

## Exploring the sources of sulfur ion deposition and runoff in forest watersheds on the northern side of Lake Biwa

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To clarify the sources of sulfur ions in precipitation and runoff from forest watersheds, bulk deposition and stream water samples were collected within the small Kutsuki and Surumi forest watersheds on the northern side of the Lake Biwa basin, central Japan. Samples were analyzed for major ions and  $\delta^{34}\text{S}$ . Continuous monitoring from 1990 to 2010 showed that the average  $\text{SO}_4^{2-}$  concentration in stream water samples was  $1.62 \pm 0.31$  (0.76, 3.58)  $\text{mg l}^{-1}$  at Kutsuki and  $6.59 \pm 1.54$  (3.68, 16.1)  $\text{mg l}^{-1}$  at Surumi (t-test,  $p < 0.01$ ). However, the average  $\text{SO}_4^{2-}$  concentration in bulk deposition samples was similar in both watersheds:  $2.15 \pm 1.31$  (SD)  $\text{mg l}^{-1}$  (range, 0.202-10.2  $\text{mg l}^{-1}$ ) at Kutsuki and  $2.24 \pm 1.29$   $\text{mg l}^{-1}$  (0.350-6.07  $\text{mg l}^{-1}$ ) at Surumi (t-test,  $p > 0.05$ ). The  $\delta^{34}\text{S}$  values in bulk deposition samples fluctuated from +8.86 to +9.14 ‰ at Kutsuki and from +9.87 to +11.1 ‰ at Surumi, whereas non-sea salt (nss-)  $\delta^{34}\text{S}$  in stream water samples varied from +6.89 to +12.0 ‰ and from +4.64 to +5.11 ‰, respectively (t-test,  $p < 0.01$ ). It is said that the  $\delta^{34}\text{S}$  values in coal products from northern China varied from -3 to -1 ‰ and Japanese oil varied from +5 to +18 ‰.

Our findings suggest that the difference in the ability of canopies in the watersheds to catch  $\text{SO}_4^{2-}$  dry deposition is the reason for the significant difference in nss-  $\delta^{34}\text{S}$  values in stream water samples. The more open canopy in the Kutsuki watershed consists of young conifers and deciduous broadleaf trees, whereas the canopy in the Surumi watershed consists of mature conifer trees at a high density. Therefore, it appears that the Kutsuki watershed was only affected by  $\text{SO}_4^{2-}$  deposition from the Asian continent. There were not any domestic air pollution sources on the north side of Kutsuki. In contrast, the dense canopy of the Surumi watershed was affected by local  $\text{SO}_4^{2-}$  pollution sources of national roads which were located 1 km northwest from Kutsuki, rather than continental sources. Overall, this investigation suggests that the difference in stream water  $\text{SO}_4^{2-}$  concentrations in both watersheds is caused by the canopy differences.