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Limitations of the isotopic composition of nitrates as a tracer of their origin

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Nitrogen and oxygen isotopes are traditionally considered and frequently used as tracers of nitrate sources in watersheds used for drinking water production. The enrichment of synthetic nitrate-containing fertilizers in ^{18}O due to the contribution of atmospheric oxygen in the production process confers a specific isotopic fingerprint to mineral fertilizers. In spite of the still widespread use on nitrate-containing synthetic fertilizers, their characteristic N and O isotope signatures are rarely unambiguously observed in nitrate-contaminated groundwater. We postulate, in line with Mengis *et al.* (2001), that fertilizer-derived nitrate is not directly and rapidly transferred to groundwater but rather retained in the soil-plant system as organic N and then mineralized and re-oxidized (termed the mineralization-immobilization turnover, MIT) thereby re-setting the oxygen isotope composition of nitrate and also changing its N isotope ratios. We show examples from watersheds on diverse alluvial/clastic and carbonate aquifers in eastern and northern France where, in spite of the use of mineral fertilizers, evidenced also through other isotopic tracers (boron isotopes), both N and O-isotope ratios are very homogeneous and compatible with nitrification of ammonium where 2/3 of oxygen is derived from soil water and 1/3 from atmospheric O_2. These field data are corroborated by lysimeter data from Canada. Even if in areas where ammonium is derived from chemical fertilizers, N values still tend to be lower than in areas where ammonium is derived from manure/sewage, this is clearly a limitation to the dual isotope method (N, O) for nitrate source identification, but has important implications for the nitrogen mobility and residence time in soils amended with synthetic fertilizers (Sebilo *et al.*, 2013).

Mengis M., Walther U., Bernasconi S. M., Wehrli B. (2001) Limitations of Using δ18O for the Source Identification of Nitrate in Agricultural Soils. *Environmental Science & Technology*, **35**, 1840-1844.

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