Accurate and precise quantification of atmospheric nitrate in streams draining land of various uses by using triple oxygen isotopes as tracers

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^{17}O anomalies were used to quantify the influence of changes in land use and population density between each catchment area on the fate of atmospheric nitrate by determining the areal distribution and seasonal variation in stable isotopic compositions including the ^{17}O anomalies ($\Delta^{17}O$) of nitrate for more than 30 streams within the same watershed. Nitrate in each inflow stream showed small annual average $\Delta^{17}O$ values ranging from +0.5‰ to +3.1‰, which corresponds to the mixing ratios of unprocessed atmospheric nitrate to total nitrate from 1.8 ±0.3% to 11.8 ±1.8%, with 5.1 ±0.5% as the average of all inflow streams. Although the annual average $\Delta^{17}O$ values tended to be smaller in accordance with the increase in annual average nitrate concentration from 12.7 to 106.2 μmol L^{-1}, the absolute concentrations of unprocessed atmospheric nitrate in the streams were almost stable at 2.3 ±1.1 μmol L^{-1} irrespective of the changes in population density and land use in each catchment area. We conclude that changes in population density and land use between each catchment area had little impact on the concentration of atmospheric nitrate. Thus, the total nitrate concentration originated primarily from additional contribution of remineralized nitrate from both natural sources, having values of +4.4 ±1.8‰ and −2.3 ±0.9‰ for $\delta^{15}N$ and $\delta^{18}O$, respectively, and anthropogenic sources having values of +9.2 ±1.3‰ and −2.2 ±1.1‰ for $\delta^{15}N$ and $\delta^{18}O$, respectively. In addition, both the uniform absolute concentration of atmospheric nitrate and the low and uniform $\delta^{18}O$ values of the remineralized portion of nitrate in the streams imply that in-stream removal of nitrate through assimilation or denitrification had small impact on the concentrations and stable isotopic compositions of nitrate in the streams, except for a few streams in summer having catchments of urban/suburban land uses. Additional measurements of the $\Delta^{17}O$ values of nitrate together with $\delta^{15}N$ and $\delta^{18}O$ enabled us to exclude the contribution of unprocessed atmospheric nitrate from the determined $\delta^{15}N$ and $\delta^{18}O$ values of total nitrate and to use the corrected $\delta^{15}N$ and $\delta^{18}O$ values to evaluate the source and behaviour of the remineralized portion of nitrate in each stream.

