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Identifying nitrate sources and transformations in surface water by combining dual isotopes of nitrate and stable isotope mixing model in a watershed with different land uses and multi-tributaries

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Nitrate is essential for the growth and survival of plants, animals and humans. However, excess nitrate in drinking water is regarded as a health hazard as it is linked to infant methemoglobinemia and esophageal cancer. Revealing nitrate characteristics and identifying its sources are fundamental for making effective water management strategies, but nitrate sources in multi-tributaries and mixed land covered watersheds remain unclear. It is difficult to determine the predominant NO_3^- sources using conventional water quality monitoring techniques. In our study, based on 20 surface water sampling sites for more than two years' monitoring from April 2012 to December 2014, water chemical and dual isotopic approaches (δ^{15}N-NO_3^- and δ^{18}O-NO_3^-) were integrated for the first time to evaluate nitrate characteristics and sources in the Huashan watershed, Jianghuai hilly region, East China. The results demonstrated that nitrate content in surface water was relatively low in the downstream (<10 mg/L), but spatial heterogeneities were remarkable among different sub-watersheds. Extremely high nitrate was observed at the source of the river in one of the sub-watersheds, which exhibited an exponential decline along the stream due to dilution, absorption by aquatic plants, and high forest cover. Although dramatically decline of nitrate occurred along the stream, denitrification was not found in surface water by analyzing δ^{15}N-NO_3^- and δ^{18}O-NO_3^- relationship. Proportional contributions of five potential nitrate sources (i.e., precipitation; manure and sewage; soil nitrogen; nitrate fertilizer; nitrate derived from ammonia fertilizer and rainfall) were estimated using a Bayesian isotope mixing model. Model results indicated nitrate sources varied significantly among different rainfall conditions, land use types, as well as anthropologic activities. In summary, coupling dual isotopes of nitrate (δ^{15}N-NO_3^- and δ^{18}O-NO_3^-, simultaneously) with a Bayesian isotope mixing model offers a useful and practical way to qualitatively analyze nitrate sources and transformations as well as quantitatively estimate the contributions of potential nitrate sources in surface water. With the assessment of nitrate sources and characteristics, effective management strategies can be implemented to reduce N export and improve water quality in this region.