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Tracing the source and fate of nitrate in groundwater using ^{15}N, ^{17}O and ^{18}O

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The stable isotopic compositions of nitrate in precipitation (wet deposition) and groundwater were determined for Toyama prefecture, Japan, so as to use the ^{17}O anomalies ($D^{17}O$) to trace the fate of atmospheric nitrate that had deposited onto the groundwater catchments. Both $d^{15}N$ and $d^{18}O$ of nitrate were also used together with $D^{17}O$ to trace the non-atmospheric sources of nitrate in the groundwater. Wet deposition (precipitation) samples were taken every week at the Kosugi Station of Toyama Prefectural Environmental Science Research Center from April 2010 to March 2011. Groundwater samples were collected at 47 sites, mostly located on Kurobe, Joganji or Shougawa alluvial fan, in Toyama prefecture. Samples were filtered through a 0.2 micro meter pore-size membrane filter and stored in a refrigerator until analysis. To determine the stable isotopic compositions of nitrate, the sample nitrate was chemically converted to nitrous oxide using a method originally developed for $^{15}N/^{14}N$ and $^{18}O/^{16}O$ isotope ratios of seawater and freshwater nitrate (McIlvin and Altabet, 2005) with slight modification (Tsunogai et al. 2008). The stable isotopic compositions of nitrous oxide were determined using our Continuous-Flow Isotope Ratio Mass Spectrometry system (Komatsu et al. 2008). Concentration of nitrate in ground water samples varied widely from less than 0.1uM to more than 100uM due to the difference in biological activity among the groundwater catchments. All the groundwater samples in this study had small but positive $D^{17}O$ values in nitrate ranging from +0.4 to +4.0 permil as compared to those in atmospheric nitrate; the annually averaged $D^{17}O$ value of atmospheric nitrate was determined to be +26.8 permil at Kosugi Station. We conclude that only less than 15% of the nitrate in groundwater originates directly from the atmosphere being processed in the soil, and substantial portion of nitrate is remineralized origins that undergo biologic processing in soil before being exported from the ecosystem.

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Keywords: triple oxygen isotopes, nitrogen isotope, nitrate, groundwater, nitrogen cycle