林冠から表層土壌にかけての大気由来硝酸の挙動の追跡：三酸素同位体組成を指標に用いて
Tracing the fate of atmospheric nitrate from forest canopy to surface soil using triple oxygen isotopes

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Increasing nitrogen (N) deposition could alter nutrient balance in forest ecosystems and increase nitrate (NO_3^-) leaching into stream, which may degrade water quality in downstream aquatic environment. Nitrogen deposition, microbial N transformation, and N uptake by plants and microbes create the complex dynamics of N in forest ecosystem, which makes it difficult to evaluate and predict the impacts of increasing N deposition on forest and aquatic ecosystems. Studies using ^{15}N tracer to determine the fate of N inputs have improved our understandings on the retention, transformation, and leaching of N in forest floor and surface soils. Studies that examine the natural stable isotopes of the precipitation and stream water have given insights into N saturation status of forested catchments against excess atmospheric N deposition. In contrast, our understanding of the N dynamics in canopy-soil continuum is still limited. Measurement of triple oxygen isotope composition ($\Delta^{17}O$) of nitrate enables us to quantify the absolute values of atmospheric NO_3^- (NO_3^-atm) concentration and flux with higher accuracy than the values estimated from $\delta^{18}O$ of NO_3^-. This makes it possible to trace NO_3^-atm after it enters forest ecosystem. Therefore, we aimed to determine the fate of NO_3^-atm from forest canopy to surface soil using $\Delta^{17}O$ of NO_3^- as a tracer. We also examined whether plant species affect the fate of NO_3^-atm in the forest.

This study was conducted in a cool-temperate broadleaved-coniferous mixed forest located in Uryu Experimental Forest of Hokkaido University, northern Hokkaido, Japan. We selected plots under canopies of four trees of *Quecus crispula* (Oak) and *Picea grehnii* (Fir) and within four canopy gaps exclusively dominated by dwarf bamboo, *Sasa senanensis* (*Sasa*), in a small watershed (ca. three ha) for water collection. Throughfall, litter leachate beneath Oa layer, and soil water at the depth of 10 cm were collected in each plot. Litter leachate and soil water were collected by tension-free lysimeter. Bulk precipitation was collected in a large canopy gap next to the watershed. Water samples were collected after a single rain event in June, September, and October in 2014, and analyzed for inorganic N concentrations and stable isotopic compositions of NO_3^- ($\delta^{15}N$, $\delta^{18}O$, $\Delta^{17}O$). The stable isotopic compositions of the nitrate were determined using Continuous-Flow Isotope Ratio Mass Spectrometry (CF-IRMS) system in Nagoya University (Tsunogai et al., 2010). The concentration and flux of NO_3^-atm were calculated from NO_3^- concentration, $\Delta^{17}O$ value, and sample water volume.

The average $\Delta^{17}O$ value of precipitation was +23.9 ‰. The average $\Delta^{17}O$ values of the throughfall were lower than that of precipitation, and were lower under Oak (+17.7 ‰) and Fir (+18.9 ‰) than under *Sasa* (+23.3 ‰). These results indicate the occurrence of microbial nitrification on the plant canopies and the magnitude of nitrification in Oak and Fir canopies is larger than in *Sasa* canopy. The litter leachate and the soil water showed much lower $\Delta^{17}O$ value of +2.3 ‰and +1.4 ‰, respectively. This indicates that

remineralizaed NO_3^- in soil becomes dominant in the leaching NO_3^- after passing the forest floor, but NO_3^-atm still exist after passing the forest floor and surface soil where biological activity is very active. The average fluxes of NO_3^-atm decreased from 0.84 mgN m^{-2} rain day^{-1} in the precipitation to 0.26 mgN m^{-2} rain day^{-1} in the soil water. The decrease of NO_3^-atm was larger between precipitation and throughfall than between throughfall and litter leachate or soil water. More than 50 % of NO_3^-atm in the precipitation was retained or consumed in the canopy, suggesting that forest canopy has a large influence on the fate of NO_3^-atm in forest ecosystem. Overall, our results suggest that retention of deposited N in the forest canopy is significant and N dynamics in the forest canopy may be very different among plant species.

キーワード：大気由来硝酸、三酸素同位体組成、林冠
Keywords: atmospheric nitrate, triple oxygen isotopes, canopy

