

15th Water-Rock Interaction International Symposium, WRI-15

## Constraints on evolution and residence time of geothermal water in granitic rocks at Grimsel (Switzerland)

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### Abstract

Thermal spring water (17–28°C) occurring in crystalline rocks at the Grimsel area, Switzerland, represents a mixture of an old geothermal component with a surface-derived, young cold water component. Comparison of chemical and isotope data of cold and thermal spring water collected in 1991/92 and 2014/15 allows deciphering the residence time and evolution of the geothermal component. Accounting for changes in atmospheric input for  $d^{18}O$ ,  $d^2H$ ,  $^3H$  and  $^{14}C$ , a meteoric origin from similar moisture source(s) for the cold and geothermal component is proposed, a  $^3H$ -PF model age of about 7 years for the cold component and infiltration of the geothermal component before 30 ka during an interglacial cycle. Deep circulation of the geothermal component is inferred from water temperatures of 110–120 °C and the uptake of geogenic  $CO_2$  and/or  $CH_4$ .

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Peer-review under responsibility of the organizing committee of WRI-15

**Keywords:** geothermal water; groundwater mixing; radiogenic isotopes; atmospheric  $^3H$  and  $^{14}C$ ; crystalline rocks

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### 1. Introduction

Knowledge about the occurrence of thermal springs in Grimsel area in the Aar Massif of the Central Swiss Alps dates back more than two centuries<sup>1</sup>. Systematic mapping and sampling of thermal and cold springs occurred between 1991 and 1993 along a tunnel constructed by Gas Transit AG for a gas pipeline in 1973<sup>2,3</sup>. Almost 25 years later, some of these thermal and cold springs were analysed again for their chemical and isotope composition including the radiogenic isotopes  $^3H$  and  $^{14}C$ .

The tunnel crosscuts the para-autochthonous crystalline unit of the southern part of the Aar Massif in the Central Swiss Alps at a depth between about 100 m and 200 m below surface (Fig. 1). The rocks are of Variscan and older age and have undergone greenschist grade metamorphism during the Alpine orogeny. The tunnel further cuts across

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the so-called Grimsel Breccia at the southern rim of the Southern Aar Granite zone (Fig. 1). Formed during the Alpine orogeny, this breccia was overprinted by a meteoric water dominated hydrothermal system during Pliocene times that resulted in an enrichment of a suite of elements (e.g. Mo, Au, As, Sb, Li, U, W) typical for epithermal systems<sup>4</sup>. Present-day thermal springs are mainly limited to the near-vicinity of the Grimsel Breccia suggesting that this system still acts as fluid pathways and that the presently ongoing thermal activity represents a late stage of the same processes that produced the breccia during Pliocene times<sup>2,4</sup>. In contrast, cold springs are mainly associated with brittle, late-Alpine deformation structures associated to uplift and cooling of the rock suite.

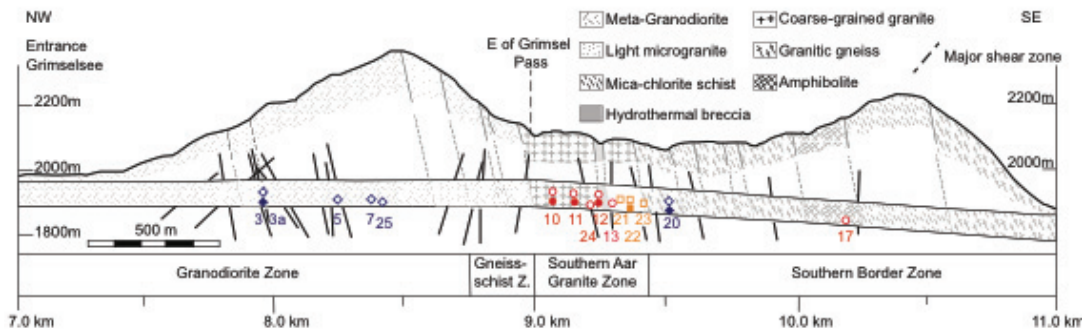


Fig. 1. Simplified geological cross-section of the tunnel section showing the localities of the 1991/93 (open symbols) and 2014/15 sample locations (closed symbols); red: thermal springs, squares: subthermal springs, diamonds: cold springs (modified from<sup>3</sup>).

## 2. Groundwater chemical composition and evolution

Groundwater from cold springs collected in 1991/93<sup>2,3</sup> and 2014/15 have temperatures between 3.5 and 10.8 °C and display variable yields between 0.2 and 50 L/min<sup>2,3</sup>. It is of a general  $\text{Ca-HCO}_3\text{-SO}_4$  chemical type with a low total mineralisation (TDS: 33–110 mg/L) and displays generally mildly alkaline (pH: 7.9–8.5) and oxidising ( $\text{Eh}_{(\text{Ag}/\text{AgCl})}$ : 100–450 mV) conditions. Little variation in chemical composition is observed over the more than 20 year period between the two sampling campaigns.

Groundwater from thermal and subthermal springs collected in 1991/93<sup>2,3</sup> and 2014/15 have temperatures of 20–28 °C and ~17°C, respectively, and display yields between about 0.2 and 4.5 L/min<sup>2,3</sup>. Thermal waters have elevated concentrations of Na, K, Li,  $\text{SO}_4$ , TIC, F and Si and are of a general  $\text{Na-SO}_4\text{-TIC-(F)-(Cl)}$  chemical type. At total mineralisations of 309–412 mg/L they display alkaline (pH: 8.9–9.2) and generally reducing conditions ( $\text{Eh}_{(\text{Ag}/\text{AgCl})}$ : –280 to –35 mV).

Temporal covariance of Na, Cl, Br, Li, K and  $\text{SO}_4$  between cold and thermal waters in 1991/93 and 2014/2015 ( $R^2 > 0.95$ ; Fig. 2a) suggests for the thermal spring waters a mixture of a geothermal end-member component from greater depth with dilute cold spring water from the surface. Based on the relationship between  $^3\text{H}$  (see below) and Cl and TDS, the mixing proportions of the geothermal component appears to have slightly increased from about 40% in 1991/93<sup>3</sup> to about 50% in 2014/15.

Cold spring groundwater is undersaturated with respect to calcite at corresponding log  $\text{pCO}_2$  values of –3.4 to –5.2. The concentrations of Ca and TIC evolve from surface to cold spring water at a molar ratio of 1 indicating dissolution of calcite from metamorphic/hydrothermally altered (saussuritised) plagioclase (Fig. 2b) besides dissolution of quartz and Al-silicates in accordance with calculated saturation states. In contrast, thermal waters are in equilibrium to slightly oversaturated with respect to calcite at corresponding log  $\text{pCO}_2$  values of –4.2 to –4.7. The higher TIC concentrations and lower Ca concentrations compared to cold spring water further suggest an additional carbon source for these waters (Fig. 2b). Thermal waters are further oversaturated with respect to quartz, kaolinite, adularia and at or close to saturation with respect to fluorite and albite.

Reconstruction of the original temperature by multi-component solute geothermometry using the GeoT computer code<sup>5</sup> reveals temperatures between about 110–120°C for the geothermal component, which is in agreement with the estimates made in 1992<sup>2</sup>.

### 3. Groundwater origin and residence time

Groundwater from cold and thermal springs have  $\delta^{18}\text{O}$  and  $\text{d}^2\text{H}$  values that plot along the local Grimsel meteoric water line<sup>6,7</sup> with the thermal waters having more negative values by  $\sim 1.5\text{‰}$  and  $\sim 10\text{‰}$ , respectively (Fig. 2c). The D-excess in cold and thermal waters is very similar and follows the trend given by the annual means of the local precipitation between 1991 and 2015. This indicates a meteoric origin from the same moisture source(s) for cold and thermal waters. Consequently, the more negative values of the thermal water are more likely to be associated with an infiltration of the geothermal component at higher altitude under present-day conditions compared to complex freezing and thawing cycles related to glacial melt water formation.

From 1991 to 2015, the  $^3\text{H}$  activity in cold and warm spring waters has dropped from  $>15$  TU to  $<5$  TU (Fig. 2d). The annual average of  $^3\text{H}$  in precipitation has strongly decreased from 1975 to 1990 and remained rather constant between 2000 and 2011 (Fig. 2d). In 2014, the  $^3\text{H}$  activity of lake water at Grimsel was  $8.8 \pm 0.7$  TU. Accounting for the variance in the  $^3\text{H}$  input and applying – as first approximation – a piston flow model (PFM) to the fracture-limited flow yields for the cold spring waters in 1991/93 and 2014/15 a consistent  $^3\text{H}$  PF-model age of about 7 years.

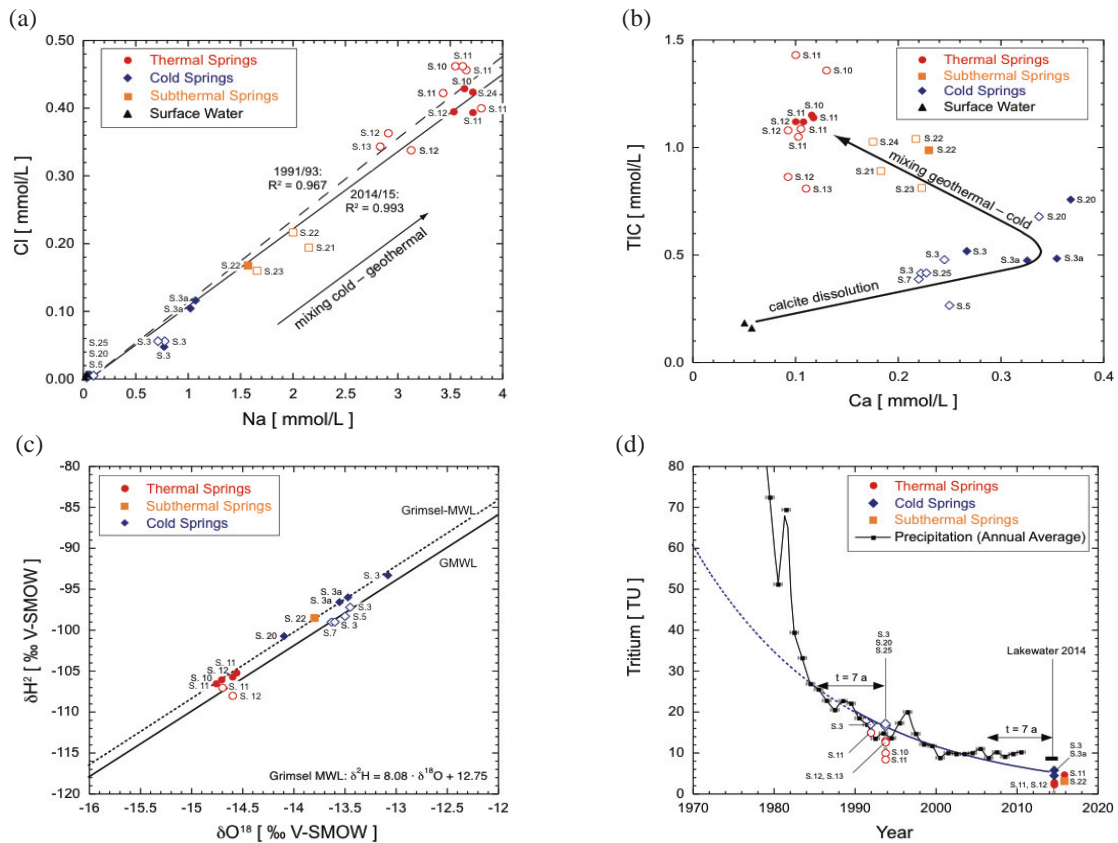


Fig. 2. (a) Linear relationship between Na and Cl concentrations and (b) between Ca and TIC concentrations in cold and thermal groundwater; (c)  $\delta^{18}\text{O}$  vs.  $\delta^2\text{H}$  of different groundwaters (Grimsel meteoric water line from<sup>5,6</sup>); (d)  $^3\text{H}$  activities of groundwater compared to  $^3\text{H}$  in precipitation at Grimsel (data from<sup>6,7</sup>), the solid and dashed blue lines indicate the PFM evolution for cold spring water from 1993 and 2014, respectively (open symbols: water samples from 1991/93, closed symbols: water samples from 2014/15; "geothermal" = old geothermal component).

Older (theoretically possible) model ages for the 2014/15 groundwater are inconsistent with the  $^3\text{H}$  input function and the 1991/93 cold spring water data (Fig. 2d). In turn, the thermal end-member must be  $^3\text{H}$ -free and has infiltrated before the bomb peak in the 1950ties based on the comparison of the  $^3\text{H}$  activity in thermal and cold spring waters combined with the linear relationships given by chemical compounds and the  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  values.

So far, carbon isotope data are available for three (sub)thermal spring waters collected in 2014/15: Thermal springs S.12 and S.11 have  $^{14}\text{C}_{\text{TIC}}$  activities of 32.7 pmc and 25.9 pmc and  $\delta^{13}\text{C}_{\text{TIC}}$  values of  $-12.2\text{‰}$  and  $-12.3\text{‰}$  V-PDB, respectively. The  $^{14}\text{C}_{\text{TIC}}$  activity of the subthermal spring S.22 is 46.9 pmc at a  $\delta^{13}\text{C}_{\text{TIC}}$  value of  $-16.6\text{‰}$  V-PDB. At the near-by Jungfrauoch monitoring station (3450 m asl)  $^{14}\text{C}$  in atmospheric  $\text{CO}_2$  has decreased drastically over the past 30 years due to dilution with  $\text{CO}_2$  from fossil fuel burning<sup>8</sup>. Adapting the residence time given by  $^3\text{H}$ , the atmospheric  $^{14}\text{C}$  input during infiltration of the cold springs in 2008 averaged about 104.7 pmc ( $\Delta^{14}\text{C}_{\text{CO}_2} - 47.3\text{‰}$  at Jungfrauoch<sup>8</sup>) and was therefore almost identical to pre-bomb peak infiltration.

Based on the identified dissolution of  $^{14}\text{C}$ -free metamorphic/hydrothermal calcite allows estimating the  $^{14}\text{C}$  activity in the cold spring waters to be about 60–50 pmc based on the increase in TIC concentrations by about a factor of 2 from the surface to the cold springs (Fig. 2b). Mixtures of the original geothermal water with cold spring water, i.e. the sampled thermal spring waters, have 2–2.5 times higher TIC concentrations but lower Ca contents compared to the cold spring water (Fig. 2b). Observed calcite precipitation in thermal waters cannot explain this concentration behaviour. Consequently, the 0.4–0.5 fraction of geothermal water present in the thermal spring waters must have an additional geogenic carbon source that must be  $^{14}\text{C}$ -free and depleted in  $^{13}\text{C}$  to account for the measured  $^{14}\text{C}$  activity and  $\delta^{13}\text{C}_{\text{TIC}}$  in the (sub)thermal spring waters. Therefore, the original geothermal water component has infiltrated more than about 30 ka ago during an interglacial period based on  $^{14}\text{C}$ .

#### 4. Conclusions

Thermal waters associated with hydrothermal breccia structures created during Pliocene times in the granitic rocks Grimsel, Switzerland, represent mixtures of an old geothermal component with recent, surface derived infiltration. Thermal spring waters collected in a tunnel some 200 m below surface contain 40–50% of an old geothermal component that is of meteoric origin, has the same moisture source(s) and has infiltrated at higher altitude under similar climatic conditions as prevailing today. For the young, cold spring water component a  $^3\text{H}$ -model age of 7 years can be derived taking the change in atmospheric  $^3\text{H}$  input since the bomb peak into account. Based on  $^3\text{H}$ ,  $^{14}\text{C}$  and  $\delta^{13}\text{C}$  measured in the thermal waters, the old geothermal component is  $^{14}\text{C}$ -free and has infiltrated some 30 ka ago during an interglacial period. Formation temperatures of the geothermal component are about 110–120°C indicating a circulation depth of several km of depth based on present-day geothermal gradient in the area (ca. 25°C/km<sup>9</sup>) during which also an uptake of geogenic carbon as  $\text{CO}_2$  and/or  $\text{CH}_4$  occurred.

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