Sulfur, nitrogen, and strontium isotope geochemistry of tributary rivers of Lake Biwa

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The water quality and biodiversity of Lake Biwa, Japan, have been deteriorating owing to expansion of human activities in the watershed, but the principal cause for the water quality deterioration has not yet been resolved. Recent studies have shown that the 15N/14N values of fish specimens (Isaza fish, Leucopsarion petersi) collected in northern Lake Biwa increased steadily from 1960 to 1999 (Ogawa et al., 2001), whereas the 87Sr/86Sr and 34S/32S values of Isaza fish decreased (Ishii et al., 2001) (Fig. 1). In order to elucidate the decadal change of sulfur, nitrogen, and strontium isotopes of Isaza fish and ambient water in Lake Biwa, we measured the concentrations and isotopic ratios of sulfur and strontium of water and the nitrogen isotopic ratios of organic particles in 41 inflowing rivers and one discharging river.

Tributary rivers of Lake Biwa can be divided into four areas based on the geology and human activity in the watershed. The concentrations of SO4, NO3, and Sr of inflowing rivers at downstream sites were generally high in the southern urban area and in the eastern area, where a large agricultural plain is situated, but low in the northern and western areas, whose watersheds are mountainous and with low population density. SO4, NO3, and Sr concentrations are also lower at upstream sites, which are closer to mountainous areas. Thus, the inflowing river receives large amounts of SO4, NO3, and Sr as it flows across the plain, where human activity levels are high. The 34S/32S or 87Sr/86Sr values of most eastern rivers at downstream sites are lower than, and the 15N/14N values of organic particles in the water are higher, than those of water in Lake Biwa, and the 34S/32S and 87Sr/86Sr values become more uniform as the proportion of the plain area in the watershed increases. River water in other areas has higher values of 34S/32S or 87Sr/86Sr than the lake water. This result indicates that the decadal decrease of 34S/32S, 15N/14N, and 87Sr/86Sr in the lake water has been caused mainly by the increased flux of SO4, NO3, and Sr from rivers in the eastern plain.

We simulated the yearly trend of 87Sr/86Sr and 34S/32S values in the lake water by assuming values for the isotopic ratios and concentrations of Sr and SO4 in the lake water in 1960 and those in the present inflowing waters from six rivers which are representative in the eastern area. We also assumed that the contribution of water mass from the eastern small rivers to Lake Biwa is 1%. The calculated trends of 87Sr/86Sr and 34S/32S values in the lake water (Fig. 1) well reproduce the observed trends of both isotopic values. Further, the Sr and SO4 contents of the present lake water are calculated as 55 g/L and 12 mg/L, respectively, values consistent with those estimated above for the lake water (59 g/L and 13.2 mg/L). This simple calculation supports our hypothesis that small rivers from the eastern paddy fields can alter lake water quality on a decadal time scale.

It is likely that in the plain, sulfur, nitrogen, and organic compounds induced by human activities generate sulfuric, nitric, and organic acids in the water, which accelerate the extraction of Sr from bedrocks, leading to the generation of Sr in the river water in the area. A good correlation between population density and the concentrations of SO4 and Sr in the inflowing river supports this idea and suggests that these ions may be used as an index of water quality to evaluate the impact of human activities.

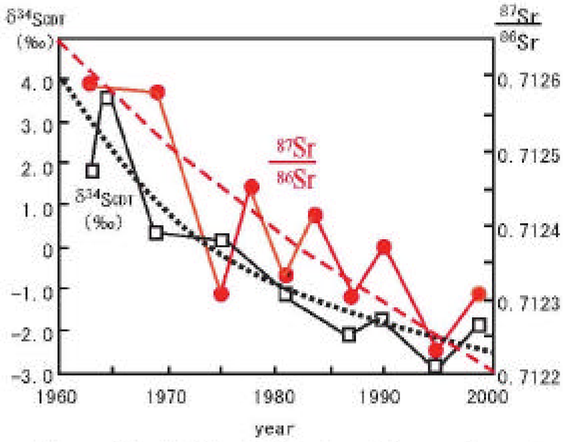

Figure 1. Annual change of the $^{87}Sr/^{86}Sr$ and $\delta^{34}S$ values of *Leucopsarion petersi* over 40 years from 1960 to 1999. Modified from Ishii *et al.* (2001) and Ito et al (unpublished data). Broken and dotted lines represent the calculated change in $^{87}Sr/^{86}Sr$ and $\delta^{34}S$ values of the lake water by inputs of small rivers from the eastern agricultural area. The $\delta^{34}S$ value of Isaza fish is assumed to be 2% lower than that of the ambient water.