Japan Geoscience Union Meeting 2011
(May 22-27 2011 at Makuhari, Chiba, Japan)

AHW023-02 Room:102 Time:May 25 08:55-09:20

Nitrate sources and processes of rivers in the Lake Biwa watershed: Synoptic surveys using nitrogen and oxygen isotopes

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In order to clarify the pathways and origins of dissolved nitrate (NO3-) in rivers flowing into Lake Biwa, Japan, three types of scale-coordinated surveys of concentrations and isotope compositions of NO3- were conducted: (1) synoptic river sampling of 32 representative inflow rivers, (2) two rivers in catchments with different land uses, and (3) intensive samplings in a headwater catchment. The d15N-NO3 was significantly positively correlated with the population density of each catchment. A mass balance model assuming the d15N-NO3 and the flow rate of sewage effluent was developed. The model simulated the relationship between the population density and the d15N-NO3 reasonably well, suggesting that the dominant source contributing to the increase in d15N-NO3 was the sewage effluent. The spatiotemporal distribution of d18O-NO3 of rivers, especially in the headwater streams, suggested the possibility of the addition of atmospherically derived NO3- through precipitation and snow, although the d18O-NO3 in soil system of forests in the headwater catchment showed the high nitrification potential and replacement of atmospheric NO3- by the microbially produced NO3-. In general, the d18O of NO3- in rainwater is remarkably higher than that produced by nitrifying bacteria in soils. Accordingly, the d18O-NO3 can often be used as an index of the impact of the atmospherically derived NO3-. While soil waters in $<$20cm depth had a strong signal of the atmospheric NO3-, the d18O-NO3 in soil water decreased in the deeper soil horizons, indicating that the dominant source of NO3- in this soil profile was nitrification. The net nitrate production of this soil profile was about 18 kg-N/ha/year, and deposited nitrate was about 6 kg-N/ha/year. Assuming that the annual mean d18O of deposited NO3- was 60 permil, and the mean value of bacterially produced nitrate in soil was about 0 permil, the average value for soil NO3- pool could be ˜15 permil. However, the observed d18O of the soil and groundwater was 0 to 6 permil and remarkably smaller than the above estimation based on annual mass balance. This suggests that the gross nitrification was sufficiently higher than net nitrification rate, and the major portion of NO3- produced in soil was reused by microbes. In forest-dominated catchments with natural drainage systems, a slightly elevated d18O-NO3 signal remained in the stream water even during base flow conditions. This study demonstrated that multi-scale, multi-isotopic investigation is a promising strategy for describing the spatial distribution of NO3- sources synoptically and is useful for evaluating the influences of land use change. The data set used in this paper is the first comprehensive collection of isotopic composition of NO3- in rivers of a large-scale basin in Asia.

Keywords: nitrate, stable isotope, river, Lake Biwa, Forest ecosystem