
Diffusion experiments at Mont Terri (Switzerland): overview and results

Savoie S.

IRSN DEI SARG Laboratoire d'Etudes des Transferts dans les Sols et le sous-sol
F-92262 Fontenay-aux-Roses

Abstract:

Several diffusion experiments have been performed at the field scale in the underground rock laboratory of Mont Terri (Switzerland) to verify the reliability of diffusion parameters obtained at lab scale. The principle of *in situ* diffusion experiments is based on the injection, in a packer-off section of a borehole, of a tracer cocktail diffusing into the rock and whose its concentration decrease is monitored by means of a circulation circuit located at the surface. Subsequently, the interval section is overcored and analysed for the tracer profiles.

Overall, the obtained tracer data confirmed that diffusion is the dominant transport process for solutes in Opalinus Clay. The diffusivity and diffusion porosity of anions was found to be lower than tritiated water, indicating anion exclusion effect. Concerning the cations, one observed that when the interaction of these species with clay surface was stronger, their decrease relative to HTO in interval was faster and their penetration depth was lower (max 3 cm after 10 months for Cs+).

Finally, *in situ* data were found to be consistent with small-scale lab diffusion experiments performed parallel to the bedding plane, suggesting that the upscaling effects for diffusion are small.

1 INTRODUCTION

One of the key issues for a repository's safety assessment is to determine the predominant radionuclide transport mechanism. Transport could occur either by molecular diffusion through the pore water or by advective flow within the rock matrix and/or fractures. Several studies indicate diffusion to be the dominant transport mechanism in consolidated argillaceous rocks (Opalinus Clay in Switzerland, argillite of Tournemire in France). Firstly, large-scale profiles of natural tracers, such as chloride, bromide and stable isotopes can be explained by slow diffusion process from the saline pore water of clayey rocks to the young ground water of the adjacent limestone formations (Rübel *et al.*, 2002; Degueudre *et al.*, 2003, Patriarche *et al.*, 2004). Secondly, the very low hydraulic conductivity measured in the clay formation limits the contribution of advective transport.

The diffusion parameters are generally derived from laboratory measurements on centimetric rock samples (Van Loon *et al.*, 2004a; Savoie *et al.*, 2006; Savoie *et al.*, in press). As these values are required in the modelling at the scale of rock formations for performance assessment, their reliability has to be verified, especially regarding potential upscaling effects. Hence, since the beginning of the Mont Terri Project, several diffusion experiments have been performed at the field scale either in the undisturbed matrix of the Opalinus clay (experiments DI, DI-A, DI-A2, DI-B, DR) or in a highly fractured zone (FM-C) to acquire such *in situ* diffusion parameters.

2 CONCEPT OF *IN SITU* DIFFUSION EXPERIMENTS

The general concept of all the experiments is based on the first *in situ* experiment, the so-called DI (Palut *et al.*, 2002).

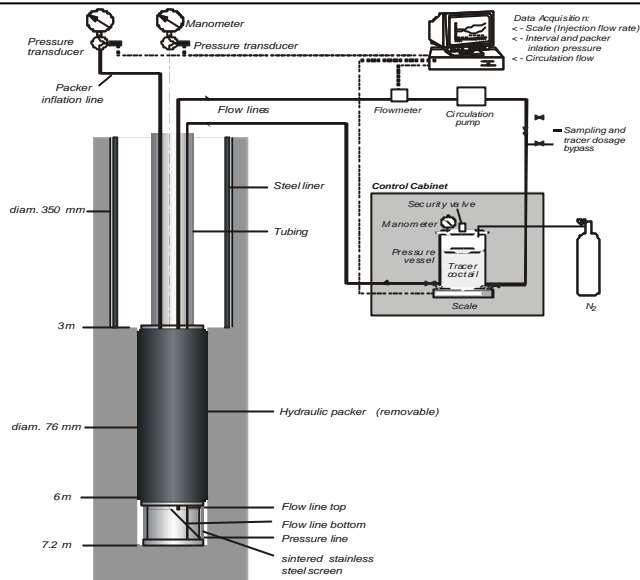


Fig. 1: Example of experiment test set-up (DI-A, Wersin et al., 2004 & van Loon et al., 2004b)

A tracer cocktail is injected into a packed-off section of a borehole as a pulse-test. Pressure in this interval is maintained equal to the pore pressure of the surrounding rock in order to prevent any hydraulic gradient around the borehole and to avoid advective transport processes. The evolution of the tracer concentration in the injection system is monitored over time with a set-up allowing fluid circulation from the interval to the surface (Fig. 1). After a certain period of diffusion (from 10 months to 2 or 3 years), the claystone section surrounding the interval is retrieved by overcoring the whole borehole and is subsequently analysed for the tracer profiles. Then, both the tracer decrease in the interval and tracer profiles in rock are simulated with the same diffusion parameters in 2D or 3D. Finally, the obtained diffusion data are compared with lab diffusion data.

3 LOCATION AND SPECIFICATION OF DIFFUSION EXPERIMENTS

Figure 2 shows the location of the Mont Terri underground rock laboratory.

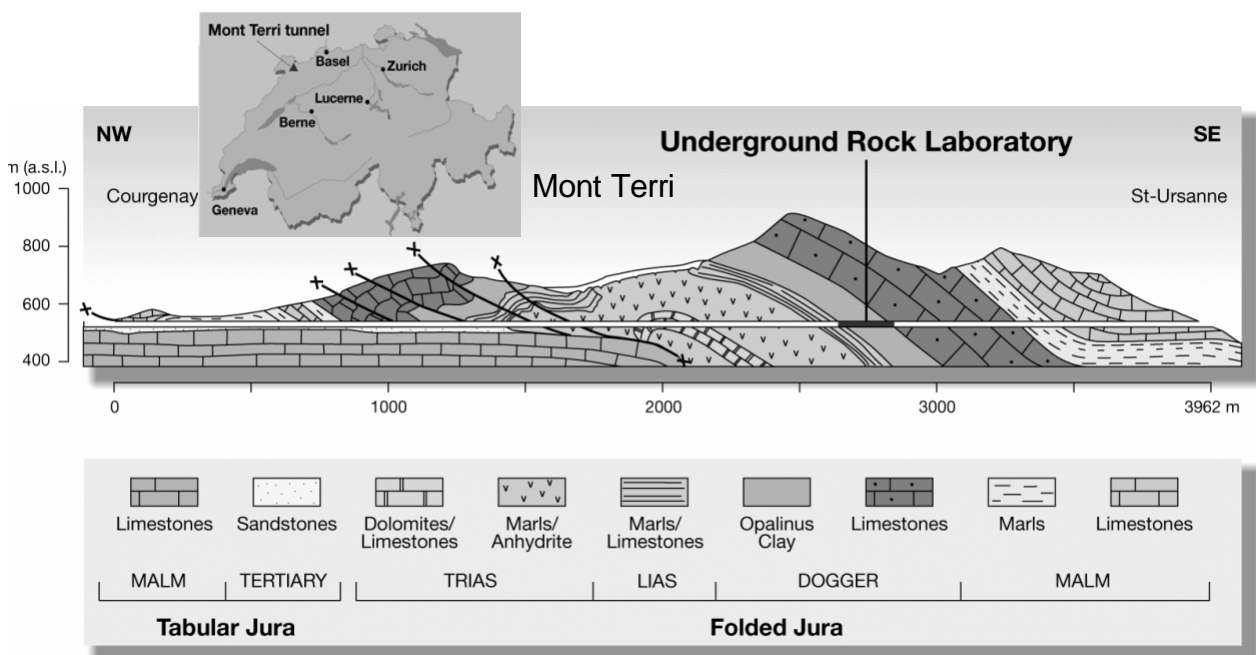


Fig. 2: Location of the underground rock laboratory of Mont Terri

The six different diffusion experiments performed at Mont Terri since 1998 and specifications are summarized in Figure 3 and Table 1.

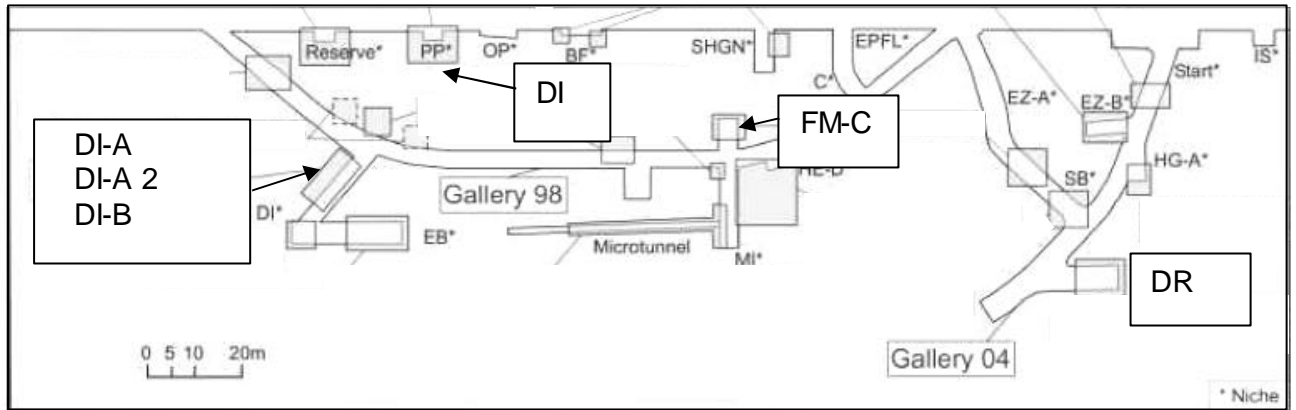


Fig. 3: Location of diffusion experiments

	DI ^a	FM-C ^b	DI-A ^c	DI-A2 ^d	DI-B ^e	DR
Facies	Shaly	Shaly & major fault zone	Shaly	Shaly	Shaly	Shaly
Orientation of Bh axis / bedding	56°	/	58°	60°	58°	90°
Hydro-test K _H (m.s ⁻¹)	No	No	3.10 ⁻¹³	2.4.10 ⁻¹³	No	6.10 ⁻¹³
Tracers	³ H, I	³ H, I (+He)	³ H, I, ²² Na, Cs	³ H, I, ²² Na, Cs, Br, Eu, ⁸⁵ Sr, ⁶⁰ Co	² H, I, ⁶ Li	³ H, I, ²² Na, ⁸⁵ Sr, Br, ⁷⁵ Se & ¹⁸ O, ⁶⁰ Co, ¹³⁷ Cs, ¹³³ Ba, ² H, ¹⁵² Eu
Start of test	Dec 98	June 2000	Jan 2002	Apr 2004	Sept 2002	April 2006
Duration	12 months	8 months	10 months	12 months	13 months	> 24months

Table 1: Main specifications of diffusion experiments performed at Mont Terri. Ref. ^a Palut et al. (2002); ^b Gomez-Hernandez et al. (2004); ^c van Loon et al. (2004b); ^d Wersin et al. (2005); ^e Yllera et al. (2004).

For technical constraints, due to the overcoring operation, most of boreholes are drilled vertically, *i.e.* inclined with regard to the bedding plane. Only the last DR experiment is performed perpendicular to the bedding in an inclined borehole.

Since the first DI experiment, the tracer cocktails have successively become more complex in terms of their chemical behaviour and now include very strongly-sorbing and redox-sensitive tracers. As certain tracers are injected as radio-isotopes, on-line measurements have been carried out with γ detectors (DIA-2 & DR).

4 RESULTS AND CONCLUSIONS

Figure 4 gives an example of both the tracer evolution in the circulating fluid in the interval vs time and of a tracer profile in the rock.

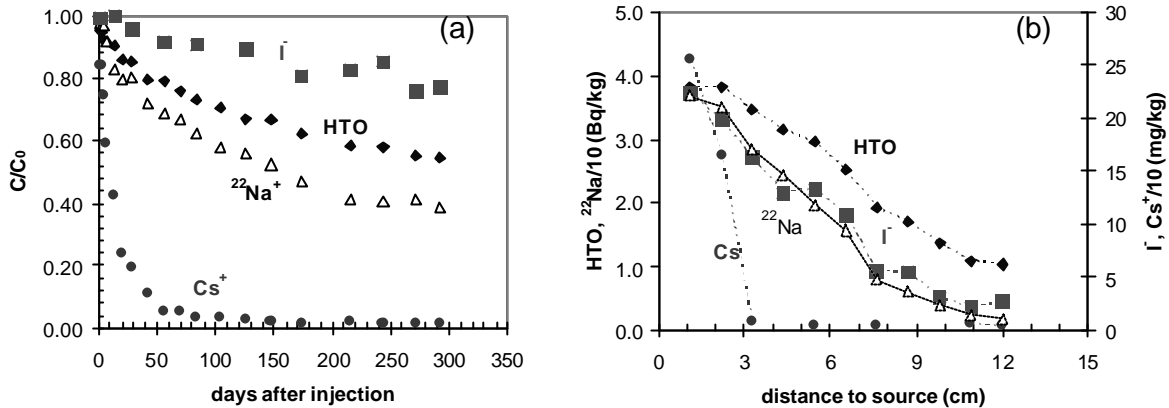


Fig. 4: Evolution of tracer concentrations (a) in interval vs time and (b) in rock vs distance to source (DI-A, Wersin et al., 2004).

The observed tracer patterns follow the generally expected trend: the flux of anions (bromide or iodide) is lower than that of HTO due to anion exclusion effects and their penetration depth in rock can reach some 15 cm, while HTO can penetrate up to some 20 cm after 1 year. The analysis of DI data revealed the occurrence of a disturbed zone around borehole in which the tracer diffusivities were higher. This disturbance was attributed to an oxidation/de-saturation process occurring for 1 month after the drilling and before installation of completion. Concerning the cations, one observed that when the interaction of these species with clay surface was stronger, their decrease relative to HTO in interval was faster and their penetration depth was lower (max 3 cm after 10 months for Cs^+). Analyses of such thin profiles in rock have necessitated the development of new approaches such as micro-spectroscopic techniques that led to high-quality data consistent with bulk data obtained from sliced rock samples.

Figure 5 shows a comparison between published data derived from in-situ and lab experiments. They are limited to HTO, iodide and Na-22. Lab data obtained at 23°C were recalculated at 14°C (the temperature of the water in the circulation system of DI-A) using the measured apparent activation energies for diffusion. It appears that *in situ* data were found to be consistent with small-scale lab diffusion experiments performed parallel to the bedding plane. The new DR experiment will address the measurement of diffusion anisotropy *in situ* by using both shorter injection intervals and a borehole normal to the bedding planes. Concerning the cations, modelling of *in situ* data led to higher diffusivities than HTO as already noted at lab scale.

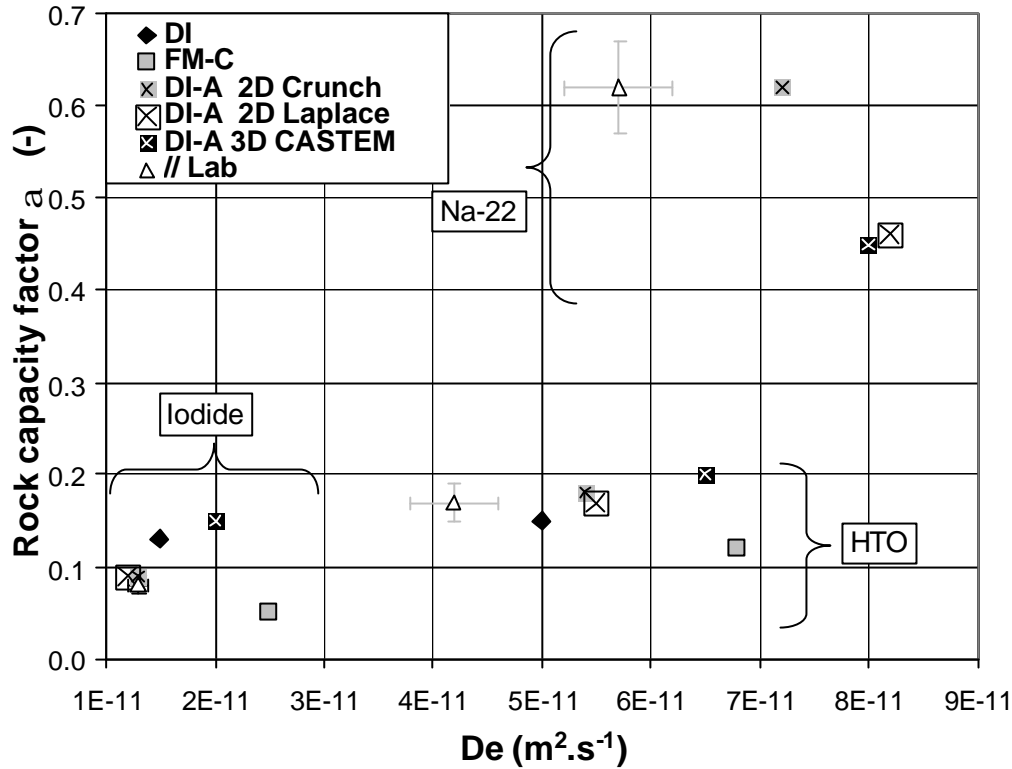


Fig. 5: Rock capacity factor a vs effective diffusion coefficient De from small-scale through-diffusion experiments (lab) and several *in situ* experiments. Crunch and Castem correspond to numerical codes and Laplace means “analytical solution via Laplace transforms”.

All these *in situ* experiments have proven to yield suitable results and to estimate the scale effect from lab to *in situ*-conditions.

5 REFERENCES

Gomez-Hernandez, J.J., Guardiola-Albert, C. (2004). Flow mechanism Experiment (FM-C): Three dimensional model predictions of tracer evolution of HTO and iodine in the main fault. Mont Terri Technical Report-2004-08.

Palut, J.-M., Montarnal, Ph., Gautschi, A., Tevissen, E., Mouche, E. (2003). Characterisation of HTO diffusion properties by an *in-situ* tracer experiment in Opalinus clay at Mont Terri. *J. Contam. Hydrol.* **61**, 203-218.

Patriarche, D., Michelot, J.-L., Ledoux, E., Savoye, S. (2004). Diffusion as the main process for mass transport in very low water content argillites : 2. Fluid flow and mass transport modeling. *Water Resour. Res.* **40**, W01517, doi : 10.1029/2003 WR0022700

Savoye, S., Michelot, J.-L., Witterbrodt, C., Altinier, M. V. (2006). Contribution of the exchange diffusive method to the characterization of pore-water in the consolidated argillaceous rocks. *J. Contam. Hydrol.* **86**, 87-104.

Savoye, S., Michelot, J.-L., Witterbrodt, C. (*In press*). Evaluation of the reversibility of iodide uptake by argillaceous rocks by the radial diffusion method. *In Radiochimica Acta*.

Tevisen, E., Soler, J.M. (2003). Mont Terri Project. DI Experiment. Synthesis Report. Mont Terri Project, Technical Report TR 2001-05.

van Loon, L.R., Wersin, P., Soler, J.M., Eikenberg, J., Gimmi, Th., Hernán, P., Dewonck, S., Savoye, S. (2004a). In-situ diffusion of HTO, $^{22}\text{Na}^+$, Cs^+ and I^- in Opalinus Clay at the Mont Terri underground rock laboratory. *Radiochim. Acta* **92**, 757-763.

van Loon, L.R., Soler, J.M., Müller, W., Bradbury, M.H. (2004b). Anisotropic diffusion in layered argillaceous rock: A case study with Opalinus Clay. *Environmental Science and Technology*, **38** : 5721-5728.

Wersin, P., van Loon, L.R., Soler, J.M., Yllera, A., Eikenberg, J., Gimmi, Th., Hernán, P., Boisson, J.-Y. (2004). Long-term diffusion experiment at Mont Terri: first results from field and laboratory data. *Applied Clay Science* **26**, 123-135.

Wersin, P., Gimmi, Th., van Loon, L.R., Soler, J.M., Dewonck, S., Eikenberg, J., Baeyens, B., Hernán, P. (2005). Diffusion of HTO, Br^- , I^- , Cs^+ , $^{85}\text{Sr}^{2+}$, $^{60}\text{Co}^{2+}$, and Eu^{3+} in a clay formation: first results from in-situ experiment in Opalinus clay. Migration 2005, Avignon, France, 199.

Yllera, A., Hernández, A., Mingarro, M., Quejido, A., Sedano, L.A., Soler, J.M., Samper, J., Molinero, J., Barcala, J.M., Martín, P.L., Fernández, M., Wersin, P., Rivas P., Hernán, P. (2003). DI-B experiment: planning, design and performance of an in-situ diffusion experiment in the Opalinus Clay formation. *Applied Clay Science* **26**, 181-196.