Geophysical Research Abstracts
Vol. 18, EGU2016-17523-1, 2016
EGU General Assembly 2016

Temporal and spatial variations in groundwater quality resulting from policy-induced reductions in nitrate leaching to the Rabis Creek aquifer, Denmark

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Twenty-five years of annual groundwater quality monitoring data from the sandy unconfined Rabis Creek aquifer were used to assess the effects of political actions aimed to reduce nitrate leaching to the aquifer. Data were collected from eight multilevel samplers along a ~3 km transect, which follows the general direction of groundwater flow. Each multilevel sampler comprises 20 screens placed with a 1 m vertical distance from near the water table downwards. The transect covers areas of livestock, plantation & heath, and agriculture. The history of nitrate leaching to the aquifer was assessed using data from screens close to the water table of multilevel samplers placed within agricultural areas. According to these data, nitrate concentrations of infiltrating 'agricultural' water peaked at 2-3 mM (120-180 mg/L) in the year 1989, and then gradually decreased and stabilized at 0.25-1.0 mM (15-60 mg/L) from year 2000. Local farmers declare having used the maximum fertilization rate allowed during the period. The timing of the observed decrease therefore suggests a direct link to the political action plans implemented in the same period. Parallel to the development in nitrate leaching, although with a transport time lag, the average concentration of nitrate in the oxic zone of the aquifer was roughly halved between 2000 and 2013. As a response to political initiatives of the late 1980'ies, part of the area covering the aquifer was changed from agriculture to non-fertilized grass for livestock; the data shows that this effectively remediated the aquifer underneath in less than 20 years, to become nitrate-free and attain background sulfate levels. The oxidized and pyritic reduced zone of the aquifer is separated by a <1 m redoxcline. Denitrification by the pyrite releases sulfate that is retained down-gradient as a tracer for historical nitrate loading to the aquifer. Thus sulfate concentrations currently increase in the multilevel samplers positioned furthest down-gradient along the transect. During the 25 year monitoring period the redoxcline has moved by one to a few decimeters, as controlled by the aquifer sediment's pyrite content. Further, the data indicate that no zero-valent sulfur is precipitated during pyrite oxidation in the aquifer, while most of the pyritic iron is precipitated. Nickel (Ni^{2+}) is released at the redoxcline resulting in concentrations more than twice the 20 μg/L Danish drinking water limit. The data clearly indicate that this Ni^{2+} contamination can be ascribed to the agricultural nitrate loading and would not occur under natural conditions. A 2D reactive transport model was constructed (PHAST 3) to simulate the temporal and spatial development in nitrate and sulfate concentrations in the aquifer while taking into account effects of dispersion. The model predictions indicate that sulfate concentrations, despite dispersive mixing, is still increasing along down-gradient stretches of the aquifer, where flow paths surface from the deeper up-gradient part of the aquifer, to eventually discharge into the Rabis Creek.