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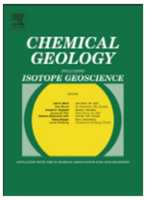

Research papers

Production, redistribution and loss of helium and argon isotopes in a thick sedimentary aquitard-aquifer system (Molasse Basin, Switzerland)

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ABSTRACT

In a rock–water system an apparent residence time for He can be derived from the bulk He concentration in the rock and the porewater and the He production rate provided that (1) the system is at steady state and (2) the He flux from external sources is negligible. This second condition is crucial and needs to be assessed by identifying the various He-sources in a given hydrologic environment.

He and Ar isotope abundances were investigated in whole rock samples and mineral separates of the alternating shale–sandstone rock sequence of the Permo-Carboniferous Trough (PCT) of the Molasse Basin in northern Switzerland. ^{4}He concentrations and ^{40}Ar/^{36}Ar ratios in groundwater in the bounding aquifers are lower than those in PCT groundwaters, thus eliminating a possible external source of radiogenic noble gases. The elements producing radiogenic helium isotopes, U, Th and Li, are mainly concentrated in the shales, where they reside in biotite, muscovite, clay minerals and organic matter. Contents of these elements as well as chronological and chemical data allow us to calculate maximum "closed system" He isotope concentrations and the retention coefficients, e.g. $^3He_{measured}/^3He_{calculated}$ ratios. As deduced from the low retention coefficients, almost all helium-3 that has been produced in these minerals since their deposition has been lost into the complementary reservoir, i.e. the porewater; the same conclusion is valid for ^{4}He. This is also indicated by the similarity between the calculated ^{4}He/^{3}He production ratio of the shales and that observed in PCT groundwater (1.3×10^7). Moreover, similar ^{4}He/^{3}He ratios are measured in quartz and plagioclase mineral grains from the sandstone layers, by far exceeding the production ratios derived for these minerals. Also, the measured concentrations of He isotopes (especially ^{3}He) are higher than the calculated production capacities of these minerals. These observations suggest that some of the He produced in the shale and released from these rocks migrates via the porewater into the sandstone layers and penetrates further into some rock-forming minerals, such as quartz and plagioclase.

Measurements of the migration rate of He atoms through quartz crystals showed that equilibrium between internal (gas–fluid vesicles) and external (porewater) He-concentrations is reached on a short time scale of $\sim10^4$ years at an in-situ temperature of $\approx70\,°C$. The equilibrium allows the He concentrations in the porewater across the PCT sequence to be quantified using quartz crystals as detectors. For the PCT segment opened by the Weiach borehole these "quartz-derived" concentrations as well as those directly measured in PCT groundwaters are rather high implying a long residence time for the He-atoms in the PCT rock-porewater system, well exceeding 10^7 years.

1. Introduction

In terrestrial rocks helium isotopes are mainly produced by α-decay of radioactive ^{238}U, ^{235}U, ^{232}Th families (radiogenic ^{4}He), and by nuclear reactions, primarily ^{6}Li (n, α) $^3H\rightarrow\beta\text{-}\rightarrow{}^3He$ (nucleogenic ^{3}He). Both processes are exothermic, so that the released particles produce damage tracks in the host material. These tracks allow radiogenic (nucleogenic) helium atoms to release readily from sedimentary, magmatic and metamorphic rocks, so that generally less than ~10% of the radiogenic or nucleogenic helium has been retained since the time of rock formation. Electron capture by ^{40}K nuclei (yielding radiogenic argon $^{40}Ar^*$) generates little energy and the $^{40}Ar^*$ atom, though a point defect in host material, is much less mobile than helium (e.g. Mamyrin and Tolstikhin, 1984).

The rate of loss of radiogenic noble gas isotopes from rocks or rock-forming and accessory minerals into the interstitial space (i.e., the pore space) can be estimated from comparison of their measured

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concentrations in a rock or mineral and those calculated from abundances of the parent elements and the formation age (for ^{3}He the whole rock chemical composition is required as well), assuming no loss or gain of the species of interest (i.e., closed system assumption). Radiogenic noble gas atoms (released from rock-forming and accessory minerals) enter pore water, the major complementary reservoir. Because these atoms are chemically conservative their removal from a rock–water system occurs either by diffusion along the concentration gradient in a situation with stagnant porewater (i.e. in an aquitard) or by advection with flowing groundwater (i.e. in an aquifer) and finally by escape into the atmosphere. Because of helium dissipation from the atmosphere (the residence time of a helium atom in this reservoir is approximately 1×10^6 years) concentration of air helium in terrestrial waters is very low, so that small contributions of helium from other sources (e.g., from rocks or from ^{3}H decay) can be detected. Because of these reasons helium is often used as a hydrological tracer.

Assuming steady state conditions and provided that there is a negligible helium flux from external sources, a helium residence time in a rock–water system can be calculated from the helium production rate and its concentrations in the rock and coexisting porewater, thus shedding light on the rate of pore- or groundwater transport.

Of the two assumptions mentioned above, that of steady state conditions is rarely valid in a rock–water system at temporal and spatial scales of ~300 Ma and ~1000 m (in thickness), respectively (as in the system discussed below). However, applying this assumption to a rock–water system of low permeability, where the porewater essentially stagnates, an apparent mean loss rate (and thus an apparent mean residence time) of helium can be derived, which can then form the basis for more complex non-steady state models of such system.

The second assumption, i.e. a negligible He flux from greater depth, is quite crucial, and this assumption was considered as invalid in a number of publications. For example, Torgersen and Clarke (1985, 1987) proposed a “whole-crustal flux” or a “flux through the crust–mantle boundary” as an explanation for high He concentrations and a slightly enhanced ^{3}He abundance (relative to a typical crustal value ^{4}He/^{3}He$\approx5\times10^7$, Mamyrin and Tolstikhin, 1984) in groundwaters of the Great Artesian Basin in Australia (GAB, see also Torgersen, 1989; Torgersen et al., 1992). Castro et al. (2000) argue along the same line based on observations and modelling of He data in three different aquifer systems with calculated groundwater residence times mainly up to 10^4 years. These authors postulate that “it has to be expected that radiogenic ^{4}He is transported into the aquifers from the deep crust by advection, diffusion and dispersion”. For one of these aquifers, Castro (2004) suggested a mantle helium flux throughout the continental crust despite the quite low (crustal) abundance of ^{3}He in helium observed in groundwaters. If such transfer of He isotopes from the mantle and deep crust into aquifer systems operated universally, He isotope data could not be used to infer residence times in rock–water systems. In the case of the GAB groundwaters, however, Lehmann et al. (2003a) re-considered the proposed residence times and origin of He isotopes using ^{81}Kr/Kr and ^{36}Cl/Cl values and helium isotope inventory in GAB sandstones and shales. These authors concluded that both helium isotopes and the subsurface-produced portion of ^{36}Cl in the GAB groundwaters of the sandstone aquifers were mainly derived from the adjacent shale aquitards. This indicates that at least for old groundwater systems the in-situ production of He isotopes and their redistribution between aquifers and aquitards cannot be neglected. The assumption of a general He flux from the mantle and/or deep crust is further brought into question by the occurrence of such a flux only in specific tectonic settings (e.g. Ballentine et al., 2002).

In some cases, in-situ production is indeed considered to be a major source of He isotopes in sedimentary basins. The central role of in-situ production follows from experimental studies of aquifer systems with low flow velocities and long residence times of helium atoms (e.g., Mazor, 1972; Marty et al., 1988; Pearson et al., 1991), of aquitards (e.g. Osenbrück et al., 1998; Rübel et al., 2002), and from modelling helium concentration fields in cores of ancient sedimentary basins undisturbed by recent tectonic events (e.g. Ivanov et al., 1978). In such systems the following processes generally control the radiogenic noble gas isotope concentrations. First, the local radiogenic isotope production provides a source for fluxes from the host rocks into adjacent porewater. Second, the radiogenic atoms migrate from rocks of low permeability (such as shale and claystone layers) into adjacent aquifers. And third, the hydrogeologic setting of the system controls fluxes at a larger scale. For example, the flux of radiogenic species from the deep crust or from the mantle towards higher-lying hydrological systems can become partly or completely interrupted by deep-seated aquifers, depending on their prevailing groundwater flow velocity. Permeable aquifers near the interface between crystalline basement and sedimentary sequences are often observed (e.g., Pearson et al., 1991). Solomon et al. (1996) and Sheldon et al. (2003) suggested in situ production as a major source of radiogenic helium even for young waters (~10 to 100 years) in shallow aquifers.

The two “end-member” models outlined above bound more general cases: mixing of water flows (or helium fluxes) from different sources (reservoirs) is certainly a very general feature of ground water evolution. The problem is to adequately characterise the sources, quantify mixing proportions, and then derive (if possible) respective time parameters. For example, helium flux into an aquifer could be related not to the “deep crust” or “crust–mantle boundary” or to “crust + mantle” reservoirs, but to an adjacent aquitard or even to specific aquitard material within an aquifer. Distinguishing between these possibilities is important, as in the latter cases taking the aquitard(s) into account allows intra-basin production to be considered as the major source of radiogenic noble gas isotopes. This in turn permits adequate estimation of their residence time scales and redistribution between the two rock types.

In order to understand He and Ar isotope inventory in terrigenic sediments and related ground- and pore- waters, Tolstikhin et al. (1996) studied contents of the parent elements (U, Th, K, Li) and concentrations of noble gas isotopes (^{3}He, ^{4}He, ^{40}Ar*) in sandstone and shale (as well as in the minerals separated from these rocks) in the shale–sandstone rock sequence of the Permo-Carboniferous Trough (PCT) of the Molasse Basin in Northern Switzerland, which appeared to be a promising hydrogeological system for estimating He residence time in pore- and ground-waters (see below last paragraph in Section 2 and Fig. 2).

The Permo-Carboniferous sediment sequence mainly consists of shales and sandstones. Contents of the elements producing helium isotopes, U, Th, and Li, in the shale sample substantially exceed those in sandstones: shale appears to be the major generator of radiogenic and nucleogenic helium isotopes, with the calculated production ratio of ^{4}He/^{3}He$=1.37\times10^7$. On the other hand, the shale has lost almost all of the helium produced into related pore waters (Loosli et al., 1995; Tolstikhin et al., 1996).

In contrast, the sandstone sample contained more ^{4}He and especially ^{3}He than what could have been produced in-situ, indicating a gain of both He isotopes. Furthermore, the ^{4}He/^{3}He ratios of the sandstone whole rock sample and its minerals vary in a narrow range between 1.1×10^7 and 1.4×10^7: these values are very close to the ratio measured in He from the Permian groundwater at the same depth (1.06×10^7, Pearson et al., 1991) and to the production ratio calculated for the adjacent shale sample. Based on these observations, Tolstikhin et al. (1996) suggested that the excess helium isotopes in the sandstone were derived from the surrounding shale layers. The uptake of large quantities of He in quartz was later experimentally confirmed by Lehmann et al. (2003b) and Tolstikhin et al. (2005).