Chlorine and bromine stable isotope diffusion through natural porous media

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Chlorine and bromide are widely assumed to be natural conservative tracers in subsurface water systems. In some groundwater and porewaters, δ 37Cl has been recognized as an efficient tool to identify diffusion, a potentially important process of transport of solute in natural water reservoirs.

Yet, the chlorine isotope fractionation factor during diffusion (α) is still debated. Free-water diffusion experimental works (such as Richter et al., 2006 or Eggenkamp et al., 2009) lead to α (Cl) of about 1.00155 and α (Br) of about 1.00064, whereas α (Cl) deduced from field-work studies (such as Groen et al., 2000 or Lavastre et al., 2005) are often superiors to 1.0020.

In this study, we attempted to experimentally explore the effective diffusion of both chloride and bromide in a natural porous media. Experiments were achieved in a diffusion cell with a Lavoux carbonate rock and two Fontainebleau sandstones. As all parameters of diffusion were precisely controlled, we developed a 1-D diffusion model allowing to optimize the effective diffusion coefficient and the isotope fractionation factor at the same time. Our results are very consistent with previous experimental studies in free-water, demonstrating that effective diffusion of two isotopes in a porous media is well described by their respective diffusion coefficients in free-water times a tortuosity parameter. We concluded that the relatively high α values deduced from field-work studies cannot be accounted for reflecting additional processes within the porosity.

Finally, we explored our results and their implications to understand high "natural" isotope fractionation factors, in the light of the recent theoretical framework for solute diffusion proposed by Bourg et al., (2007).