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Is groundwater age the main control for slow turnover of nitrate in a fractured groundwater system?

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Slow transformation processes are known to control the chemical, isotopic, and redox evolution of large-scale aquifers (Edmunds et al., 1982; Katz et al., 1995). However, at the field scale some of the crucial biogeochemical processes governing pollutant turnover and their interrelations with hydrology are poorly understood. Particularly, only little is known about denitrification in fractured rock aquifers. Therefore, the main objective of the presented study is to assess where and how slow turnover of nitrate ans other pollutants in the deeper subsurface take place.
The studied fractured and partly karstified aquifer consisting of Triassic black limestones and dolomites is located in the catchment of the Ammer river (ca. 350 km^2) close to Tübingen in southern Germany. Near the recharge area, the aquifer is covered by loess allowing intensive agriculture. Further downgradient, the cover consist of a series of mudstones and sandstones of variable permeability. The aquifer is used for drinking water purposes by regional water suppliers. Land-use is dominated by agriculture with arable land covering nearly 50% of the catchment.
Over the last years a variety of groundwater samples have been collected from the groundwater system including 6 water supply wells, 4 karstic springs, and 9 monitoring wells in the recharge area. This allowed to identify spatial and temporal patterns of water quality including concentrations of major ions, dissolved organic carbon (DOC), organic pollutants (e.g., pesticides), and environmental isotopes. Groundwater age distributions at most of these locations were derived from tritium, 3He, CFCs and SF6.
Groundwaters in the recharge area show high concentrations of nutrients (e.g. 20-51 mg/L of nitrate and 0.2 to 0.05 μg/L of phosphate). Of special concern are disparate nitrate concentrations ranging from below 0.4 to 20 mg/L in water supply wells although screen depths of the production wells are similar. Concentrations of dissolved oxygen comply with nitrate concentrations (negligible oxygen when nitrate is low and more than 1.5 mg/L of oxygen when nitrate is high). At the same time, apparent groundwater ages (derived by tritium-3He) show a narrow range of 14 to 23 years, implying that all wells should be affected by high nitrate inputs from recent agricultural practices. The latter results in average concentrations of e.g., 32 mg/L in the karstic Ammer springs close to the recharge area. The much lower than expected nitrate concentrations along longer groundwater flowpaths towards some of the production wells indicate the occurrence of denitrification. Due to the geochemical composition of the rock matrix, nitrate reduction may proceed through autotrophic and mixotrophic denitrification using organic carbon and pyrite as electron donor.