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# Ongoing stress of transboundary pollution: Assessment of atmospheric N deposition influence by means of nitrate isotopes

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Anthropogenic emissions of reactive nitrogen (N) due to fossil fuel combustion and modern agriculture practices have dramatically increased in global scale. In the Japanese watershed along the Sea of Japan, large amounts of reactive N emitted from northeastern Asia have deposited onto forest ecosystems, and thus N concentrations in rivers and groundwaters have been increased year to year. Remotely modified N concentrations of terrestrial waters in local watersheds would affect coastal ecosystems. However, quantitative influence of N deposition on the watershed along the Sea of Japan is still unclear. In this study, we assessed the influence of atmospheric reactive N deposition on the watershed in Wakasa Bay by means of the $^{17}O$ anomaly ($\triangle$ $^{17}O$) in nitrate, which is an alternative tracer for atmospheric nitrate ($NO_3{}^-{}_{atm}$). As a result, fractions of $NO_3{}^-{}_{atm}$ in stream waters of the Kita River observed in 2014 changed from $>$10% in winter to 6% in summer. This seasonal variation corresponded to monthly changes in reactive N (= $NO_3{}^-$, $NH_4{}^+$ and others) deposition rates onto the watershed. However, most of $NO_3{}^-$ in stream water was derived from remineralized $NO_3{}^-$ by nitrification ($NO_3{}^-{}_{re}$) within the watershed. Apparent nitrification rates estimated by $\triangle$ $^{17}O$ were significantly higher than total N deposition rates. Although fractions of $NO_3{}^-{}_{atm}$ in groundwater and spring water (mean$\pm$SD = 4.3$\pm$0.3%), which was dated from 2001 to 2009 by $SF_6$, were significantly lower than that in stream water, yearly variation in $NO_3{}^-{}_{atm}$ fractions showed exponential increasing trend in the recent decade. These results imply that atmospheric reactive N deposition would affect severe influence on not only the forest ecosystem but also the coastal ecosystems.

Keywords: transboundary pollution, atmospheric nitrogen deposition, triple oxygen isotope, nitrogen saturation, retrospective analysis, Sea of Japan