

## Rise and fall of nitrogen concentration in a river draining to the Sea of Japan driven by atmospheric deposition

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Large amounts of reactive nitrogen (N) emitted from northeastern Asia are deposited onto forest ecosystems in watersheds draining to the Sea of Japan. Consequently, N concentrations in some Japanese rivers and aquifers have been increased until around 2010. In recent years, this trend has been changed possibly due to the decrease of atmospheric N emissions in China. Here, we hypothesize that Japanese groundwater and river water record the recent atmospheric deposition of N transported from East Asia. We rely on the <sup>17</sup>O anomaly of nitrate and groundwater dating via SF<sub>6</sub> to reveal atmospheric nitrate contributions to the Kita River watershed in Japan in the last three decades. Our latitudinal survey in central Japan revealed that the meteoric water sources were mostly supplied from the Sea of Japan in the northern area and from the Pacific Ocean in the central and southern areas. The mean atmospheric nitrate fraction in surface river water and groundwater in the Kita River watershed located in the northern area were 4.1 ± 1.3% and 5.2 ± 3.0%, respectively. Combining the SF<sub>6</sub>-derived groundwater dating with the <sup>17</sup>O anomaly shows an increase in atmospheric nitrate fraction in riverine nitrate until 2010 but a decrease since 2010, consistent with long-range atmospheric transport and deposition.

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