Recovery of stream water from acidification due to declining atmospheric sulfur deposition in a Japanese cedar forest near the Sea of Japan

*Hiroyuki Sase[1], Tatsuyoshi Saito[1,2], Tsuyoshi Ohizumi[3], Naoyuki Yamashita[4], Yayoi Inomata[5], Masayuki Morohashi[1], Naoko Take[3], Akiomi Takahashi[1], Masato Kamisako[1], Ryo Kobayashi[1], Shigeki Uchiyama[1], Ichiro Tayasu[6], Ki-Cheol Shin[6], Takanori Nakano[6], Makoto Nakata[2]

1. Asia Center for Air Pollution Research, Japan Environmental Sanitation Center, 2. Niigata University, 3. Niigata Prefectural Institute of Public Health and Environmental Sciences, 4. Forestry and Forest Products Research Institute, 5. Kanazawa University, 6. Research Institute for Humanity and Nature

Introduction: Atmospheric environment in Northeast Asia has been dynamically changing recently. Although SO_2 emissions in China peaked in 2006 and started declining thereafter, emissions of N compounds and related air pollutants have still been increasing. As shown in previous studies in Europe/North America, decline of S deposition promotes recovery of ecosystems from acidification but its process is not uniform. Since the deposition in Asia has just started declining, response of ecosystems should be studied as one of the current subjects in biogeochemistry. In this paper, in addition to the long-term monitoring data, isotopic analysis was applied to discuss the recovery process of ecosystems from acidification due to declining S deposition in a forested catchment.
Methods: The Kajikawa site (3.84 ha) was established in a small catchment within a Japanese cedar plantation in Shibata City, Niigata Prefecture. Rainfall outside the canopy (RF), throughfall and stemflow (TF+SF), and stream water (SW) have been collected biweekly or monthly since January 2002. Discharge of the stream water was measured continuously using the weir. The pH, electrical conductivity, alkalinity (only for SW) and major ions were measured for the water samples. Additionally, soil solutions have been collected since 2012, and then measurement of the sulfur isotopic ratio ($\delta^{34}S$) was started. Moreover, isotopes in Sr, Pb and water (H and O) were also measured for part of the water samples. The water year (WY) in the site was defined from June to May in the next year.
Results and discussion: Fluxes of SO_4^{2-} by RF and TF+SF increased in winter with those of Cl^- and Na^+. This suggested that the area suffered from long-range transported air pollutants due to seasonal winds through the Sea of Japan in winter. The long-term data indicated that the annual flux of non-sea salt SO_4^{2-} by TF+SF started declining after the peak in WY 2006/2007, reflecting the sulfur emissions in China. SO_4^{2-} concentrations in SW declined significantly, and pH and alkalinity increased. This may be the recovery process from the past acidification. The $\delta^{34}S$ values in RF and TF+SF were also lowering in summer (ca. 4‰) and increasing in winter (ca. 12‰). In winter, the rainwater may be affected by high-$\delta^{34}S$ sulfur derived from sea-salt (20.3‰) and coals in China (6.6‰: Ohizumi et al. 2016). On the other hand, the $\delta^{34}S$ values in SW were stable (ca. 9‰). The annual weighted mean $\delta^{34}S$ value in RF was also around 9‰. According to the input-output budget until WY 2013/2014, approximately 76% of the sulfur input was discharged into SW in the study catchment. The $\delta^{34}S$ value in RF and SW suggested that the sulfur input was once cycled and/or retained in the soil-plant system and then the isotopically homogenized sulfur was discharged into SW. Therefore, in the recovery process, the circulation/retention in the plant-soil system may sensitively respond to decline of the sulfur input. In seven lakes monitored by the Ministry of the Environment of Japan (MOEJ), similarly, SO_4^{2-} concentrations started declining after peaks in 2006/2007/2008, and the $\delta^{34}S$ values were stable without seasonality. This suggests that inland water in Japan sensitively responded to decline of the atmospheric sulfur input. In the presentation, results of other isotopic measurements will also be discussed.

Acknowledgements: The surveys in Kajikawa site were conducted under approval by Niigata Prefecture and assisted by the manager, Mr. Takanori Funayama. Monitoring on lake water and part of isotopic measurements were conducted as the Long-term Monitoring on Transboundary Air Pollution and Acid Deposition by MOEJ. Isotopic measurements of elements other than sulfur were conducted as Environmental Isotope Study in Research Institute for Humanity and Nature. The authors thank relevant persons and organizations.
References: Ohizumi, T. et al. 2016. Atmospheric Environment 140, 42-51.

Keywords: sulfur, acidification, recovery

