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Helium diffusivity and fluxes from a sedimentary basin (Permo-Carboniferous trough, Northern Switzerland)

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ABSTRACT

Abundances of radiogenic He and Ar isotopes, observed in ground waters of the Permian-Carboniferous terrigenous sediments (PCT) in Northern Switzerland, exceed those in the overlaying (Muschelkalk) and underlying (Crystalline basement) aquifers, eliminating external sources of these species. In this case a comparison of the observed abundances with those in situ generated sheds light on mobility of noble gases generated in the PCT.

Detailed studies of the parent element and He isotope concentrations in the PCT rocks, mineral separates, pore and ground waters along with the data on γ-logging and porosity allow He isotope production and redistribution in the PCT rock-water system to be understood and quantified. The PCT shales generate most of ^{3}He and ^{4}He and both isotopes are almost completely released from the shales into pore space and migrate either into the aquifers with movable waters and further out of the PCT, or into gas-fluid inclusions of the rock forming minerals, quartz and plagioclase.

Here we present a He concentration profile, based on: (i) helium measured concentrations in ground water samples from 5 aquifers available within the cross section studied; (ii) helium concentrations (in 15 PCT pore water "samples"), derived from the He partial pressure in gas-fluid inclusions, recovered by special extraction/saturation experiments.

In order to estimates the rates of He migration through the PCT, we compare the produced and observed abundances within the frame of 1D diffusion model. Only two parameters, the diffusion coefficient for the whole sequence, $D_{ALL} = 4.1 \times 10^{-3}\,m^2\,year^{-1}$, and the rate of He removal from the crystalline aquifers, $\theta_{CR} \approx 1.4 \times 10^{-6}\,year^{-1}$, are required to obtained a good agreement between the observed and calculated He concentrations in PCT waters: the minimal square deviations are $\approx (2\text{ to }3) \times 10^{-8}\,mol\,cm^{-3}\,H_2O$, comparable with the accuracy of measurements. The following parameters were derived from the modeling: the reduction factor $D_{ALL}/D_0 = 0.013$; the He diffusion fluxes into the overlaying Muschelkalk and underlying Crystalline aquifers, $F_{MU} \approx 1.4 \times 10^{-7}\,mol\,m^{-2}\,year^{-1}$ and $F_{CR} = 0.24 \times 10^{-7}\,mol\,m^{-2}\,year^{-1}$, respectively. From these values we calculated the mean helium residence times in PCT, $< \tau_{PCT} > \approx 120$ Ma, and in Muschelkalk aquifer, $< \tau_{MU} > \approx 6$ ka. The relationships between He diffusivities of the PCT and the underlying Crystalline basement are discussed. We further compare our results with other estimates of He mobility in low-permeable sediments.

1. Introduction

Noble gases, the most conservative tracers of terrestrial fluids in subsurface environments, are widely used to study different geochemical problems, such as the safety assessments of nuclear waste disposals (De Marsily et al., 2002; Osenbruck et al., 1998), secure storage of movable gaseous phases, such as CO_2 (Jeandel et al., 2010; Holland and Gilfillan, 2013), understanding of processes of hydrocarbon formation, migration and estimation of the respective time scales (Prinzhofer, 2013). Generally radiogenic He atoms, derived from the decay of U and Th, readily migrate from sedimentary, metamorphic and magmatic rocks into pore waters. Therefore a comparison of He production in the host rocks with its presently observed abundance in the rock – water system allows He mobility and removal rate from this system to be constrained, assuming negligible external He fluxes. The major mechanisms, that govern He mobility, are: (i) movement of He atoms as constituents of water flow, followed by He discharge into the atmosphere; (ii) diffusion through porous (fractured) media towards

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domains with lower He concentrations; (iii) ascent along with abundant gases (CH_4, CO_2, N_2) with subsequent discharge into the atmosphere or accumulation in a gas field.

The residence time of ground waters vary over a large range, from ≈1 day or less (Snow and Spalding, 1997) to ≈10^9 years (Holland et al., 2013), therefore a number of different tracers are used (Lehmann et al., 1993). He and heavier radiogenic noble gas isotopes (^{21}Ne*, ^{40}Ar*, ^{136}Xe*) allow dating of the ancient ground waters and studying of respective transport processes. Since the early contribution of Savchenko (1935) these issues were widely discussed (Marine, 1979; Osenbruck et al., 1998; Rubel and Sonntag, 2001; Rubel et al., 2002; Battani et al., 2011). In the last paper the authors compared the observed He concentrations in pore waters with those calculated from He production by U and Th decay in the host rocks since the sedimentation age; U and Th abundances were obtained from gamma-logging. In the deep segment of the cross section the measured and calculated concentrations were similar, indicating very long He residence times, and these authors included into their 1D model formation of the sedimentary layers with zero concentrations of radiogenic species at the formation times. In many respects these approaches are similar to what is discussed in our contribution.

Tolstikhin et al. (1996, 2011) and Lehmann et al. (2003a) studied noble gas isotope and parent element abundances in rocks, minerals, pore and ground-waters of the alternating shale-sandstone rock sequence of the Permo-Carboniferous Trough, Northern Switzerland, opened by the Weiach borehole (hereafter abbreviation PCT is used for the studied cross section). These authors have presented a self-consistent scenario of He isotope production, redistribution between aquitards and aquifers as well as between pore waters and fluid inclusions in minerals, and slow removal from the rock–water system.

The objectives of this contribution are: to constrain parameters governing He migration through PCT, such as the He diffusivity in shale-sandstone porous media and the He flux into the overlying and underlying aquifers; to compare results obtained with data available for other low-permeable hydrological systems, and to assess the PCT as a potential store of nuclear waste.

Two important issues are to be taken into account in our approach. (1) The pore water residence time in PCT is comparable with the sedimentation ages of the water bearing sediments (Tolstikhin et al., 2011); therefore the time dependent model, taking into account these ages, is applied, in line with Battani et al. (2011). (2) The sandstones contain trapped excess He, generated in the adjacent shale aquitards (Lehmann et al., 2003a; Tolstikhin et al., 2011; Gannibal and Tolstikhin, 2013); hence, there is an internal sink of He atoms from pore waters, which for the first time is included in the He migration model.

2. Regional geology and hydrology

There are several Palaeozoic troughs in the Alp foreland (Fig. 1a) including the Permo-Carboniferous trough (PCT) of Northern Switzerland (Fig. 2). The foreland was considered as a potential site for radioactive waste disposals and seven deep boreholes were drilled in this area by the Swiss National Cooperative for the Disposal of Radioactive Waste. Results of careful studies of rocks, minerals, and groundwater samples from one of these, the Weiach borehole, are used in this contribution (Pearson et al., 1991; Tolstikhin et al., 2011).

The Weiach borehole is located near the Rhine river, 25 km northwest of Zürich (Fig. 1a, b). We model He diffusivity of the Weiach cross section below the Buntsandstein aquifer (982 to 991 m, Fig. 2). The cross section consists of Permian and Carboniferous terrigenic sediments, mainly shales and sandstones, down to 2020 m; below the borehole intruded into the crystalline basement, from 2020 m to its final depth 2482 m (Fig. 2).

The crystalline basement is represented by a monotonous series of highly metamorphosed biotite-plagioclase paragneisses crossed by aplitic dykes. During the Late Variscan (320–300 Ma ago) the gneisses were subjected to a last hydrothermal overprint.

The Carboniferous is composed of a ca. 600 m thick sequence of alternating layers of shale, clay, silt and sandstone. The shale/sandstone mass ratio is approximately 1, thickness of the layers ~ 10 m (Matter et al., 1988; Pearson et al., 1991). The Permian cross section (Fig. 2) is similar to the Carboniferous and is overlain by the fluvial sediments of the Buntsandstein and the shallow marine sediments of the Muschelkalk. Several groups of meter-thick coal seams occur between 1551 m and 1751 m depth.

The present day temperature of the PCT ranges between 55 °C at the top and 88 °C at the bottom. The porosity decreases from ≈15% at ≈1000 m depth to below 3% in the crystalline basement. The hydraulic conductivity of PCT sediments is generally very low ($< 10^{-6}$ m year^{-1}) and the Carboniferous sediments contain only stagnant ground waters. The Permian ground waters were collected from two intervals (average depths of 1117 m and 1411 m) with an elevated hydraulic conductivity, the high values (> 1 m year^{-1}) of which are observed in the upper part of the cross section for the Buntsandstein and Muschelkalk aquifers (Matter et al., 1988).

According to Pearson et al. (1991) the degree of the mineralization, enrichment in ^{18}O and ^{2}H, absence of ^{14}C and high contents of radiogenic noble gases indicate a very long residence time for the ground waters within the PCT. The two Permian ground water samples show a substantial contribution of radiogenic ^{40}Ar* indicated by high ^{40}Ar/^{36}Ar ratios, > 370. The deep Permian ground water (Pm1) also displays the highest ^{4}He concentration (4.48×10^{-3} cm^3 STP/g H_2O).

The ground water in the Buntsandstein aquifer just on top of the Permian sediments has a ^{40}Ar/^{36}Ar ratio of only 306, and these ratios in ground waters from the overlying Muschelkalk and the underlying Crystalline basement aquifers are similar the atmospheric value of 296. Further, these aquifers show lower He concentrations than those in the Pm1 (see Fig. 4). These data suggest a local production for ^{3}He, ^{4}He and ^{40}Ar* in the PCT rock-water system, because fluxes from external sources could only dilute the highly mineralized, He- and ^{40}Ar*-rich PCT ground waters.

3. Production and redistribution of He (and Ar) isotopes in the PCT Weiach cross-section

A large data set on parent element concentrations (U, Th, K, Li) and He and Ar isotope abundances in sedimentary rocks, mineral separates and ground (pore) waters allow production of radiogenic species, ^{3}He, ^{4}He and ^{40}Ar*, to be estimated and their behavior in the PCT shale/sandstone interlayering to be understood (Pearson et al., 1991; Tolstikhin et al., 1996, 2005, 2011; Lehmann et al., 2003a; Gannibal and Tolstikhin, 2013; Fig. 3). These results are used to determine the model, developed in this contribution.

3.1. Radiogenic production of He isotopes

Taking into account the previous studies, we exclude external sources of radiogenic noble gases for the PCT cross section (Section 2). In this case a comparison of the observed abundances of the radiogenic noble gas species with those in situ generated allows mobility of the noble gases to be quantified.

The parent element concentrations [μg g^{-1}], obtained either by γ-logging of the Weiach borehole (Weber et al., 1986) or by X-radiography of the rock samples (Tolstikhin et al., 1996), the rock ages (Pearson et al., 1991), and the porosity data (≈300 measurements, Matter et al., 1988) constrain the closed system (no loss/gain of species of interest) pore water concentrations of ^{4}He [mol cm^{-3} H_2O], accumulated since the beginning of sedimentation T years ago (Savchenko, 1935; Zartman et al., 1961):

$$^{4}He^{*}_{PW} \approx (5.40 \times 10^{-18}\,U + 1.28 \times 10^{-18}\,Th) \times T \times L \times \rho \times (1-\varphi)/\varphi \quad (1)$$