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Multi-isotopic approach (δ^{15}N, δ^{13}C, δ^{34}S, δ^{18}O and δD) for tracing nitrogen groundwater pollution and evaluating natural attenuation processes in Empordà basin (NE Spain)

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Nitrate pollution has become a major threat to groundwater quality as the maximum nitrate concentration allowed by the European Directive 98/83/CE in waters for human consumption, 50 mg/L, is reached in most of the regional aquifers in Europe. Numerous studies have been carried out over the last decades in order to assess isotope data (N, C, S, O) as a useful tool to evaluate the origin of nitrate contamination and the occurrence of several chemical processes during transport, such as natural attenuation (Aravena and Robertson, 1998).

The Baix Ter basin in the Empordà region (Catalonia, NE Spain) was declared as vulnerable region to nitrate contamination by the local Catalan government in 1998. The regional hydrological system presents a widespread non-point source of pollution due to a large amount of nitrogen which is generally used as a fertilizer in excess all over the area. The main sources of nitrate in groundwater in that zone are either the manure derived from intensive activity of pig farm exploitations or the use of synthetic fertilizers due to agriculture activity (mainly maize).

The sampled area is 200 km^2 and consists geologically of Paleogene detritic and carbonate sedimentary rocks (east and south), Neogene clay facies (west), and Holocene alluvial deposits of Ter river, the main stream in the area, and its tributary Daró. Because of a high lithological diversity, the area presents a complex distribution of hydrogeological units. The basic units are: 1) an unconfined aquifer with sand and gravel with some clay in the matrix (mainly from Quaternary) and 2) a confined and sometimes unconfined fractured aquifer with thickness discontinuity (mainly from Paleogene). Alluvial aquifers are those located at the river terraces and considered as productive. Groundwater extractions for agriculture use and urban supply are mostly in Quaternary formations.

This study is intended for characterizing chemically and isotopically nitrate contaminated groundwater and distinguishing between the contribution of each pollutant source (pig manure, fertilizers). Likewise, another important aim is to identify the fractionation processes of NH_4^+ volatilisation, nitrification and denitrification in order to assess feasible natural attenuation conditions.

Chemical data for groundwater samples collected in two field samplings in Jan-2004 and Aug-2004 show that groundwaters are mainly bicarbonate calcium type waters and with a wide range of nitrate concentrations achieving values as high as 480 mg/L.

δ^{18}O and δD values of groundwaters are very similar in all studied samples and plot very close to the Local Meteoric Water Line (MWL), so that rainwater is the main recharge of the area. The isotope composition of the groundwater nitrate ranges between +5.0 and +32.5%¸ for $\delta^{15}N_{NO3}$, but most of the samples present values higher than 8%¸. As a preliminary interpretation, groundwaters are mainly influenced by pig manure ($\delta^{15}N_{NH4}$ between +8 and +15%¸; Vitòria et al., 2004a). The higher $\delta^{15}N_{NO3}$ values can be explained as a result of denitrification processes in groundwater or ammonium volatilization when the fertilizers are spread onto the fields. The combination of $\delta^{15}N_{NO3}$ with $\delta^{18}O_{NO3}$ permits discriminate between these two processes. The negative values of $\delta^{34}S_{SO4}$, which ranges from -16.0 to +14.7%¸, indicate pyrite oxidation, reaction involved in some denitrification processes.

Thus, multi-isotopic characterisation of groundwaters can provide a useful tool to trace the origin and processes of nitrate contamination. This information should allow to improve manure handling and disposal practices as well as water management.

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