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Safety-relevant hydrogeological properties of the claystone barrier of a Swiss radioactive waste repository: An evaluation using multiple lines of evidence

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Abstract In Switzerland, the Opalinus Clay – a Jurassic (Aalenian) claystone formation – has been proposed as the first-priority host rock for a deep geological repository for both low- and intermediate-level and high-level radioactive wastes. An extensive site and host rock investigation programme has been carried out during the past 30 years in Northern Switzerland, comprising extensive 2D and 3D seismic surveys, a series of deep boreholes within and around potential geological siting regions, experiments in the international Mont Terri Rock Laboratory, compilations of data from Opalinus Clay in railway and motorway tunnels and comparisons with similar rocks.

The hydrogeological properties of the Opalinus Clay that are relevant from the viewpoint of long-term safety are described and illustrated. The main conclusions are supported by multiple lines of evidence, demonstrating consistency of conclusions based on hydraulic properties, porewater chemistry, distribution of natural tracers across the Opalinus Clay as well as small- and large-scale diffusion models and the derived conceptual understanding of solute transport.

Keywords Opalinus Clay · Hydraulic conductivity · Diffusion · Porewater · Natural tracers · Self-sealing

Sicherheitsrelevante hydrogeologische Eigenschaften der Tonsteinbarriere eines Schweizer Tiefenlagers für radioaktive Abfälle: Eine Evaluation anhand mehrfacher Argumentationslinien

Zusammenfassung In der Schweiz wurde der Opalinuston – eine jurassische (Aalenium) Tonstein-Formation – als prioritäres Wirtgestein für geologische Tiefenlager (Endlager) für radioaktive Abfälle vorgeschlagen, sowohl für schwach- und mittelaktive Abfälle, wie auch für hochaktive Abfälle. Während der letzten 30 Jahren wurden in der Nordschweiz umfangreiche Standort- und Wirtgesteinsuntersuchungen durchgeführt, mit 2D- und 3D-reflexionsseismischen Untersuchungen, einer Serie von Tiefbohrungen in und im Umfeld der potenziellen geologischen Standortgebiete, Experimenten im internationalen Felslabor Mont Terri und Kompilationen von Daten aus Opalinustonstrecken in Straßen- und Eisenbahntunneln.

Die hydrogeologischen Eigenschaften des Opalinustons, die unter dem Gesichtspunkt der Langzeitsicherheit relevant sind, werden beschrieben und veranschaulicht. Die wichtigsten Schlussfolgerungen sind durch mehrere Argumentationslinien unterstützt und zeigen ein konsistentes Gesamtbild von hydraulischen Eigenschaften, Porenwasserchemie, Verteilung natürlicher Tracer sowie klein- und großmaßstäblicher Diffusionsmodelle und den abgeleiteten konzeptionellen Modellvorstellungen.

Introduction

In Switzerland, the Nuclear Energy Act requires the disposal of all radioactive waste in deep geological repositories. The Swiss programme foresees two types of reposi-

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tories: one for low- and intermediate-level waste (L/ILW) and one for high-level waste (HLW) consisting of spent fuel, vitrified high-level waste and long-lived intermediate-level waste with the option to dispose of all wastes in one repository – the so-called combined repository. The engineered barrier system for L/ILW and for long-lived ILW is concrete-based and, in the HLW repository, the disposal canisters (carbon steel or copper-coated carbon steel) are surrounded by compacted bentonite (Nagra 2016).

The procedure for selecting the repository sites is defined in the Sectoral Plan for Deep Geological Repositories (Swiss Federal Office of Energy 2008) and is managed by the Swiss Federal Office of Energy. Nagra, the Swiss National Cooperative for the Disposal of Radioactive Waste, is the responsible implementer and conducts all necessary site investigations, safety assessments and studies related to construction feasibility. The Sectoral Plan foresees the selection of sites in three stages, the third stage leading to the granting of a general licence. Stage 1 of the Sectoral Plan concluded with the proposal of four different sedimentary host rocks for the L/ILW repository; the Opalinus Clay formation in Northern Switzerland was selected for the HLW repository (Nagra 2008a, 2008b). Nagra also proposed three geological siting regions for the HLW repository and six for the L/ILW repository; all siting regions and potential host rocks were approved by the Federal Government in November 2011. Currently, the envisaged repository depth in these sites is between 400 and 900 m. The minimum depth is related to possible erosion scenarios in the siting region, the maximum depth to issues in the context of construction feasibility (for details see Nagra 2014a and ENSI 2017).

The ongoing Stage 2 of the Sectoral Plan aims at narrowing down the potential geological siting regions. Nagra's proposals were submitted and published by the SFOE in January 2015 (Nagra 2014a, 2014b, 2014c). Of the six L/ILW regions and three HLW regions identified in Stage 1, Nagra proposed the two regions, Jura Ost and Zürich Nordost, for further investigation (Fig. 1). The other four siting regions would also meet the safety requirements, but they were placed in reserve because in relative terms they showed clear disadvantages from a safety viewpoint. The disadvantages include limited length of migration pathways, challenges due to great depth of the repository and less favourable situation with respect to erosion and neotectonics. ENSI (the Swiss Federal Nuclear Safety Inspectorate) and its experts reviewed the submitted documents (ENSI 2017). ENSI approved Nagra's proposals for: (a) the choice of Opalinus Clay as the host rock for both repository types, (b) the two regions for further investigation in Stage 3, (c) placing three of the remaining four regions in reserve, but (d) recommended to include in addition Nördlich Lägern in Stage 3 (Fig. 1). The review continues with a review by federal commissions and a broad public consultation. A final decision by the Swiss Federal Council on the result of Stage 2 is expected at the end of 2018.

In Stage 3, the remaining sites will be investigated in detail, with 3 to 5 deep boreholes per site comprising extensive test programmes and 3D seismic surveys.

Lithology, mineralogy and porosity of the Opalinus Clay

The Jurassic (Aalenian) Opalinus Clay formation – named after the ammonite *Leioceras opalinum* – was deposited in an epicontinental shallow marine environment (Wetzel and Allia 2003) and has a thickness of 80–130 m in boreholes in Northern Switzerland. It consists of dark grey to black, partly silty claystones that are subdivided into several lithostratigraphic units based on the content of thin calcareous silty-sandy lenses or siderite nodules (Nagra 2002a). Compared to other Mesozoic sedimentary rocks in the area, it is a very homogeneous formation with rather small vertical and lateral lithological variability. At the decimetre to millimetre scale, the preferred alignment of platy clay particles is responsible for a distinct fabric ('bedding') which causes anisotropic hydraulic and mechanical properties.

The average clay mineral content of the Opalinus Clay is 59 ± 12 (mean value in wt.-% ± 1 sigma, for details see Nagra 2014b, Dossier VI), consisting of illite (22 ± 8%), illite/smectite mixed layers (13 ± 7%), kaolinite (19 ± 6%) and chlorite (7 ± 3%). Other main components are quartz (20 ± 6%) and calcite (14 ± 10%, partly bioclasts, partly diagenetic cement); siderite and feldspars are in the range of a few %. Important accessory components are pyrite (1.2 ± 0.9%), dolomite/ankerite (0.6 ± 0.2%) and organic carbon (0.7 ± 0.4%).

The Opalinus Clay was subject to a complex burial history (Mazurek et al. 2006), during which a maximum depth of 1650–1700 m was reached in the area of the potential siting regions in Northern Switzerland in the Swiss Molasse Basin and the Tabular Jura, and approximately 1350 m in north-west Switzerland in the area of the Mont Terri underground rock laboratory (URL) located in the Jura Fold-and-Thrust Belt (Fig. 1).

The porosity of the Opalinus Clay reflects the burial history, with values in the range of 11–12% in the area of the potential siting regions with deeper maximum burial and approx. 12–18% in the Mont Terri URL with somewhat shallower maximum burial. The porosity values are based on water-loss measurements at 105 °C or on bulk and grain density measurements (definitions and nomenclature of porosities in clay rocks are addressed in more detail in Nagra 2014b, Dossier VI, Ch. 2.3.1.1). Pore sizes (equivalent pore radii) are mainly in the range of 1–25 nm (Nagra 2002a, Ch. 5.4 and Nagra 2014b, Dossier VI, Ch. 2.3).

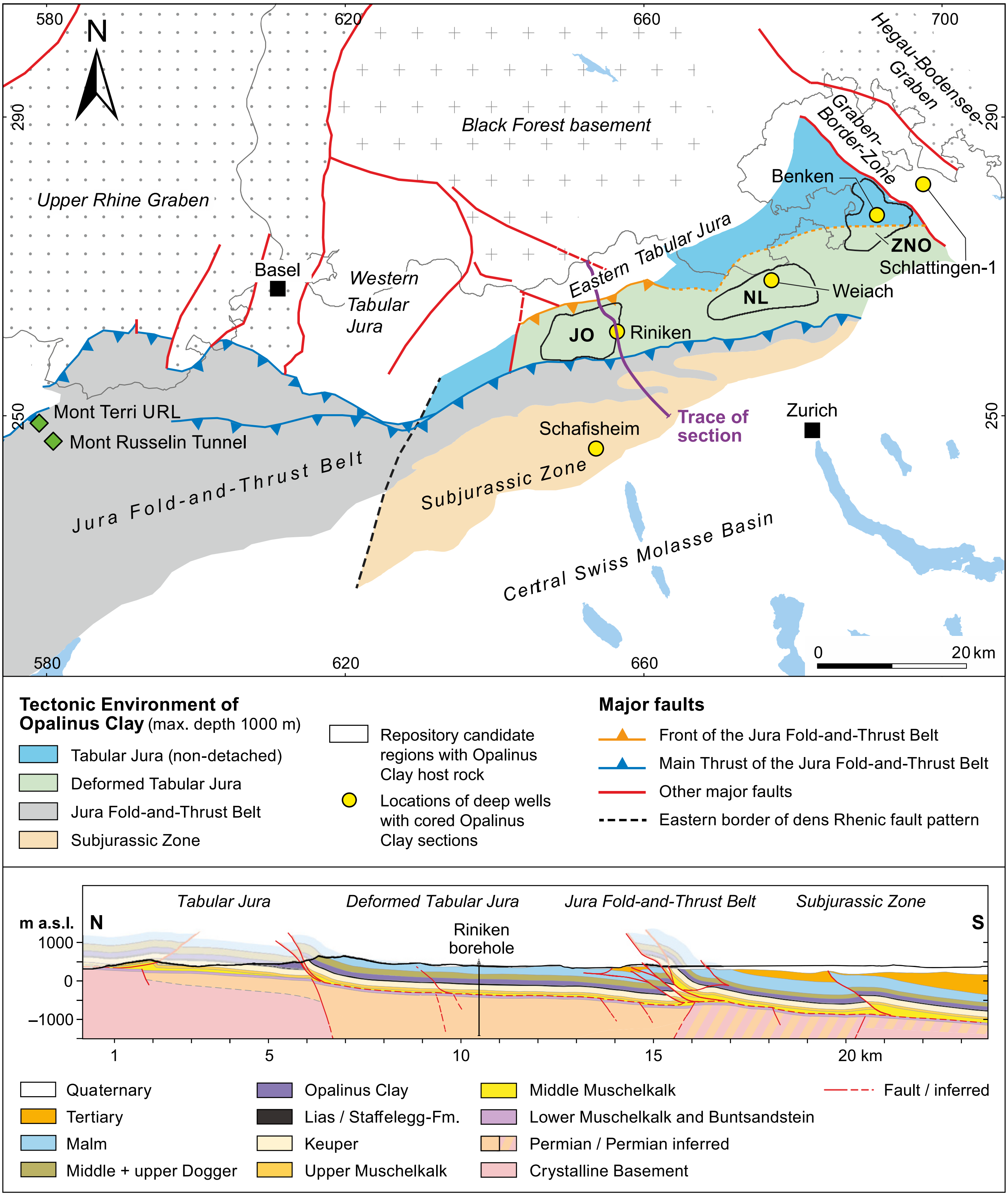


Fig. 1 Tectonic environment of the Opalinus Clay in Northern Switzerland (modified after Nagra 2014b) and locations of deep boreholes and tunnels with investigations of Opalinus Clay. Candidate siting regions: ZNO Zürich Nordost, NL Nördlich Lägern, JO Jura Ost

Porewater geochemistry

Origin of Opalinus Clay porewaters

Diffusion has been shown to be the dominant mechanism of solute transport in the Opalinus Clay and in other clay-rich formations (Gimmi et al. 2007; Mazurek et al. 2011). Driven by concentration gradients, the porewater of the Opalinus Clay adjusts slowly to changing conditions in the adjacent aquifers. While the characteristics of mobile groundwaters in aquifers may change rapidly in response to evolving surface conditions and climate, porewaters in clay-rich formations are geochemical archives with a much longer memory. Thus, the porewater composition in the Opalinus Clay is, on the one hand, a result of chemical interaction (buffering) with solid phases and, on the other hand, it reflects the palaeo-hydrogeological evolution in the embedding aquifers since deposition of the formation at 174 Ma (e. g. Nagra 2002a, Ch. 4.8; Pearson et al. 2003). The most thorough attempt to reconcile all available hydrogeochemical and isotopic data of the Opalinus Clay in the Mont Terri/Mont Russelin area is provided by Mazurek and De Haller (2017) by unravelling the complex palaeo-hydrogeological evolution in great detail. It is shown that Opalinus Clay porewaters are not simple binary mixtures of seawater and meteoric water, but their chloride and stable water isotope signatures can potentially be explained by the addition of a component of partially evaporated seawater, followed by mixing with meteoric water. While the Bromide/Chloride ratios of the porewaters in the Mont Terri/Mont Russelin area are in the range of modern seawater, the ratios of Opalinus Clay porewaters in Northern Switzerland do not have a marine signature (Pearson and Waber 2001). This indicates a different palaeohydrogeological evolution of the Mont Terri/Mont Russelin and Northern Switzerland realms.

Methodology for deriving a 'reference porewater' for safety assessment

A thorough understanding of the porewater chemistry of a host rock is indispensable for the assessment of radionuclide solubility and retention. However, the extremely low permeability of claystones places limitations on the artefact-free sampling of porewaters. For this reason, the complete composition of a clay porewater cannot be derived directly from measured concentrations, a measured pH value and partial pressure of CO_2. While measured concentrations of conservative (non-reactive) species, such as chloride, are considered to closely represent in-situ values, assumptions on the saturation state with respect to relevant minerals (calcite, dolomite, celestite, quartz, fluorite, pyrite, siderite) are made for non-conservative (reactive) species. A few parameters need to be estimated based on *expert opinion*, such as the partial pressure of CO_2. The measured occupancy and selectivity of the cation exchange site population on clay minerals is directly related to the cation proportions in the porewater and can also be taken into account in the geochemical models.

A fundamental understanding of the porewater chemistry of the Opalinus Clay was built up through extensive investigations in the Mont Terri Rock Laboratory and investigations on drillcores in the laboratory (Pearson et al. 2003).

Based on this, and taking into account data from cores of the Benken borehole which is located in the Zürich Nordost siting region (Nagra 2002a, Ch. 5.12), Mäder (2009) used geochemical model calculations to define a so-called 'reference porewater' as the basis for the safety assessment, covering the possible variation of porewaters in the Opalinus Clay of Northern Switzerland, as well as the higher salinity porewaters observed in the Mont Russelin tunnel mentioned below (Table 1). In order to reduce the uncertainties, Pearson et al. (2011) emphasised the need to improve geochemical modelling efforts, e. g. by a more detailed identification of controlling phases in the Opalinus Clay (including clay minerals and redox-sensitive elements) together with an evaluation of their thermodynamic properties or by the measurement of the composition and the stability constants of clay minerals that are present in the formation.

Porewater salinity, chloride content

An intrinsic property of claystones lies in the electrostatic interactions of the permanently negatively charged clay surfaces with dissolved substances; this results in a reduction of the porosity accessible to anions (e. g. Appelo and Wersin 2007; Altmann et al. 2012). Details on this topic in the context of the Opalinus Clay and implications for porewater sampling are summarised and discussed in Wersin et al. (2013), Van Loon (2014) and Mazurek et al. (2015).

For the porewater salinities of the Opalinus Clay in Northern Switzerland, the proportion of the anion-accessible pore space is around 50% of the total pore space, while the other half is anion-free (bound) water.

The salinity of the porewater in the Opalinus Clay varies both in the vertical dimension and, with much lower gradients, also laterally. It depends strongly on the evolution of the salinity in the aquifers above and below (see also section on 'natural tracer profiles'). For the 'reference porewater' of the Opalinus Clay in Northern Switzerland, the maximum chloride value from the Benken borehole was selected ($0.16\,\text{mol/kg} \approx 6\,\text{g/kg}$). This is less than half of the maximum content in the Mont Terri Rock Laboratory ($0.4\,\text{mol/kg} \approx 14\,\text{g/kg}$; Pearson et al. 2003; Mazurek et al. 2011) and much less than the maximum chloride content

Table 1 Porewater compositions of the Opalinus Clay (Mäder 2009; updated for Nagra 2014b, Dossier VI, also taking into account fluorite saturation)

		Reference (Benken)	High pCO_2 (Benken)	Low pCO_2 (Benken)	High salinity (Mont Russelin)
pH	–	7.20	7.00	7.50	7.01
pε	–	–2.78	–2.55	–3.13	–2.56
Eh	[mV]	–164	–150	–185	–151
Ionic strength	[mol/kg$_{H_2O}$]	0.23	0.23	0.23	0.76
log pCO_2	[bar]	–2.20	–1.80	–2.80	–2.50
Na	[mol/kg$_{H_2O}$]	1.64E-01	1.65E-01	1.64E-01	5.28E-01
K	[mol/kg$_{H_2O}$]	2.60E-03	2.62E-03	2.60E-03	1.77E-03
Mg	[mol/kg$_{H_2O}$]	9.65E-03	9.77E-03	9.55E-03	3.76E-02
Ca	[mol/kg$_{H_2O}$]	1.25E-02	1.27E-02	1.24E-02	4.97E-02
Sr	[mol/kg$_{H_2O}$]	2.11E-04	2.12E-04	2.09E-04	4.22E-04
Fe^{II}	[mol/kg$_{H_2O}$]	5.24E-05	5.43E-05	5.09E-05	2.46E-04
Cl	[mol/kg$_{H_2O}$]	1.60E-01	1.60E-01	1.60E-01	6.62E-01
F	[mol/kg$_{H_2O}$]	1.53E-04	1.52E-04	1.53E-04	1.11E-04
S^{VI}	[mol/kg$_{H_2O}$]	2.47E-02	2.47E-02	2.47E-02	2.11E-02
C^{IV}	[mol/kg$_{H_2O}$]	2.51E-03	4.15E-03	1.21E-03	1.06E-03
Si	[mol/kg$_{H_2O}$]	1.78E-04	1.78E-04	1.79E-04	1.72E-04

observed at Mont Russelin of 0.66 mol/kg ≈23 g/kg (Koroleva et al. 2011). More recent data from the Schlattingen-1 borehole in Northern Switzerland (Wersin et al. 2013) show similarly moderate salinities as those in Benken. It is expected that the salinities in the Opalinus Clay of the potential siting regions in Northern Switzerland will not significantly exceed those of the reference porewater of Benken. Nevertheless, a highly saline variant of the reference porewater based on the situation at Mont Russelin (Table 1) takes into account the uncertainty related to the salinity evolution in the aquifers overlying and underlying the Opalinus Clay. This uncertainty also affects the predictability of the vertical salinity profiles across the region of interest.

Redox conditions

Redox conditions strongly affect the mobility and sorption of redox-sensitive radionuclides in a nuclear waste repository, such as uranium, neptunium, plutonium, technetium and selenium. Generally, reducing conditions are favourable with respect to suppressing solubilities and promoting retention of radionuclides (e. g. Altmaier and Vercouter 2012; Berner 2014a, 2014b, Baeyens et al. 2014a, 2014b).

During repository construction and operation, oxygen will be introduced into the repository system. After a relatively short initial oxic phase (for the Swiss HLW repository <1 year, Garitte et al. 2016), the conditions will become and remain reducing due to iron corrosion and pyrite oxidation in the repository near-field, provided that reducing conditions prevail in the undisturbed pristine clay host rock (Wersin et al. 2003; Bradbury et al. 2014).

Determining the redox state and redox-active reactions of porewaters in clay rocks is not straightforward. Redox measurements in the Opalinus Clay in the Mont Terri URL were carried out over nearly 20 years. Pearson et al. (2003) summarise the early experience, while later developments and the improvement in the understanding of the measurement results are documented in Wersin et al. (2011a, 2011b) and in a series of papers in a special issue of Applied Geochemistry (Volume 26, Issue 6, June 2011). The most challenging issues in measuring redox conditions are O_2 gas contamination at trace level in laboratory glove boxes, glycerol release from the gel filling of reference electrodes or electrode drift, unknown or not measured redox-sensitive species and co-existing redox couples providing mixed redox potentials; the understanding and the modelling of the redox conditions can thus be very challenging. There is also a correlation between pH and redox potential, depending on the specific redox-active chemical couple, which is implicit in the interpretative and predictive modelling of redox.

Despite trying several approaches to collect in situ pH and redox potential data, none of them were entirely satisfactory (Pearson 2009). For this reason, Eh values were calculated assuming that ferrous iron is controlled by pyrite and siderite equilibria for an upper and lower limit of CO_2 partial pressure (or pH respectively) and for a given (measured) sulphate content (Mäder 2009) (Table 1). These are thought to be relatively robust assumptions.

Radionuclide solubility and retention

The long-term safety of radioactive waste repositories in clay rocks relies mainly on a very low solubility of the waste form and an effective interplay between slow advection and diffusion, as well as strong nuclide retention. For the Swiss case, the solubilities and sorption coefficients of all safety-relevant radionuclides have been evaluated during Phase 2 of the Sectoral Plan (Berner 2014a, 2014b; Baeyens et al. 2014a, 2014b).

Under reducing conditions, the UO_2 (and PuO_2) pellets in the spent fuel have very low solubilities, and, because of the very low quantities of mobile porewater present in the engineered bentonite barrier and in the Opalinus Clay, the masses dissolved are likely to be exceedingly small (Bradbury et al. 2014), i. e. except for the instant release fraction (IRF), the fission products trapped in the pellets are only released very slowly. The IRF consists of those radionuclides that are released from spent fuel immediately upon canister breaching. It is the combined release of radionuclides from grain boundaries and cracks of the spent fuel pellets and from the gap between the pellets and the cladding (Nagra 2002b, Fig. 4.5-2).

Under reducing conditions, the most radiotoxic actinides have very low solubilities and they are also strongly sorbed on clay minerals in the bentonite backfill and in the clay-rich host rock. In combination with slow advection and diffusion (see sections below), sorption will retard transport of many positively charged ionic species of radionuclides until complete decay within a few metres distance from the waste canister surface (e. g. Nagra 2002b, Fig. 6.6-2; Grambow 2016). Only negatively charged long-lived radionuclide ions (^{129}I, ^{79}Se, ^{36}Cl) or neutral species (^{14}C in small organic molecules) can migrate through the natural barrier system because they are non-sorbing or weakly sorbing (Nagra 2002b). These nuclides also determine the annual individual radiation dose to the local population, which is quantified by dose calculations for different scenarios of a safety case (Nagra 2002b).

Hydraulic properties

Hydraulic packer tests in boreholes and laboratory tests on cores

An extensive database is available for the Opalinus Clay from hydraulic packer tests in boreholes (Fig. 2). Data are available from three Nagra deep boreholes in Northern Switzerland (data compilation in Nagra 2010, Tab. A3-1), from numerous boreholes drilled in all directions in the Mont Terri Rock Laboratory (Jaeggi et al. 2014; Yu et al. 2017) and from shallow boreholes in the Swabian Alb in Germany (Hekel 1994). The hydraulic conductivity of the Opalinus Clay is depth-dependent (Fig. 2). Near the surface, it can have K values up to 10^{-4} m/s due to decompaction effects and weathering processes. At depths between 10 and 30 m, K decreases by several orders of magnitude. This decrease correlates with a marked increase in the chloride content of the porewaters which confirms that slow groundwater movement has persisted for a very long period (Hekel 1994). There are no data from hydraulic tests in boreholes for depths between 50 and 250 m. The most comprehensive and reliable dataset comes from the Benken exploratory borehole where the Opalinus Clay was encountered in a depth of 539–652 m (Nagra 2001; Beauheim 2013). It shows consistent K values with little scatter, typically between $2 \cdot 10^{-14}$ and $1 \cdot 10^{-13}$ m/s. These values are more than 1000 times lower than the minimum requirements for the hydraulic conductivity of host rocks for HLW in the German and the Swiss site selection programmes: $K < 10^{-10}$ m/s (AkEnd 2002, Tab. 4.1; Kommission Lagerung hochradioaktiver Abfallstoffe 2016, Kap. 6.5.5.1), $K_V \leq 10^{-10}$ m/s (Nagra 2008b, Tab. 4.2-1). In the German programme K values $< 10^{-12}$ m/s are classified as 'favourable' (AkEnd 2002, Tab. 4.3; Kommission Lagerung hochradioaktiver Abfälle 2016, Tab. 25).

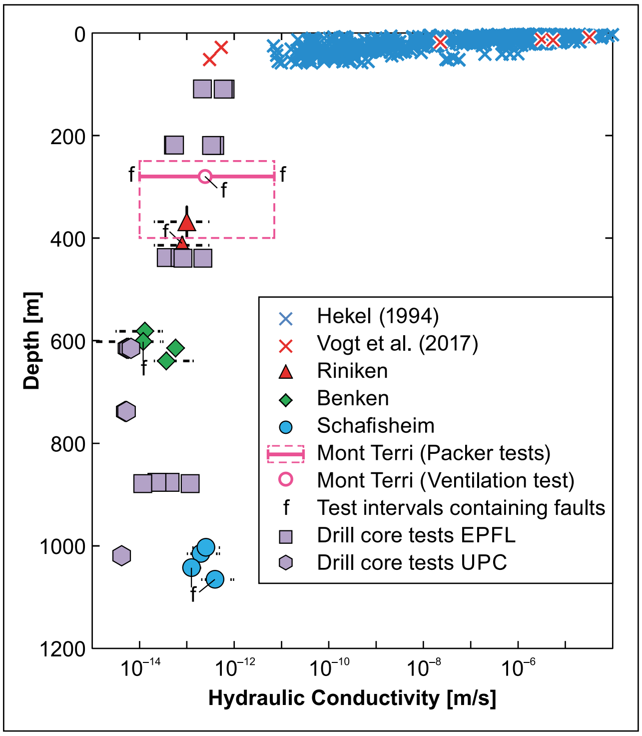


Fig. 2 Hydraulic conductivity of the Opalinus Clay in Northern Switzerland versus depth: Results from double packer tests in boreholes, from a large-scale ventilation test in the Mont Terri URL and from selected laboratory tests on drillcores (average experimental confining pressure converted to depth). EPFL samples: Ferrari et al. (2012), UPC samples: Romero and Gómez (2013)

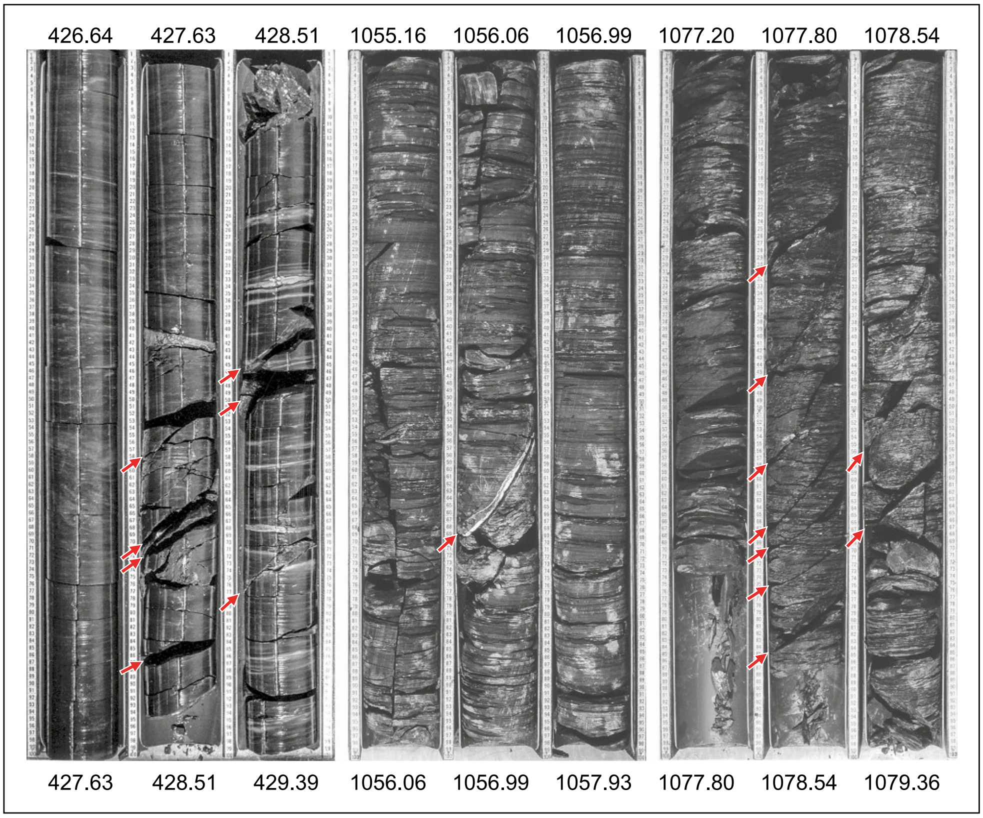


Fig. 3 Cores of faulted sections in the Opalinus Clay in the Riniken (*left*) and Schafisheim (*right*) boreholes. The depth of the cores is indicated above/below the cores. *Red arrows:* Fault planes. The hydraulic conductivity measured by double packer tests in these sections is comparable with tests in undisturbed sections (cf. Fig. 2)

In the Opalinus Clay of the Mont Terri Rock Laboratory, which has a higher porosity compared with Benken due to lower maximum burial (see above), K tends to be somewhat higher and reaches values up to $7 \cdot 10^{-12}$ m/s (Jaeggi et al. 2014; Yu et al. 2017) (Fig. 2). Values $\geq 10^{-11}$ m/s may reflect relaxation of stresses around underground structures. Results from hydraulic tests in an inclined borehole crossing the entire Opalinus Clay (Yu et al. 2017) show higher values (in the order of 10^{-12} m/s) for the Opalinus Clay shaly facies than for its sandy facies (in the order of 10^{-13} m/s), which is attributed to diagenetic cementation of the latter. Such a trend cannot be recognised in a compilation of previous tests (Jaeggi et al. 2014; Fig. 18).

In many of the boreholes in the Mont Terri Rock Laboratory and in the deep boreholes of Northern Switzerland (Benken, Riniken, Schafisheim), intervals with large and small fault zones were also hydraulically tested (for example at Riniken and Schafisheim, see Fig. 3). The K values of these intervals do not differ significantly from those of undisturbed sections (Fig. 2) and are evidence of the excellent self-sealing capacity of the Opalinus Clay (Bock et al. 2010).

Numerous hydraulic conductivity measurements on cores parallel and perpendicular to bedding have been performed in the laboratory under various confining pressures (selected values are shown in Fig. 2). All data and the corresponding references are compiled in Nagra (2014b, Dossier VI, Table 2.5-2). An anisotropy in hydraulic conductivity due to bedding is largely the result of microscopic heterogeneity (orientation of clay platelets, diagenetic cementation of thin silty and sandy layers). The anisotropy coefficient varies in the order of 1–10, depending on the sedimentary fabric (Nagra 2002a, Ch. 5.8). An average value of 5 ($K_H = 5 \times K_V$) was chosen as the reference value for radionuclide transport models for safety case considerations at formation scale (Nagra 2002a, Table 9.3-2, 2014b, Dossier VI, Ch. 4.7.2.1).

Large-scale ventilation test

A long-term ventilation test has been performed in the Mont Terri URL in a 10 m long unlined microtunnel with a diameter of 1.3 m. The test can be considered as a large-scale pumping test in which the humidity increase of injected dry air, i. e. the outflow rate of vapour, is measured. An average hydraulic conductivity of $2.5 \cdot 10^{-13}$ m/s of the tested block of Opalinus Clay, including several small faults, was estimated using appropriate models (Mayor et al. 2007; value plotted in Fig. 2).

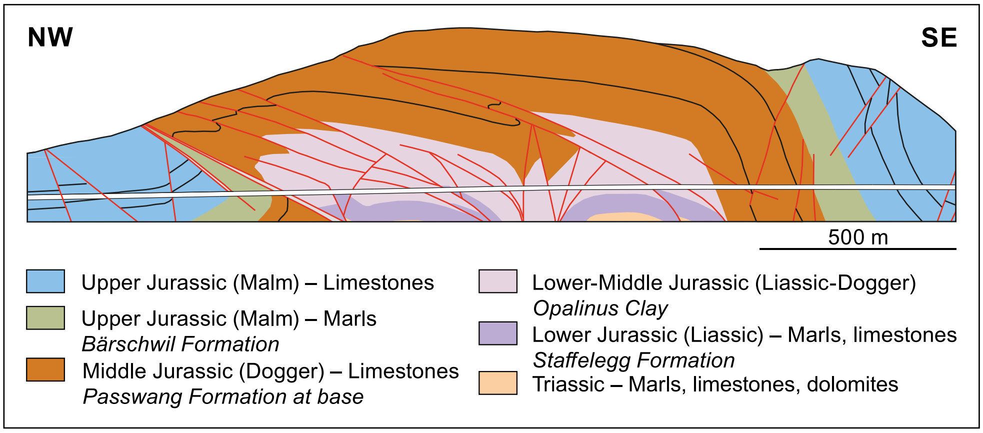


Fig. 4 Geological cross-section along the Mont Russelin highway tunnel (extract from Fig. 1 of Mazurek and De Haller (2017), after Bureau Technique Norbert (1993)). Despite extensive faulting of the Opalinus Clay, no traces of water inflows have been detected

Hydrogeological observations in underground structures

Hydrogeological observations in more than 6000 m of tunnel sections in the Opalinus Clay, mostly in the Jura Fold-and-Thrust Belt, occasionally show small water inflows at depths of less than 200 m, generally associated with faults (Gautschi 2001). Where information is available, the waters are always saline. No water inflows or damp patches have been reported from sections with an overburden of more than 200 m. The visual recognition of inflows in tunnels depends on various factors such as ventilation, humidity and local hydraulic gradients. The detection limit for transmissivities of discrete inflows at faults in ventilated tunnels is in the range $T = 10^{-9}$–$10^{-10}\,\mathrm{m^2/s}$ (Eugster and Senger 1994); in sections with no or low ventilation (e. g. tunnel end) much lower inflows would probably be detectable.

More recent observations in the Mont Terri Rock Laboratory showed isolated indications of localised water flow in the Opalinus Clay – so-called *wet spots* – with a maximum overburden up to 300 m (Müller and Leupin 2012). Their localisation does not show any systematic correlation with mineralogy, lithology or facies of the Opalinus Clay, and only a partial correlation with brittle structures could be found. The inflow rates in boreholes in different facies types of the Opalinus Clay which intersect *wet spots* are often in the range of a few 10 s of ml/day in the shaly and sandy facies. The inflows delivered porewater for periods of months to years. The inflow rates can also change with time, particularly during excavation work in the rock laboratory. This points to stress redistributions and associated poromechanical effects (e. g. squeezing of water due to local compaction). An exceptional inflow rate of up to 450 ml/day was measured in a sandy carbonate-rich facies which has not been observed in the siting regions in Northern Switzerland (Vogt 2013). The chemical composition of the porewaters of *wet spots* follows the general distribution pattern of the data for Mont Terri porewaters, i. e. they do not represent chemical anomalies (see e. g. Mazurek et al. 2010a, 2010b; Vogt 2013).

In the exploration tunnel of the Mont Russelin highway tunnel crossing the Jura Fold-and-Thrust Belt, more than 1000 m of Opalinus Clay with numerous faults with an overburden of 300–400 m were penetrated by an open mode tunnel boring machine (Fig. 4). During excavation, detailed hydrogeological mapping at the 1:100 scale focusing on indications of even minor water inflow and damp patches was performed (Bureau Technique Norbert 1992). No dripping water or *wet spots* were detected in the Opalinus Clay section. The very low transmissivity of the faults is attributed to a very effective self-sealing capacity of Opalinus Clay at high overburden.

Comparison with data from other clay rocks

In a compilation of hydraulic conductivity data from various rock types (Appel and Habler 2002), values from numerous packer tests in clays/claystones with an overburden >200 m are $\leq 3.3 \cdot 10^{-11}\,\mathrm{m/s}$ (except for one test from 480 m with a value of $2.1 \cdot 10^{-10}\,\mathrm{m/s}$). The hydraulic conductivity values measured in undeformed and deformed Opalinus Clay at depths below the near-surface decompaction zone (Fig. 2) are generally consistent with the experience from other clay rocks.

A compilation of 127 packer tests conducted in sedimentary strata in Nagra's deep boreholes in Northern Switzerland and at the Wellenberg site in the Helvetic Alps in various geological units (Nagra 2009) shows a clear trend of decreasing hydraulic conductivity K with increasing clay mineral content (Nagra 2014b, Dossier VI, Fig. 2.5-7; only data from depths below the near-surface decompaction zone were considered). Specifically, it can be shown that, for clay mineral contents >25 wt.-%, K reduces to values $< 10^{-9}\,\mathrm{m/s}$, and for clay mineral contents of >40 wt.-% to values of $< 10^{-11}\,\mathrm{m/s}$.

Jolley et al. (2007) compiled data from a large number of permeability tests on fault rock in cores from a large depth range from the Brent Province (North Sea) as a function of clay mineral content. All samples with clay contents >40 wt.-% yielded K values $\leq 10^{-12}$ m/s (original permeability data transferred into hydraulic conductivity values, see Nagra 2014b, Dossier VI, Fig. 2.5-8).

Mazurek et al. (2008) show that, for clay rocks, there is a positive correlation between porosity and the logarithm of hydraulic conductivity for porosities larger than ca. 5%. However, for highly compacted clay formations with smaller porosities, the trend is broken and effective hydraulic conductivity may increase sharply. This is probably due to the fact that highly overconsolidated clay rocks lose their plasticity and their swelling potential and hence their self-sealing capacity. In the hydraulic conductivity vs. porosity diagram of Mazurek et al. (2008), the K values of Opalinus Clay plot in the lower range of hydraulic conductivities measured for clay rocks.

Clay-rich confining units above and below the Opalinus Clay as supplementary migration barriers

Above and below the Opalinus Clay, a series of low-permeability clay-rich or evaporitic formations are found between the host rock and the regional Jurassic and Triassic limestone aquifers in Northern Switzerland. Particularly in a vertical, but also in a lateral, direction these confining units act as supplementary radionuclide transport barriers (Nagra 2014b). The confining units locally contain a few metres of thick sandy, calcareous or dolomitic layers that represent potential water-conducting pathways for radionuclides. The effect of the confining units is that radionuclides that may not have decayed during transport through the Opalinus Clay host rock will only reach the biosphere after a further considerable time delay (Nagra 2002b, Ch. 7.4.6).

Diffusion properties

Laboratory and field experiments

Diffusion coefficients for Opalinus Clay were measured in the laboratory using 0.8–2 cm thick discs of rock, taking into account the effect of confining pressure, sample depth, sample orientation and temperature (Van Loon et al. 2003; Van Loon 2014; Van Loon and Mibus 2015). Data are available for HTO (tritiated water = mainly ordinary H_2O plus ^{3}HOH in trace amounts), $^{36}Cl^{-}$, $^{125}I^{-}$, $^{22}Na^{+}$, $^{85}Sr^{2+}$ and $^{134}Cs^{+}$ and relate mostly to transport perpendicular to bedding, with some information on transport parallel to bedding. Besides values for effective diffusion coefficients D_e, the accessible porosity ε for each species can also be derived from the experiments with conservative species. Opalinus Clay data for anions and HTO are shown in Fig. 5 in comparison with data from other clay rocks. Samples from Northern Switzerland show somewhat lower effective diffusion coefficients and porosities than those from the Mont Terri URL, which is attributed to the deeper maximum burial of the Opalinus Clay in Northern Switzerland.

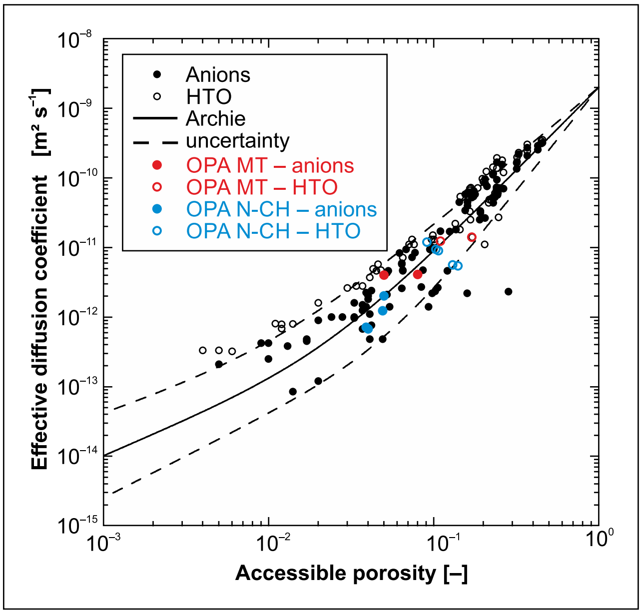
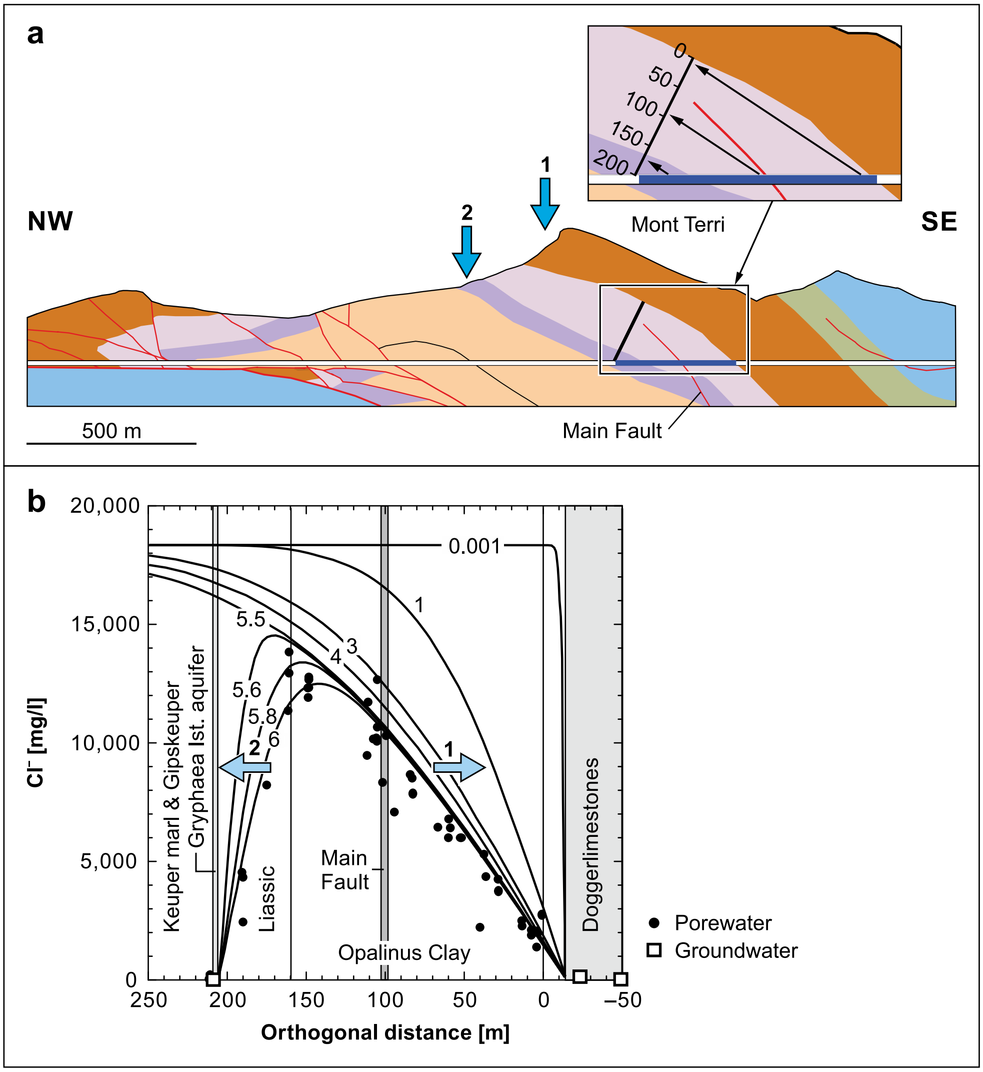


Fig. 5 Relationship between the effective diffusion coefficient (perpendicular to bedding) and the diffusion-accessible porosity for HTO and anions (Cl^{-} and I^{-}). Modified Fig. 3 from Van Loon and Mibus (2015). Highlighted Opalinus Clay samples are from the Mont Terri URL (MT) and from deep boreholes in Northern Switzerland (N-CH), *black symbols* represent values from other clay rocks. The *curves* represent the extended Archie's relation, data were taken from the literature (for details see original paper)

In situ diffusion coefficients for HTO, $^{125}I^{-}$, $^{22}Na^{+}$ and some other species were measured during field experiments in the Mont Terri Rock Laboratory (Van Loon et al. 2004; Wersin et al. 2006, 2008; Soler et al. 2008, 2013); these mostly relate to transport parallel to bedding. In the experiments, which extended over one to four years, diffusion distances of up to 30 cm (depending on species) were reached, i. e. around one order of magnitude more than in the laboratory experiments. The laboratory and in situ values are consistent and therefore do not show any scale dependence in the range 1–30 cm (see compilation of all in situ and lab values in Nagra 2014b, Dossier VI, Tab. 2.6-1). From the numerous measurements perpendicular and parallel to the bedding, an anisotropy factor of around 4 can be derived for the diffusion coefficients. In a recent experiment (Gimmi et al. 2014), anisotropy ratios were determined in field experiments. For HTO and $^{22}Na^{+}$, the anisotropy factor is around 5; for the iodide anion it is around 4.

Fig. 6 Distribution of chloride in the Opalinus Clay and surrounding units and results from a large-scale diffusion model at formation scale (slightly modified after Mazurek et al. (2011), combined with the upper part of Fig. 1 from Mazurek and De Haller (2017)), see explanations in the text. Geological legend see Fig. 4



Natural tracer profiles

From the spatial distribution of porewater tracers (e. g. $\delta^{18}O$, $\delta^{2}H$, Cl^{-}, He) in the Opalinus Clay and in the clay-rich confining units in the Benken borehole, in the Mont Terri URL and in the Mont Russelin tunnel in Switzerland, Gimmi et al. (2007), Mazurek et al. (2009, 2011) and Koroleva et al. (2011) estimated the diffusion times needed for the build-up of the present-day tracer profiles using model calculations. In all cases, the shapes of the profiles can be explained by diffusion acting as the dominant transport process over periods of several hundreds of thousands to several millions of years and at the length scales of the profiles.

The most broadly based case study is that for the chloride profile through the Opalinus Clay in the southern limb of the Mont Terri anticline (Fig. 6a and b). Diffusion coefficients from small-scale laboratory experiments were used for all model calculations. These were performed considering recharge of meteoric water into the upper (Dogger) aquifer (dark blue arrow 1 in Fig. 6a) at a time when the clay-rich formation covering the Dogger aquifer was removed by erosion. At a later stage, when erosion cut through Opalinus Clay elsewhere and exposed underlying strata, recharge of meteoric water in the lower (Liassic) aquifer was initiated (dark blue arrow 2). This means that out-diffusion of chloride from the Opalinus Clay into the upper aquifer started long before that towards the underlying aquifer (light blue arrows 1 and 2 in Fig. 6b). The out-diffusion times into both aquifers were treated as fit parameters in the model calculations (Mazurek et al. 2011). As shown in Fig. 6b, a fit that well reproduces the observed asymmetry of the Cl^{-} contents is obtained when the upper aquifer is activated at 6 Ma and the lower aquifer at 0.5 Ma. These times are within the geologically plausible range of activation times, taking into account the time of the formation of the Jura Fold-and-Thrust Belt and landscape evolution (detailed discussion in Mazurek et al. 2011). Mazurek et al. (2011) also explored effects of additional advection in the direction normal to the contacts to the aquifers on the shapes and evolution times of the tracer profiles. They found that model fits considering continuous advection are worse than those based on diffusion alone, and advection velocities $\geq 4 \cdot 10^{-13}$ m/s in either direction are inconsistent with the data.

Although information concerning the palaeohydrogeological boundary conditions determined by tectonic and landscape evolution does contain uncertainties, it is concluded from the consistency between the calculated build-up times and the independent palaeohydrogeological evidence that the small-scale diffusion coefficients are applicable on the formation scale, and that the permeability of the Opalinus Clay is also very low at this scale.

Summary and conclusions

In Switzerland, the Opalinus Clay formation has been selected as the first-priority host rock for a deep geological repository for both low- and intermediate-level and high-level radioactive waste.

The properties of the Opalinus Clay that are relevant from the viewpoint of long-term safety can be summarised as follows:

- Below a near-surface decompaction zone with elevated hydraulic conductivity (up to 10^{-4} m/s), the hydraulic conductivity is very low ($\sim 10^{-13}$ m/s) and diffusion is the dominant transport mechanism, with advection playing a secondary role at best; clay-rich confining units over- and underlying the Opalinus Clay host rock act as a supplementary barrier to migrating radionuclides.
- Faults in the Opalinus Clay do not represent preferential flow paths, which is attributed to an efficient self-sealing mechanism.
- The self-sealing capacity of the Opalinus Clay minimises the effects of perturbations caused by the repository (excavation damaged zone around underground structures) or by potentially induced or reactivated fractures in the far future.
- Stable, reducing geochemical conditions prevail and, due to its high clay mineral content, the host rock has favourable sorption properties.

The main conclusions are supported by multiple lines of evidence demonstrating consistency among hydraulic properties (tests and observations at various scales, comparisons with data from other clay rocks), porewater geochemistry, laboratory and in situ diffusion experiments, the distribution of natural tracers across the Opalinus Clay as well as small- and large-scale diffusion models and the derived conceptual understanding of solute transport.

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