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Constraining nitrogen cycling hotspots in contaminated aquifers

Naomi Wells and Kay Knoeller
Helmholtz Centre for Environmental Research - UFZ, Catchment Hydrology, Halle (Saale), Germany (naomi.wells@ufz.de)

Accurate assessments of the fate of inorganic nitrogen (N) in groundwater are needed in order to mitigate the threat that ammonium (NH_4^+) and nitrate (NO_3^-) pose to water quality and the long-term health of down-gradient ecosystems. However, such assessments are currently limited by difficulties in measuring the biological attenuation (via either denitrification or anaerobic ammonia oxidation (anammox)) of these reactive species in-situ. Based on the knowledge that both of these processes can create unique fractionation patterns in the residual N pools, the objective of this research was to build a template for identifying and quantifying N removal hotspots within complex aquifers using isofluxes. The variations in concentration and isotopic abundance of multiple dissolved inorganic N species (δ^{15}N of NH_4^+, and δ^{15}N and δ^{18}O of NO_2^- and NO_3^-) were measured in 100 wells across two contaminated megasites in Western Europe. The sampling locations were selected span the NH_4^+ (the dominant N form in both sites) concentration gradient (0 to 900 mg NH_4^+-N l-1) over depth and distance, which coincided with gradients in co-contaminants BTEX and sulphate of 0 to 5 mg l^{-1} 4 to 11000 mg l^{-1}, respectively. Although NO_2^- is a key component of both anaerobic and aerobic ammonium oxidation, it is rarely detected in groundwater. Yet, by analysing for it on-site, we found that NO_2^- concentrations reached up to 0.7 mg NO_2-N l^{-1} and had a highly sensitive isotopic composition (mean of -5 $\pm$23‰ (δ^{15}N) and +11 $\pm$12‰ (δ^{18}O)). The largest NO_2^- concentrations coincided with those of NH_4^+ levels, meaning that attenuation fluxes could be partitioned between anammox and denitrification using simple isotope mass balance calculations based on Rayleigh type isotope fractionation and established nitrate (δ^{15}N and δ^{18}O) isotope dynamics during denitrification. The constraints on N attenuation within these complex hydrological and chemical setting created by overlaying isoflux maps for each N species provide a template for a new means of assessing a site's in-situ remediation capacity.