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## Hydrochemical and isotopic ( $\delta^2\text{H}$ , $\delta^{18}\text{O}$ , $^3\text{H}$ ) characterization of fracture water in crystalline rock (Grimsel, Switzerland)

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### Abstract

Knowledge on water circulation through crystalline rock is of key interest for radioactive waste disposal, geothermal projects or groundwater supply. This study presents first results of long-term monitoring of fracture groundwater chemical and isotope compositions combined with structural data on fracture networks developed in the crystalline rocks at the Grimsel Test Site (GTS), Switzerland. Groundwater circulation in the two major lithologies, the Central Aar Granite (CaGr) and the Grimsel Granodiorite (GrGr) is mainly associated to NE-SW trending brittle deformation structures. Within the different lithologies groundwater chemical and water isotope ( $\delta^{18}\text{O}$ ,  $\delta^2\text{H}$ ) compositions remain constant over the 2 years monitoring period. This suggests the absence of seasonal variations at the sampling depth of 420–520 m below surface. Differences in chemical and isotope composition occur, however, between groundwater collected from the CaGr and GrGr and can be related to differences in infiltration area, flow paths, residence time and water-rock interaction in the two rock units. The  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  signatures indicate a meteoric origin from the same moisture sources under present-day climatic conditions and the absence of measurable  $^3\text{H}$  suggests residence times > 65 yrs for groundwater from both units.

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### 1. Introduction

In crystalline rocks, groundwater circulation is essentially limited to brittle fractures that locally increase the rock permeability and enable advective water flow. Groundwater mineralisation in crystalline rock depends on the water pathway, the interaction with the host rock, the residence time, and mixing of groundwater from different fracture networks. The Grimsel Test Site (GTS) underground rock laboratory (Fig. 1) located in the Grimsel area, Central

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Swiss Alps, and operated by the Swiss National Cooperative for the Disposal of Radioactive Waste (Nagra) offers a unique possibility for studying the evolution of crystalline groundwater in combination with deformation structures developed in the rocks. The bedrock in the rock laboratory and surroundings is made of three different rock types: (i) Grimsel Granodiorite (GrGr), (ii) Central Aar granite (CAGr) and (iii) metabasic dykes<sup>1</sup>. The GrGr and the CAGr form a calc-alkaline differentiation series of Permian age<sup>2</sup> whereas the metabasic dykes are younger and are of various magmatic origins. During Alpine Orogeny, the bedrock was subjected to greenschist metamorphism and ductile as well as brittle deformation<sup>1</sup>. This brittle deformation resulted in ubiquitous brittle fractures and fault zones (Fig. 1) with enhanced permeability. Fault zones mainly strike NE-SW and dip steeply towards S, reaching today's topography, and so crosscut the entire rock mass above the rock laboratory. These zones constitute the major flow paths for infiltrating meteoric water. The brittle fault zones are thin (< 1dm) and form fault gouges with nearly no neo-formation of clay minerals<sup>3</sup>. The GTS underground laboratory is located some 420-520 m below the surface offers the unique possibility to collect fracture groundwater from such fault zones from instrumented intervals in numerous boreholes besides detailed mapping of the water-conducting structures.

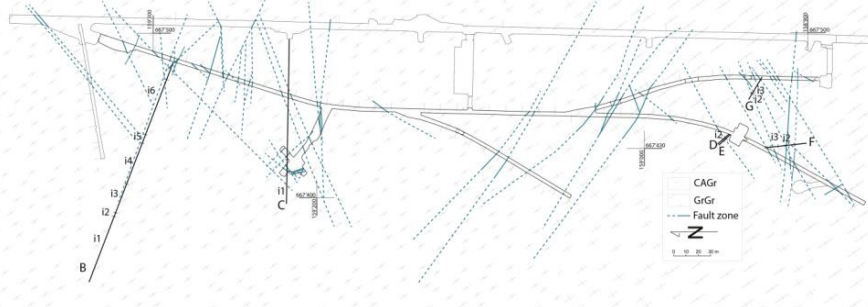


Fig. 1. Situation plan of the GTS underground rock laboratory with mapped traces of brittle fault zone (solid lines) and interpreted traces (dashed lines). Boreholes (n= 6) and packed of intervals (n= 13) are shown by black solid lines.

## 2. Fracture water chemical composition and evolution

Fracture water within the underground rock laboratory has been sampled in the 1980s<sup>1</sup> and in the 1990s<sup>4</sup>. In this study, fracture water was sampled five times (April 2014, August 2014, October 2014, January 2015 and August 2015) from 15 intervals in 7 boreholes (Fig. 1). Sampled boreholes are located either within the CAGr (A, B, C) or within the GrGr (D, E, F, G).

Regardless of the bedrock and with very little variation, the large majority of the fracture waters are of a general Na-Ca-TiC-F-(SO<sub>4</sub>) chemical type and of low total mineralization (TDS: 51.9 – 71.4 mg/L). Within the individual rock units, the chemical and isotope composition of fracture water at a specific location remained constant over the 2 years sampling period and – where available – also compared to the early surveys in the 80ies<sup>1</sup> and 90ies<sup>4</sup> of the last century.

Although similar in the general aspects, the chemical and isotopic composition of fracture water collected from the CAGr and GrGr differ systematically in several parameters. Differences include temperature, which is on average higher ( $13.7 \pm 0.9^\circ\text{C}$ ) in the GrGr compared to that of water collected from the CAGr ( $12.5 \pm 0.6^\circ\text{C}$ ). This difference can be associated with the higher overburden in the GrGr (ca. 500m) compared to that in the CAGr (450m) part of the rock laboratory based on a geothermal gradient of  $25^\circ\text{C}/\text{km}^5$ , which would suggest a temperature difference of  $1.2^\circ\text{C}$  for the different depths below surface. Furthermore, fracture water in the GrGr part is more alkaline (pH = 8.85 – 9.78) than that in the CAGr part (pH = 8.13 – 9.43). Related to the differences in pH the partial pressure of CO<sub>2</sub> is below that in the atmosphere and lower in the GrGr part ( $P[\text{CO}_2] = 10^{-5.26}$  bar) compared to that in the CAGr part ( $P[\text{CO}_2] = 10^{-4.91}$  bar). Fracture water in the GrGr part has higher concentrations of Na, K, Li, and Cl, but lower Ca and TIC concentrations compared to fracture water in the CAGr part (Fig. 2a). Higher Cl concentrations in the waters from the GrGr may be associated with greater interaction with pore water in the low-permeability rock matrix (known to be more saline) because no easily soluble Cl-bearing minerals occur in the fractures. The more intense diffusive exchange with such porewater, which is of moderately mineralised Na-Cl type ( $\text{Cl} \approx 330\text{--}440 \text{ mg}/\text{kg}_{\text{H}_2\text{O}}$ <sup>6</sup>) in the GrGr, might also indicate a somewhat longer residence time for the GrGr fracture

waters. Mg concentrations are close to detection limit and do not show variation, indicating minor dissolution of Mg-bearing minerals.

The mineralogical composition of the GrGr and the CaGr is very similar except for modal abundance of mafic minerals (mainly biotite), which is higher in the GrGr<sup>1</sup>. This allows using ion-ion ratios to deduce the evolution and reaction progress of fracture groundwater within the two lithologies. Ratios of Na/Ca and Sr/Ca are higher in fracture water of the GrGr compared to those of fracture water in the CAGr, whereas the Na/Cl ratio is considerably lower (Fig. 2b). These ratios indicate an increased dissolution of plagioclase (albite composition), a larger addition of Na and Cl from the matrix porewater, and a more advanced precipitation of calcite in fracture water of the GrGr. The latter explains the lower TIC concentrations and the higher concentrations of Sr, which comes from feldspar dissolution, in these waters whereas Ca concentrations are controlled by calcite equilibrium. In both types of fracture groundwater, the increase in pH is explained by the effect of feldspar dissolution under concurrent precipitation of kaolinite and calcite. Such an evolution is consistent with calculated mineral saturations indices for the fracture waters: All analysed fracture waters are saturated with respect to quartz (CAGr, SI =  $-0.01 \pm 0.05$ ; GrGr, SI =  $-0.11 \pm 0.09$ ) and calcite (CAGr, SI =  $-0.51 \pm 0.82$ ; GrGr, SI =  $-0.07 \pm 0.19$ ). Kaolinite saturation index varies strongly (CAGr, SI =  $0.35 \pm 0.96$ ; GrGr, SI =  $-0.29 \pm 0.94$ ) but most waters are close to saturation. Mg phases as well as fluorite, K-feldspar, and plagioclase are undersaturated.

It is concluded that the general reactions of fracture water in the CAGr and GrGr are similar, but the reaction progress and the exchange with porewater from the rock matrix is different. This implies longer circulation times of fracture water in the GrGr compared to that in the CaGr what is consistent with their isotope composition (see below).

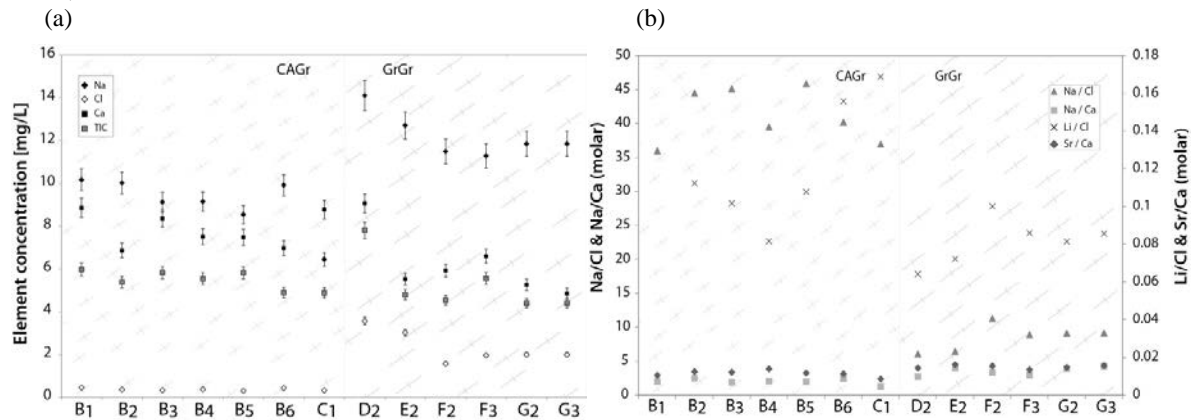


Fig. 2. (a) Concentrations of Na, Cl, Ca, and TIC in fracture water collected from various borehole intervals (x-axis) in the CAGr and GrGr (b) Molar ion ratios in fracture water collected from various borehole intervals (x-axis).

### 3. Groundwater origin and residence time

Fracture waters collected from within the CAGr and GrGr in the GTS underground rock laboratory show  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  values that plot on the Local Meteoric Water Line (LMWL)<sup>7</sup> defined by the precipitation on the Grimsel pass (Fig. 3a). The deuterium excess (d) of all samples is very similar and follows the trend given by the annual means of the local precipitation. Fracture water is therefore of meteoric origin in both rock units, infiltrated under present-day climatic conditions and stems from the same moisture sources. No systematic variation was observed between the different sampling campaigns and seasonal variations got erased by homogenization through mixing during infiltration and circulation in the rock mass down the depth of the GTS underground rock laboratory.

Conversion of the  $\delta^{18}\text{O}$  values into infiltration altitude based on surface water isotope data<sup>7</sup> yields in higher infiltration altitudes of about 110m for the fracture water collected in the GrGr (Fig. 3b), which is in good agreement with present-day topography. As the dip of the water-conducting structures in the CAGr and GrGr is similar, this could indicate longer flow paths and longer residence times of fracture water in the GrGr as suggested by the chemical differences and assuming similar flow rates in both units.

So far the absolute residence time of the fracture waters could not be constrained. In 2016, all sampled waters have  $^3\text{H}$  activities below a detection limit of 0.6 TU. Given the tritium concentration in surface waters in the area ( $7.0 \pm 1.1$  TU, 2016), the  $^3\text{H}$  input at Grimsel<sup>8</sup> and using a piston flow model – as a first approximation – the fracture waters must have infiltrated before the bomb peak in 1950s and thus have residence times of more than 65 years.

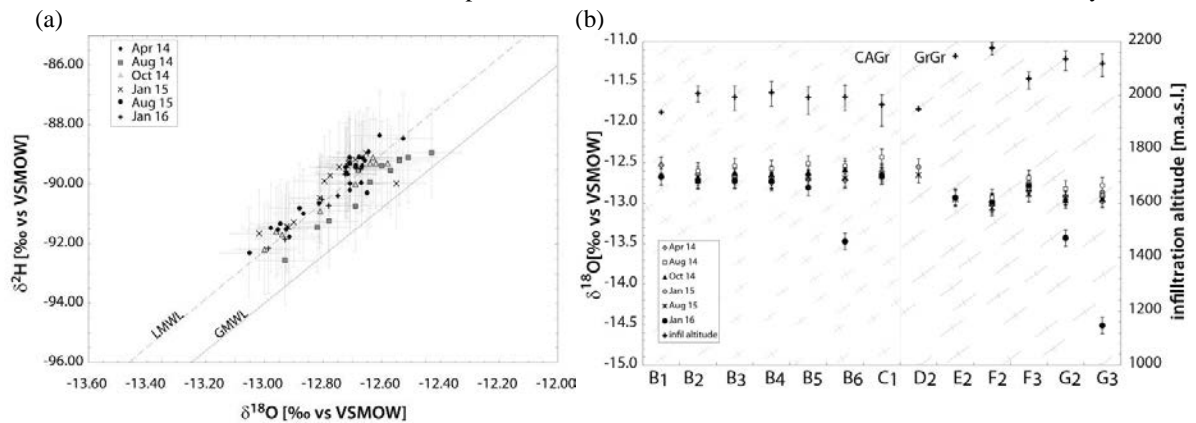


Fig. 3. (a)  $\delta^{18}\text{O}$  versus  $\delta^2\text{H}$  of fracture water in the GTS underground laboratory compared to Grimsel Local Meteoric Water Line (LMWL, data from <sup>7</sup>). (b)  $\delta^{18}\text{O}$  values of fracture water and infiltration altitude derived from these as function of sampling intervals (x-axes) and rock type.

#### 4. Conclusions

Sampling over 2 years enabled the definition of a baseline of fracture water chemical and isotope composition at the GTS underground rock laboratory. Chemical and isotope compositions remain constant within specific water-conducting zones encountered by packed-off borehole interval, but vary between intervals in the two main lithologies of the Grimsel Granodiorite (GrGr) and Central Aar Granite (CAGr). Fracture water flow is related to fault zones that were established by brittle deformation of the rocks and that are encountered in the rock laboratory as well as at the surface.

The isotopic composition and deuterium excess indicate a meteoric origin under present-day climatic conditions and from similar moisture source for the fracture water in both rock units. In both lithologies the major water-rock interactions are identical, but fracture water in the GrGr is chemically more evolved. Consistent with differences in chemical compositions, temperature and isotope signatures, longer flow paths related to the larger overburden and higher residence times are proposed for the GrGr fractures waters assuming similar flow rates in the water-conducting zones of both units. Based on measured  $^3\text{H}$  activities the minimum residence time is more than 65 years for all fracture waters.

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