the Vienna Standard Mean Ocean Water (VSMOW) scale, IAEA-NO-3 was used, as it was the most frequently used and best characterized international nitrate reference material. To ascertain the isotope scale linearity, the additional reference materials USGS-34 and USGS-35 were used as well, as they had a larger difference in both ^{17}O and ^{18}O content.

As a result, we found the $D^{17}O$ values of the nitrate in Lake Mashu to be around +3.0 per mil. Using the $D^{17}O$ values of nitrate, we could estimate the atmospheric nitrate inputs into Lake Mashu to be about 10-15%. Besides, we clarified that 90% of the nitrate in Lake Mashu were derived from the remineralized nitrate produced through nitrification reactions.