

When does back-diffusion from low-permeability porous media into aquifers create isotope fractionation?

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At many historical polluted sites with chlorinated solvents, diffusion has transported dissolved compounds into aquitards. Remediation of the residual solvents in aquifers is possible by e.g., chemical, or biological reduction processes, but after source removal, it is highly probable that back-diffusion of the dissolved compounds from aquitards will occur and will require a prolonged post-remediation monitoring.

The objective of this work is to propose stable isotopes as a tool to monitoring processes at sites impacted by back-diffusion. The main question is does back-diffusion create isotope fractionation in a magnitude that would interfere with classical use of isotope analysis for monitoring of reductive processes during remediation.

To this end, we developed analytical reactive transport equations which simulate the carbon isotope fractionation of chlorinated hydrocarbons during forward- and back-diffusion in aquitards. With these equations which include linear sorption to aquitard solids, a systematic view is obtained under which conditions isotope shifts in chlorinated solvents diffusing back from aquitards will be generated. We simulate a case where trichloroethene diffused forward into a clay aquifer of either infinite or finite thickness, and then diffused back for 100 years. Our model finds that although forward diffusion creates isotope fractionation in the aquitard, it is later cancelled out during back-diffusion when the aquitard is of infinite thickness, and when no degradation occurs. An aquitard with a finite thickness will create an isotope fractionation with time, in an insignificant magnitude compared to fractionation by reduction reactions. We show that aquitards can only contribute to isotope fractionation when fractionating degradation reactions are present in the aquitard (e.g. abiotic reductions). To confirm absence of isotope fractionation during back-diffusion, an experiment with three chlorinated solvents diffusing from water to agar gels and then diffusing back to clean water was performed. No significant isotope fractionations were found for either dichloromethane, trichloroethene or monochlorobenzene. This result agrees with findings in literature made on the basis of numerical simulations of diffusion.