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## Helium in porewater and rocks of crystalline bedrock from the Fennoscandian Shield, Olkiluoto (Finland)

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### Abstract

Dissolved noble gases in matrix porewaters of low-permeable crystalline rocks are useful tracers for investigating porewater migration and interactions between porewater and groundwater. Samples from the 1160 m deep OL-KR56 borehole, drilled at the Olkiluoto site (southwestern Finland), have been analysed for concentrations and isotope signatures of helium in porewater, groundwater and in the rock matrix. These are the first <sup>3</sup>He/<sup>4</sup>He ratios combined with <sup>4</sup>He concentrations in porewaters and rocks from the Fennoscandian Shield. High <sup>4</sup>He porewater concentrations suggest long porewater residence times, while concentration differences of one order of magnitude between porewater and groundwater indicate a transient state between these reservoirs. Concentrations of <sup>4</sup>He in porewater show small variations with depth, which are mimicked in the concentrations of conservative chemical tracers as well as in <sup>4</sup>He concentrations of groundwaters from various regional boreholes. <sup>3</sup>He/<sup>4</sup>He ratios in porewaters are higher than in the rock matrix, requiring either fractionation during redistribution of He between these reservoirs or the addition of an isotopically light component.

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### 1. Introduction

In low-permeable crystalline bedrock such as from the Fennoscandian Shield, porewater resides in the connected inter- and intragranular porespace of the rock matrix. Chemical and isotopic characterisation of such waters offers the potential to investigate diffusive transport within the porewater, and interactions between porewater and

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advectively flowing groundwater in local and regional fracture networks as a function of time and space. Dissolved noble gases are useful tracers for investigating porewater migration, as they are chemically inert and neither their concentration nor isotope signature is known to be affected by chemical or biological processes within the porewater or groundwater system.

The subvertical borehole OL-KR56 was drilled in winter 2011/2012 to a depth of 1160 m below surface (b.s.) in predominantly unaltered gneissic basement rocks of the eastern part of Olkiluoto island (southwestern Finland)<sup>1</sup>. Below 700 m b.s. three groundwater conducting fractures with transmissivities between  $3 \times 10^{-9}$  and  $1.6 \times 10^{-8}$  m<sup>2</sup>/s were encountered, of which the last one was sampled. A series of core samples were taken for investigations including chemical tracers in porewater and noble gases in porewater and bulk rocks<sup>2</sup>. This dataset comprises the first <sup>3</sup>He/<sup>4</sup>He isotope ratios and - together with data from the subhorizontal borehole ONK-PH9 at approximately 300 m b.s.<sup>3</sup> – the first <sup>4</sup>He concentration data in porewaters and rocks of the Fennoscandian Shield.

## 2. Sampling and analytical methods

The samples for porewater noble gas analyses were conditioned on-site to minimize sample degassing and contamination with air. Immediately after core recovery the drillcore samples were broken to a length of approximately 9 cm with a hammer, wiped clean, weighed and sealed into stainless steel cylinders. After repeated flushing with 99.99% pure N<sub>2</sub> to remove all air, the cylinders were evacuated, sealed and the dissolved porewater gases were allowed to quantitatively outgas over 13 months into the cylinder's void volume at a constant temperature of 21°C. Separation and purification of the extracted porewater gases by sequential gettering and cryocondensation as well as measurement of their <sup>4</sup>He concentrations by mass-spectrometry was performed at the University of Bern, Switzerland. Analysis for <sup>3</sup>He/<sup>4</sup>He ratios of the purified gas was done at the University of Bremen, Germany and radionuclide concentrations, <sup>4</sup>He concentrations and <sup>3</sup>He/<sup>4</sup>He ratios of the rock phase were measured by the Kola Scientific Centre in Apatity, Russia. Water content and water-loss porosity were determined gravimetrically and using the bulk wet density. A fracture groundwater sample was obtained from a packed off interval between 1113.8 and 1117.7 m b.s. using pressurized sampling vessels and analysed at the University of Heidelberg. Analytical uncertainties are <7% for <sup>4</sup>He concentrations and <1% for <sup>3</sup>He/<sup>4</sup>He ratios in porewater and <1% and <6%, respectively, in groundwater. For rock samples, analytical uncertainties are ≤10% for <sup>4</sup>He<sub>rock</sub>, U and Th concentrations, ≤20% for Li concentration and ≤40% for (<sup>3</sup>He/<sup>4</sup>He)<sub>rock</sub> isotope ratios. Porewater chemical tracers were extracted by out-diffusion into ultrapure water and measured by ion chromatography<sup>2</sup>.

## 3. Helium in the rock matrix

The Olkiluoto bedrock lithologies show significant variability in texture and mineralogy<sup>4</sup>, with the principal lithologies encountered in the OL-KR56 borehole listed in Table 1. In these rocks, concentrations of the parent nuclides of <sup>4</sup>He (U, Th) and <sup>3</sup>He (Li) vary within a factor of 2 and 4, respectively, with exception of a notably lower Th concentration in the pegmatitic granites<sup>2</sup>. <sup>4</sup>He<sub>rock</sub> concentrations show uniform values of  $4.0 - 6.6 \times 10^{-4}$  ccSTP/g<sub>rock</sub> in the various rock types, while (<sup>3</sup>He/<sup>4</sup>He)<sub>rock</sub> ratios cluster at  $1.3 - 1.9 \times 10^{-8}$  except for the tonalitic-granodioritic-granitic (TGG) gneisses.

Table 1. <sup>4</sup>He and radionuclide concentration and <sup>3</sup>He/<sup>4</sup>He ratio of the main lithologies from borehole OL-KR56

Lithology	U (ppm)	Th (ppm)	Li (ppm)	<sup>4</sup> He <sub>rock</sub> (ccSTP/g <sub>rock</sub> )	( <sup>3</sup> He/ <sup>4</sup> He) <sub>rock</sub>
veined gneiss	4.2	11.8	74.3	6.6E-4	1.3E-8
diatexitic gneiss	7.1	7.3	18.6	6.3E-4	1.5E-8
mica gneiss	4.2	13.0	74.3	4.0E-4	1.7E-8
pegmatitic granite	9.9	0.9	13.5	5.3E-4	1.9E-8
TGG gneiss	5.2	10.5	79.0	6.5E-4	0.8E-8

## 4. Helium in porewater

The analysed samples contain between 0.34 and 0.73 g of porewater, which equals water contents of 0.09 - 0.17 wt.% relative to the water saturated rock. Measured <sup>4</sup>He concentrations in these porewaters range from

$9.8 \times 10^{-2}$  to  $2.3 \times 10^{-1}$  ccSTP/g<sub>pw</sub> (Table 2), which is over 6 orders of magnitude higher than air-saturated water ( $< 4 \times 10^{-8}$  ccSTP/g<sub>H<sub>2</sub>O</sub> at 20°C, at a porewater salinity of 33.9‰ total dissolved solids)<sup>5</sup>. With respect to in-situ conditions, the porewater <sup>4</sup>He concentrations are at least a factor 2 below saturation ( $> 5 \times 10^{-1}$  ccSTP/g<sub>pw</sub> at P<sub>hydrostatic</sub> > 7 MPa, T < 60°C), precluding the existence of He as a free gas phase.

Table 2. <sup>4</sup>He concentration and <sup>3</sup>He/<sup>4</sup>He ratio of porewater (pw) and groundwater (gw) from borehole OL-KR56

sample ID	depth (m b.s.)	pw   gw (g)	<sup>4</sup> He (ppmv)	<sup>4</sup> He (ccSTP/g <sub>pw/gw</sub> )	<sup>3</sup> He/ <sup>4</sup> He
KR56-2b (NG)	751.9	0.73	931	9.81E-02	-
KR56-3 (NG)	772.2	0.45	8119	1.98E-01	3.16E-08
KR56-5 (NG)	877.5	0.65	5349	2.30E-01	-
KR56-6 (NG)	930.4	0.66	13838	1.45E-01	4.25E-08
KR56-7 (NG)	979.9	0.58	4571	1.52E-01	5.53E-08
KR56-9 (NG)	1053.5	0.43	2552	1.08E-01	-
KR56-13 (NG)	1153.7	0.34	11391	1.99E-01	2.85E-08
KR56-14 (NG)	1156.3	0.43	6839	1.43E-01	8.52E-08
KR65 groundwater	1116.6	-	-	1.61E-02	2.63E-08

## 5. Discussion

Despite <sup>4</sup>He concentrations in porewater varying by about a factor of 2 over the sampled depth interval there is no clear relationship with distance to the nearest water conducting fracture (Fig. 1). With increasing depth, however, measured <sup>4</sup>He concentrations show a slight increase between 750 and 800 m b.s. followed by a roughly gradual decrease down to 1050 m b.s., intersecting a water conducting fracture at 956 m b.s.

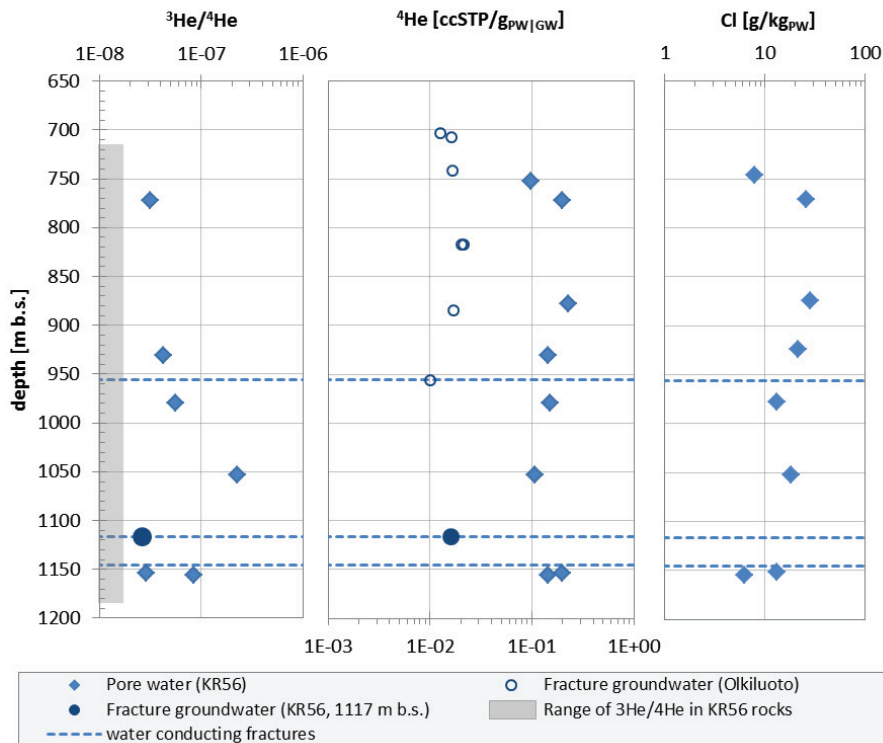


Fig. 1. <sup>4</sup>He concentration and <sup>3</sup>He/<sup>4</sup>He isotope ratio of porewaters and groundwater from borehole OL-KR56 as a function of depth. Additional <sup>4</sup>He groundwater concentrations at similar depths from the Olkiluoto site (obtained from various boreholes) and chloride concentration in OL-KR56 porewater samples are given for comparison. Note: the <sup>4</sup>He concentration of the fracture groundwater at 950 – 960 m b.s. is suspected to be underestimated due to sampling problems and expected to be closer to the value of the groundwater sample above.

While these trends are tentative at best (considering the roughly 30% variability in  $^4\text{He}$  concentrations between the two closely adjacent lowermost samples), it is interesting to note that the observed pattern is mimicked in the OL-KR56 porewater concentrations of conservative tracers such as chloride and bromide (Fig. 1,  $\text{Br}^-$  not shown)<sup>2</sup>. Furthermore, a similar pattern can be observed in the  $^4\text{He}$  concentrations from fracture groundwaters obtained from various boreholes from the Olkiluoto site (Fig. 1), allowing for a (currently unspecified) regional phenomenon.

Measured  $^3\text{He}/^4\text{He}$  ratios in porewaters vary between  $2.8 \times 10^{-8}$  and  $2.2 \times 10^{-7}$  (Table 2), which is mostly higher than the general range of crustal values ( $1 - 3 \times 10^{-8}$ )<sup>6</sup>. More importantly, all OL-KR56 porewater  $^3\text{He}/^4\text{He}$  values are distinctly higher than the range of  $0.8 - 1.9 \times 10^{-8}$  of the porewater's host rocks (Fig. 1). This indicates that the current porewater He inventory cannot be sourced solely by radiogenic in-situ production but requires an additional, less radiogenic component and/or processes such as isotopic fractionation during redistribution of He between the rock's mineral phases and the porewater, as has been observed in the case of the Swiss Molasse Basin<sup>7</sup>. Addition of relevant amounts of isotopically light He by contamination of the samples with air ( $^3\text{He}/^4\text{He} = 1.34 \times 10^{-6}$ ) can be excluded, as the He partial pressure of air (5.24 ppmv) is over 2 orders of magnitude lower than its equivalent in the extracted porewater gases (931 – 13838 ppmv, Table 2). While addition of even small amounts of isotopically light He (e.g. with lithospheric mantle signature,  $^3\text{He}/^4\text{He} = 6.5 - 8.5 \times 10^{-6}$ )<sup>8</sup> could result in the observed isotopic signatures in the porewater, the existence or exact source of such a potential isotopically light He contribution is currently not clear. The isotopic signatures of the sampled groundwater at 1116.6 m b.s. ( $^3\text{He}/^4\text{He} = 2.6 \times 10^{-8}$ ) and another groundwater from the same borehole at approximately 400 m b.s. ( $^3\text{He}/^4\text{He} = 3.1 \times 10^{-8}$ ) are, however, similar to the porewater, suggesting a similar origin for both reservoirs.

$^4\text{He}$  concentrations in the analysed porewater are generally about one order of magnitude higher than in fracture groundwater at similar depth, both from other boreholes and from OL-KR56, regardless of the distance between the two reservoirs (Fig. 1). These high  $^4\text{He}$  concentrations suggest a long residence time in the order of millions of years for the porewater. At the same time, the transient state between porewater and fracture groundwater with respect to the inert tracer He indicates that the timespan during which groundwater with present-day composition is flowing in the sampled fracture at 1116.6 m b.s. has been insufficient for the groundwater/porewater system to reach equilibrium.

Knowledge of  $^4\text{He}$  concentrations and  $^3\text{He}/^4\text{He}$  isotope ratios in the rock and porewater domain is an important factor for interpreting helium data in groundwaters. The continuous production of helium in the rock's various minerals, its release into the porewater and subsequent diffusive redistribution - all in the context of lithological and petrophysical constraints - leads to a complex input function for the assessment of groundwater helium inventories, residence times and interpretation of potential groundwater provenance.

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