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Origin of halides (Cl- and Br-) and of their stable isotopes (d37Cl and d81Br) at the Tournemire URL (France) - Experimental and numerical approach

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This work is part of research conducted by the Institute of Radiological and Nuclear Safety (IRSN) on the geological disposal of High-Level and Intermediate-Level Long-Lived (HL-ILLL) radioactive waste in deep clayrocks. In France, the choice of the potential host rock for the geological storage is focused on the Callovian-Oxfordian (COx) of Meuse/Haute-Marne from its low permeability, capacity for self- sealing, high sorption and ability to radionuclide (RN) transport by diffusion. IRSN, which plays an expert role for ASN has its own underground research laboratory in a clayrock which has strong analogies to the COx. This is the Toarcian/Domerian clayrock located at Tournemire in southern Aveyron in France.

The purpose of this study was to assess the transfer of RN in the Tournemire clayrock through the study of halides contents and of their stable isotopes (Cl-, Br-, Cl-/Br-, d37Cl, d81Br). The approach used was multiple and consisted for halides to: 1) Assess their stock in different fractions of the rock by applying several techniques including i) alkaline fusion for their total stock, ii) leaching to access their stock in porewater and to mineral phases sensitive to dissolution iii) cubic diffusion for their stock in porewater, 2) Get their diffusive transport parameters of a selection of samples from the upper Toarcian by cubic diffusion experiments modelled using the Hytec transport code developed by Mines ParisTech and 3) Model their transport after palaeohydrogeological known changes of the Tournemire massif.

The experimental approach, conducted at the LAME lab, did not lead to an operational protocol for the alkaline fusion due to an incomplete rock dissolution. Leaching was used to characterize the concentrations of halides in the fractions of pore water and of minerals sensitive to dissolution. The results show levels of halides much higher than those of pore water with very low Cl/Br ratios likely resulting from the dissolution of mineral species. The cubic diffusion produced the pore diffusion coefficients for Cl and Br as well as their concentration in the porewater. Cubic diffusion also allowed to estimate a Cl to Br pore diffusion coefficient ratio, necessary to calculate the profiles of Cl/Br. These estimates have required the use of the transport code Hytec i) for dimensioning and implementing the experiment in a time frame compatible with the work period, ii) for analysing the sensitiveness of the model to the accessible porosity and to the diffusion coefficient which act respectively to the steady phase and transient phase of the experiments, and finally, iii) for adjusting the pore diffusion coefficients of Cl and Br to an accessible porosity of 3-4%.

The Hytec code was then used to check the consistency of the current profiles of chlorides, bromides, 35Cl , 37Cl , d37Cl, Cl/Br in 1D, a fake drilling assumed crossing the entire clayrock. The assumption is that halides have undergone a diffusive transport between seawater trapped during sedimentation and meteoric waters infiltrated at different times to domain boundaries. Four scenarios were tested according to the paleohydrogeological history of the massif. All tracers and scenarios are consistent with a unique marine source of halides more or less diluted by meteoric waters. The duration of the diffusive exchange initially suggested 85 $\pm$ 10 Ma (Bensenouci, 2010) is never contradicted despite uncertainties related to changes in boundary conditions. This body of evidence would suggest that molecular diffusion is the transport process which has affected and still affect the Tournemire clayrock, outside fault zones. The d37Cl results expected on the surrounding carbonated aquifers, leachates and fracture waters (including d81Br values) should help to refine the models and the results.