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Fate of nitrate and origin of ammonium during infiltration of treated wastewater investigated through stable isotopes

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The EU FP7 project MARSOL addresses water scarcity challenges in arid regions, where managed aquifer recharge (MAR) is an upcoming technology to recharge depleted aquifers using alternative water sources. However, a potential impact to water quality is increasing ammonium concentrations, which are known to be a problem resulting from bank filtration. In the context of MAR, increasing ammonium concentrations have received little attention so far.

A soil column experiment was conducted to investigate transformations of nitrogen species when secondary treated wastewater (TWW) is infiltrated through a natural soil (organic matter content 5.6%) being considered for MAR. The TWW contains nitrate and dissolved organic nitrogen (DON), but typically very low ($<$0.2 mg/L) concentrations of nitrite and ammonium. In addition to the nitrate and DON in the inflow water, nitrogen in the soil organic matter is a third possible source for ammonium produced during infiltration.

The experiment simulated MAR using a series of wetting-drying cycles. At the end of the wetting phases, pore water samples were collected from six depths. Results show that the largest decreases in nitrate concentration occur in the upper part of the soil, with on average 77% attenuated by 15 cm depth and 94% by 30 cm depth. Starting at 30 cm and continuing downward, ammonium concentrations increased, with concentrations reaching as high as 4 mg-N/L (the EU drinking water limit is 0.41 mg-N/L).

Selected samples were also measured for stable nitrogen and oxygen isotopes. Nitrate became isotopically heavier (both N and O) with increasing depth (samples collected at 5 and 15 cm below the soil surface), with most results forming a linear trend for δ^{18}O vs. δ^{15}N. This pattern is consistent with denitrification, which is also supported by the fact that the ammonium concentration first increases at a depth below where most of the nitrate is consumed. However, the relationship between δ^{15}N-NO_3^- and nitrate concentration is not clearly logarithmic, so processes other than denitrification are not ruled out for explaining the fate of nitrate.

The δ^{15}N of ammonium in the water samples and of nitrogen in the soil were also measured. With increasing depth and time, the δ^{15}N-NH_4^+ (mean 4.3‰) decreases and approaches the δ^{15}N of the pre-experimental soil of 2.4‰. This suggests that ammonium is formed at least in part from the soil organic matter, likely through a combination of leaching and microbial processes.

Although most nitrate attenuates by 15 cm depth and very little ammonium is observed here, some nitrate (usually $<$0.5 mg-N/L) was observed at depths of 30 cm and below, especially early in the experiments. Starting at 30 cm depth, organic carbon concentrations and thereby also C:NO_3^- ratios become high ($>$10), which are conditions sometimes found to be favorable to dissimilatory nitrate reduction to ammonium. Rayleigh enrichment factors also suggest that nitrate may be the source of some of the ammonium. Measurements of additional samples and organic nitrogen isotopes are planned, in order to further evaluate the fate of nitrate and the source(s) of the ammonium.