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Modelling of biodegradation and isotope fractionation of petroleum hydrocarbon plumes in 2D bench-scale tank experiments

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In case of the natural attenuation of an organic contaminant in porous aquifers, biodegradation together with transverse mixing represent the main processes that determine plume length and contaminant concentration. In order to study the interplay of biodegradation processes in detail, aerobic and anaerobic biodegradation of ethylbenzene were investigated in 2D bench-scale tank experiments. This set-up reduces natural complexity and allows a spatially and temporally resolved evaluation of individual parameters and processes. Flow of oxic and reduced groundwater through the homogeneously and heterogeneously packed porous media was maintained by simultaneously injecting water into inlet ports and extracting from outlet ports at the sides of the tank at a transport velocity of 1.2 m d^{-1}. A mixture of unlabeled and fully deuterium-labeled ethylbenzene isotopomers (ratio 3:1) was supplied through one central inlet port, generating an ethylbenzene plume. In the first phase of the experiment, the system was recharged with water containing oxygen as electron acceptor and inoculated with the strain *Pseudomonas putida* F1 to initiate aerobic biodegradation of ethylbenzene. Later, nitrate was added as an additional electron acceptor and competitive degradation under denitrifying conditions was studied after inoculating strain *Aromatoleum aromaticum* EbN1. The spatial distribution of anaerobic degradation was investigated using measurements of compound-specific stable isotope fractionation along vertical profiles at the tank outlet.

High resolution reactive transport modelling was applied to substantiate the experimental results and to elaborate the factors controlling biodegradation. For the numerical evaluation of the laboratory experiments, the FEM code OpenGeoSys (OGS) was used. OGS is a software for the simulation of thermo-hydro-mechanical-chemical (THMC) processes in porous media that allows simulation of multi species reactive transport, kinetic and equilibrium geochemical reactions, biodegradation reaction following multiplicative Monod kinetics as well as stable isotope fractionation. Sensitivity analysis showed that although an increase of water flow velocity and sediment porosity results in higher transverse mixing, the reduced contact time between electron donor and acceptor limits ethylbenzene degradation. Simulated and measured ethylbenzene and oxygen concentrations showed a good agreement for the aerobic degradation phase, while the evaluation of the anaerobic phase turned out to be more difficult as there are uncertainties in the correct stoichiometry of the denitrification reaction. The model results, calibrated on the stable isotope signatures, showed that for the case of aerobic/anaerobic degradation, the reproduction of the observed isotopic patterns is strongly dependent on the assumed initial distribution of microbial biomass.

Key words tank experiments; numerical modelling; biodegradation; isotope fractionation.