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Groundwater denitrification in two agricultural river catchments: influence of hydro-geological setting and aquifer geochemistry

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Identifying subsurface environments with a natural capacity for denitrification is important for improving agricultural management. At the catchment scale, a complex hierarchy of landscape, hydro-geological and physico-chemical characteristics combine to affect the distribution of groundwater nitrate (NO_3-). This study was conducted along four instrumented hillslopes in two ca. 10km2 agricultural river catchments in Ireland, one dominated by arable and one by grassland agriculture. Both catchments are characterised by well drained soils, but have differing aquifer characteristics. The arable catchment is underlain by weathered Ordovician slate bedrock which is extensively fractured with depth. The grassland catchment is characterised by Devonian sandstone bedrock, exhibiting both lateral (from upslope to near stream) and vertical variations in permeability along each hillslope. The capacity for groundwater denitrification was assessed by examining the concentration and distribution patterns of N species (total nitrogen, nitrate, nitrite, ammonium), dissolved organic carbon (DOC), dissolved oxygen (DO) and redox potential (Eh) in monthly samples from shallow and deep groundwater piezometers (n=37). Additionally, the gaseous products of denitrification: nitrous oxide (N2O) and excess dinitrogen (excess N2) were measured seasonally using gas chromatography and membrane inlet mass spectroscopy, respectively. The slate catchment was characterised by uniformity, both laterally and vertically, in aquifer geochemistry and gaseous denitrification products. The four year spatial mean groundwater NO_3–N concentration was 6.89 mg/l and exhibited low spatial and temporal variability (temporal SD: 1.19 mg/l, spatial SD: 1.185 mg/l). Elevated DO concentrations (mean: 9.75 mg/l) and positive Eh (mean: +176.5mV) at all sample horizons indicated a setting with little denitrification potential. This non-reducing environment was reflected in a low accumulation of denitrification products (excess N2 mean: 1.57 mg/l, N2O mean: 1.61μg/l). Groundwater in the sandstone catchment had a comparable mean NO_3–N concentration to that of the slate site (6.24mg/l) and while temporal variation was low (SD: 0.9 mg/l), spatial variation was substantially greater (SD: 3.63 mg/l). The accumulation of denitrification products in the sandstone catchment showed a large contrast to that of the slate with excess N2 ranging from 0.16-8.77 mg/l and N2O from 0.07-66.42 μg/l. Mean dissolved oxygen concentration and redox potential were 5.6mg/l and 67.5mV respectively. The near stream zones in particular were marked by favourable denitrifying conditions: hydraulic conductivity (<2m/day), Eh (<50mV) and DO (<5mg/l). Winter recharge had a diluting effect, increasing the concentration of DO and Eh with a concurrent decrease in excess N2 and N2O. The evolution of groundwater geochemistry along a subsurface flow path is a function of residence time. While both catchments are characterised as permeable, the slate catchment exhibits greater hydraulic conductivity values, particularly at depth, with groundwater geochemistry in all horizons reflective of recently recharged water. The deeper groundwater pathways and near stream zones in the sandstone catchment have a lower hydraulic conductivity. As such, dissolved oxygen and redox gradients occur with depth, causing the development of NO_3- reducing zones.