## The Low Permeability of the Earth's Precambrian Crust

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The large volume of deep groundwater in the Precambrian crust has only recently been understood to be relatively hydrogeologically isolated from the rest of the hydrologic cycle. The paucity of permeability measurements in Precambrian crust below 1.3 km is a barrier to modeling fluid flow and solute transport in these low porosity and permeability deep environments. Whether permeability-depth relationships derived from measurements shallower than 1.3 km can be extended to greater depths in unclear. Similarly, application of a widely-used permeability-depth relationship from prograde metamorphic and geothermal systems to deep Precambrian rocks may not be appropriate. Here, we constrain permeabilities for Precambrian crust to depths of 3.3 km based on fluid residence times estimated from noble gas analyses. Our analysis shows no statistically significant relationship between permeability and depth where only samples below 1 km are considered, challenging previous assumptions of exponential decay. Additionally, we show that estimated permeabilities at depths >1 km are at least an order of magnitude lower than some previous estimates and possibly much lower. As a consequence, water and solute fluxes at these depths will be extremely limited, imposing important controls on elemental cycling, distribution of subsurface microbial life and connections with the nearsurface water cycle.

### Introduction

Precambrian crust, which makes up  $\sim$ 72% of the Earth's continents by area <sup>1</sup>, has been estimated to host between  $\sim$ 8.5 and 13 million km<sup>3</sup> of groundwater <sup>2,3</sup>. This deep store of mostly saline fluids accounts for 20 to 30% of total continental groundwater. Estimates of groundwater residence times in Precambrian rocks (Figures 1 and 2a), can exceed 1 billion years <sup>2,4–6</sup>, with the longest residence times found in Archean age rocks.

These deep and ancient groundwaters are estimated to contain a substantial portion of the Earth's biomass, with microbial activity found to depths of up to 2-3 km <sup>7-12</sup>. The degree of hydrogeologic – and associated geochemical – isolation from near-surface environments exerts control on the habitability, abundance, and diversity of subsurface microbial life <sup>6,9,11-13</sup>. At these depths, life is isolated from the photosphere and increasingly dependent on chemosynthesis. The supply of electron donors and acceptors exerts and important control on subsurface microbial activity<sup>11</sup>, which is influenced by permeability.

The crystalline rocks of the Earth's Precambrian crust are inherently a low permeability hydrogeologic regime where what fluid flow occurs primarily via fractures. Despite representing a significant proportion of the crust globally (Figure 1), detailed permeability measurements are few, particularly in deep (>1 km) crystalline rock <sup>14–17</sup>. Permeability values are necessary to constrain fluid and solute fluxes in the crust, to define the degree of interconnectivity that might occur between subsurface biomes, and to provide insights into the distribution and connectivity of fracture networks in deep Precambrian rock.

## **Previous Permeability-Depth Relationships**

Permeability typically decreases with depth due to tectonic stresses, compaction, diagenesis and weathering<sup>14,18,19</sup> and permeability-depth relationships have been derived for a variety of environments using various approaches (Table 1). A prime example of such a relationship was generated using data from geothermal and metamorphic environments <sup>15,20</sup> (Figure 2b), which extended down to depths of 40 km, including the 10 km thickness of brittle crust nearest to the ground surface. Permeabilities of geothermal systems were estimated by determining the amount of fluid flow required to produce enough advection

to replicate observed temperature distributions. In contrast, in metamorphic systems, permeabilities were estimated using the time-integrated fluid flux required to produce the observed amount of mineralization. The resulting permeability-depth relationship for geothermal and metamorphic systems has been widely applied to studies of generic regional flow systems <sup>21</sup>, geomechanics of the crust <sup>22</sup>, circulation of deep meteoric fluids<sup>23</sup>, biogeochemical cycles<sup>24</sup> and, following scaling for gravity, Martian hydrogeology<sup>25</sup>.

Ingebritsen and Manning<sup>20</sup> noted that stable tectonic crystalline rock settings ('cratons') where a significant proportion of the world's oldest rocks, including those of Archean age, are located, are likely to have even lower permeability values than the models they developed for areas of tectonic or magmatic activity would predict. We note that there are exceptions to cratons being stable, such as the Precambrian basement in the Yellowstone region, where the Wyoming Craton appears to have become more permeable due to magmatic activity<sup>26</sup>. More recent studies, based on compilations and regression analysis of permeability estimates from a range of in situ and laboratory hydraulic testing techniques, support the idea of lower permeability values in stable crystalline rock <sup>14</sup> and batholiths down to depths of 1.3 km (Figure 2b). Evaluating whether those relationships hold at greater depths is difficult with only one hydraulic test beyond a depth of 1.3 km available in those databases; a value of 10<sup>-19</sup> m<sup>2</sup> measured over a depth interval of 4.3 to 9.0 km in the Kola Superdeep Borehole in Russia<sup>27</sup>.

These previous attempts to estimate permeability are inadequate to understand the past behaviour of fluids in Precambrian rocks. Numerical models have used a wide range of permeability values for these settings, from as low as  $10^{-21}$  for Atikokan, Canada<sup>28</sup> to  $10^{-16}$  m<sup>2</sup> for Fennoscandia<sup>29</sup>. A continental-scale simulation of the Canadian Shield that used a

value of 10<sup>-17</sup> m<sup>2</sup> predicted residence times of 800 ka at a depth of 3.7 km near Kidd Creek, Canada for the mid Pleistocene Epoch and much younger ages under current conditions following flushing during inflow of subglacial meltwater<sup>30</sup>. These values are inconsistent with residence times of 400 to 1,700 Ma estimated from noble gas measurements<sup>2,4</sup>, suggesting that the permeabilities used in the model were far too high.

# **Constraining Permeability with Residence Time Estimates**

To constrain the permeability of stable crystalline rocks beyond 1.3 km, we carried out a novel approach to estimate in situ permeabilities by incorporating noble gas-based residence times of groundwaters in Precambrian rocks at depths from the ground surface to 3.3 km<sup>2,4,5,31</sup> (Figure 2a). Radiogenic noble gases, such as <sup>4</sup>He and <sup>40</sup>Ar, are produced due to radiogenic reactions in the crust and accumulate in groundwaters <sup>32–34</sup>. From measured concentrations of uranium, thorium, and potassium, rates of production of these noble gases can be determined. Combining these with measured concentrations of the noble gases in the groundwater phase can provide effective means to estimate residence times beyond those provided by unstable radioisotopes such as <sup>3</sup>H, <sup>14</sup>C and <sup>81</sup>Kr <sup>35</sup>. Analysis of the noble gas content of fracture fluids sampled from Canada, Fennoscandia and South Africa have revealed mean fluid residence times ranging from a few thousand to over one billion years <sup>2,4,31,36–43</sup>.

The maximum distance groundwater can travel since exposure to the atmosphere is limited by the size of topography-driven flow systems, which is defined by distance between recharge and discharge points as determined by the distribution of hydraulic head. We estimate permeabilities by applying the working assumption that the flow system size provides an upper constraint to permeability because water found within a regional

defined by hydraulic divides. In many cases, the distance travelled will be considerably smaller, particularly where other mechanisms of emplacement, such as burial or tectonics, were involved. While some continental-scale groundwater models have predicted hydraulic divides that indicate the presence of flow systems extending over 100s of km in Precambrian rock<sup>29,44</sup>, these are at odds with smaller scale models that have incorporated more geological complexity and local site geologic and hydrogeologic details. Groundwater models produced for Whiteshell<sup>45,46</sup> and Atikokan<sup>47</sup> in the Canadian Shield, Foresmark<sup>48,49</sup> and Okiluoto<sup>50</sup> in the Fennoscandian Shield, and Moab Khotsong, South Africa<sup>51</sup> have all found that local topography and surface water bodies along with geologic structures exert strong controls on the hydraulic divides that delineate groundwater flow systems. Flow system extents were typically on the order of 10 to 25 km and similar in size to HydroBASINS level 12 catchment size<sup>52</sup>, which we use to estimate the size of topography-driven flow systems at sites where noble gases have been analyzed (see methods section).

We acknowledge that the flow system geometries may have varied over time due to deposition and erosion of sediments overlying cratons <sup>53</sup>, along with folding and faulting<sup>4</sup>. However, the Precambrian rocks themselves have been subject to slow erosion rates over much of their history, suggesting that flow system size may not have changed dramatically for long periods of time<sup>54</sup>. Samples taken from these systems will have travelled less than the full length of the system. Forces other than topographic-relief may also be important (e.g. compaction, tectonic stresses, dewatering reactions, free convection), but those drivers are generally less important and limited in both time and space in terms of developing hydraulic gradients <sup>55</sup>. We estimate permeabilities by assuming that these

fluids have travelled less than 14.7 to 30.0 km based on the dimensions of catchments in Precambrian rocks, which are similar to flow system dimensions used in previous analysis of permeabilities in metamorphic and geothermal environments 15 (see methods section). This distance provides an upper bound for flow system size in Precambrian rocks, where flow systems will likely be shorter and could be driven by factors other than topography.

The permeabilities estimated from the noble gas residence times have the following relationship with depth (Figure 2):

$$log k = -17.07 - 1.44 log z$$
 [1]

where k is permeability in  $m^2$  and z is depth in km. This linear regression has an  $R^2$  value of 0.579, which is significant at a p-value of 0.001. The vast majority of the locations examined here are from Archean settings, with only one sample from Sweden and one from Finland both in Proterozoic rock. Whether Archean cratons and other Precambrian rocks of Proterozoic age that reflect a broader range of structural features, such as rifts, accretionary complexes, and/or metasediments, have significantly different permeability-depth relationships is beyond the scope of this paper, as permeability measurements or estimates for a diverse global set of Proterozoic settings are even fewer than for the Archean systems.

Equation [1] produces log k values that are over 2 lower than the Ingebritsen and Manning (1999) curve at a depth of 3 km. Our log k estimates would increase by 2.0 if either hydraulic gradients were decreased by approximately two orders of magnitude or flow systems were increased by an equal amount. These higher permeabilities are unlikely because longer flowlines are uncommon in Precambrian rock due to the limited geologic continuity and the hydraulic gradients of  $4.7 \times 10^{-4}$  to  $1.4 \times 10^{-3}$  used here are below or near

the global median value of  $1.3 \times 10^{-2}$  <sup>56</sup>. It is plausible these noble gas samples were collected in systems that had higher hydraulic gradients in the geologic past, perhaps approaching 0.1 <sup>56</sup>, which would result in a decrease in our estimated permeabilities by approximately an order of magnitude. There is also considerable evidence that solute transport in deep Precambrian rock is dominated by diffusion <sup>6,57,58</sup>, which indicates that permeabilities less than  $10^{-20}$  m<sup>2</sup> are common<sup>59</sup> (Figure 2).

This study suggests that permeabilities are at least one order of magnitude lower than those predicted by the regression of in situ hydraulic tests compiled by Achtiziger-Zupancic et al.  $^{14}$  (Table 1) for stable crust (i.e. cratons). Those tests likely had support volumes on length scales of a few m up to a few  $100 \text{ m}^{60}$  and may not reflect the effect of discontinuities in the fracture network on solute transport that would affect groundwater ages. In order for the permeabilities found here to match those found from hydraulic testing, flow system lengths would need to increase by an order of magnitude, which is inconsistent with detailed studies of the regional scale flow systems in cratons  $^{28,45,47-51}$ . These results could also be achieved if porosity was increased to  $\sim 10\%$ , which is inconsistent with measurements for cratons  $^{58}$ , or if hydraulic gradients were reduced by an order of magnitude. However, it should also be noted that the median log k for depths > 1 km in the Achtziger-Zupancic et al.  $^{14}$  database is  $^{-19.0}$ , which is similar to those found in the analysis presented here. Data at those depths were not considered in the regression analysis for cratons and shields in that study.

It is instructive to compare the results obtained here with those for batholiths (large masses of relatively homogeneous intrusive igneous rock)<sup>16</sup> (Table 1; Figure 2), which likely reflect the lower-end of permeability relative to Precambrian rock as a whole. The

upper limit of permeabilities estimated here are approximately two orders of magnitude greater than those predicted by a relationship proposed for the equivalent porous media values for batholiths. The higher permeabilities appear to be plausible given that environments considered here include settings that with higher degrees of fracturing such as ore deposits  $^{4,42}$ , areas within impact structures  $^{2,6}$  and fracture zones within batholiths  $^{16}$ . Reducing flow system lengths to  $\sim 100$  m could bring the estimates here into closer agreement with the relationship for batholiths. However, if the flow system lengths used here are approximately correct, unrealistically low porosity ( $\sim 0.01\%$ ) $^{61}$  or excessively high hydraulic gradients, approaching 1, would need to be present.

Permeability is elevated in the upper 1 km in Precambrian rock, which supports the concept that enhanced permeability in shallow (<1 km) crystalline rocks is largely a function of weathering  $^{62,63}$  along with unloading and tectonics (Figure 3). This shallow zone to  $\sim 1$  km also corresponds to the approximate depth where meteoric and paleometeoric waters are typically found to penetrate in the Canadian Shield, Fennoscandian Shield and Witwatersrand Basin  $^{5,23}$ . Groundwater flow is more active in the upper 1 km and limited by permeability at greater depths.

Likely the most important outcome of this exercise is when only data deeper than this 1 km zone is considered, there is no significant correlation between estimated permeability and depth (log k and log z, respectively) at the p=0.1 level. Permeability below 10 km has also been shown to have a weak relationship with depth, although that has attributable due to its position below the brittle-ductile transition<sup>20</sup> and does not explain the lack of relationship between 1.0 and 3.3 km. In sedimentary environments, permeabilities generally decrease from the ground surface to depths of several km due to

compaction and diagenesis <sup>19,64</sup> but a similar trend is not obvious in Precambrian rock below 1 km. Any trends related to geomechanical and geochemical processes that are a simple function of depth could be overwhelmed by the long and often complex burial and exhumation histories of Precambrian rocks<sup>53</sup>. The apparent increase in permeability from Canada to Fennoscandia to South Africa hints at the importance of differences in the geological histories of these settings that promise to be important issues for future studies. The presence of younger groundwaters at depth in the Witwatersrand Basin, and in the Sudbury Impact Crater on the Canadian Shield, may be the result of the high degree of fracturing related to the impact events forming both basins<sup>2,6</sup>. The widespread presence of paleometeoric waters at depth in the Fennoscandian Shield<sup>65</sup> suggests the presence of interconnected fracture networks and elevated permeability. There is also the possibility that these fluids were emplaced in the past when permeabilities were elevated <sup>17,66</sup>, which would make current permeabilities lower than those estimated here.

These estimated permeability values overestimate actual values because the transit distances of groundwater in Precambrian rock are likely substantially less than the full length of the topography-driven flow system and associated flow system lengths used here (Figure 3). Sleep and Zoback <sup>67</sup> proposed that 1 km long flow systems in fractured Precambrian rock could support sufficient geochemical fluxes to sustain microbial activity. If this length was used rather than full flow system length, estimated permeabilities would be approximately an order of magnitude lower than the upper estimates presented here, reinforcing the overall conclusion. The noble gas analyses determine the period of hydrogeologic isolation from atmospheric recharge events, but it is important to note that these fracture fluids are the net product of groundwater circulation, original syn-

depositional fluids, and subsequent fluid history and water-rock reaction <sup>4,5</sup>. Hence burial, negative buoyancy, and tectonic forcing may have been important mechanisms that would result in shorter transit distances and lower permeability estimates, as would the inherently hydrogeologically discontinuous nature of sparsely fractured rock.

The overall coherence between the He-Ne-Ar-Xe derived noble gas residence times at each site support a model of hydrogeologic isolation<sup>2</sup>. However, Warr et al<sup>6</sup> recently demonstrated that these settings actually represent a spectrum from being fully isolated to fully open to diffusive transport. At sufficiently low diffusion coefficients (10<sup>-15</sup> m<sup>2</sup>/s), there will be no appreciable loss of any noble gases <sup>57</sup>. At slightly higher diffusion coefficients, noticeable diffusive transport of He and Ne will occur, while Ar, Kr, and Xe are retained 6,57. Relating these low rates of diffusion to permeability is not straightforward. While laboratory testing of core samples has found correlations between permeability and diffusion coefficients <sup>68–70</sup>, there is no universally agreed upon relationship between these two parameters. The lowest diffusion coefficients found in the laboratory studies were a few orders of magnitude higher than those required to prevent differential diffusion of noble gases and suggest permeabilities < 10<sup>-21</sup> m<sup>2</sup>. For porosities and hydraulic gradients similar to those estimated in this study, fluids could migrate distances up to a few 100 m over a time period of a billion years, keeping them isolated from the surface. If the relationships found by Kuva et al.<sup>69</sup> holds for lower diffusion coefficients, bulk permeabilities in Precambrian crust could be as low as  $10^{-25}$  m<sup>2</sup>. The focus in these studies is on diffusive transport, as advective transport would be limited to distances of a few cm over billion-year time scales at these low permeabilities. Manning and Ingebritsen<sup>15</sup>

suggested a transition between diffusion-dominated environments and advection-dominated environments occurs at a permeability of  $10^{-20}$  m<sup>2</sup>.

The permeabilities estimated here, along with previous findings on diffusion rates of noble gases, bring into question the results of prior numerical simulations of groundwater flow in Precambrian rock at depths >1 km. Simulations that used higher permeabilities will have overestimated flow rates and underestimated residence times, potentially by a few orders of magnitude.

# Permeability and Life in the Deep Subsurface

Studies of the deep subsurface biosphere have to date suggested there is evidence for a depth component associated with microbial communities, with Proteobacteriadominated communities at shallower depths of  $\sim$ 1 km  $^{10}$ , while Firmicutes-dominated communities are thought to be more common at depths > 1 km. This pattern has been observed in South Africa <sup>10,36,71</sup> and Fennoscandia <sup>72,73</sup>, while patterns in Canada are not yet apparent due to a sample bias to date towards very deep sites alone. These biome boundaries correspond to a general geochemical transition, with changes observed in total dissolved solids (TDS), redox conditions (Eh) and a general transition from meteoric and paleometeoric waters to shield-type brines with  $\delta^2 H$  and  $\delta^{18} O$  values that plot to the left of the global meteoric water line (GMWL) 5,37,74. This shift in stable water isotope values at depths > 1 km is the result of water-rock reactions, including oxygen isotopic exchange between waters associated with hydrothermal/metamorphic activity and the host rocks over very long (Ma+) time periods 5,75. The position of this transition zone approximately corresponds to the transition from the upper more permeable zone of the Precambrian crust to lower permeability environments at depth (Figure 3). The reduced permeability at

these depths restricts fluid and solute fluxes and transport is likely dominated by diffusion on a regional scale, and fracture-controlled fluid only locally<sup>76,77</sup>. These lower fluxes can affect cycling and migration of elements in the subsurface related to microbial life (e.g. CHNOPS) and have been proposed to exert an important control on the distribution and composition of microbial communities<sup>74,76,78</sup>.

## **Conclusions**

Residence times estimated from noble gas analyses of deep groundwaters suggest that the permeability of Precambrian crust in stable cratons is at least approximately 2 orders of magnitude lower than previous estimates from prograde metamorphic and geothermal environments and an order of magnitude lower than a relationship based on hydraulic tests in the upper 1 km of Precambrian crust <sup>14</sup> and possibly approaching values measured in batholiths<sup>16</sup>. Importantly, permeability estimates based on noble gas data measured between 1.0 and 3.3 km no longer show a statistically significant correlation with depth. The limited diffusion rates in these environments imply that the permeabilities are likely even lower than those estimated here, and considerably more dependent on lithologic setting and local geological history, including events such as impact fracturing, than previously considered.

The low permeabilities of Precambrian rock suggest that microbiological processes in this deep biosphere are more likely to be limited by fluid and solute fluxes and more dependent on diffusive transport than they are in other environments. As a consequence, microbial communities at depths in Precambrian rock will likely be more isolated than in other geological environments and, as a consequence, will be slower to respond (if at all) to changes in surface and near-surface Earth system processes.

### Methods

Groundwater residence time  $(\tau)$  is calculated with:

$$\tau = \frac{L}{\frac{k\rho g}{un}\nabla h}$$
 [2]

where L is flow system length, k is permeability,  $\rho$  is fluid density,  $\mu$  is viscosity,  $\eta$  is porosity, and  $\nabla h$  is the hydraulic gradient (Figure 1). Here, we rearrange this equation to estimate permeability (k):

$$k = \frac{L\nabla h\mu\eta}{\tau\rho g}$$
 [3]

L was determined from the maximum length of the HydroBASINS level 12 polygons<sup>52</sup> containing the sample site. This length was similar to flow system lengths found in detailed studies of the flow systems at a subset of the sample sites, including Whiteshell<sup>45,46</sup>, Okiluoto<sup>50</sup>, Foresmark<sup>48,49</sup> and Moab Khotsong<sup>51</sup>, which found that flow systems are affected by local topography and surface water bodies and bound by geological structures on scales of  $\sim$ 10 to 25 km. This length is also similar to previous treatment of flow system dimensions in metamorphic and geothermal environments<sup>15</sup>. Hydraulic gradients were estimated using L from HydroBASINS and the minimum and maximum topographies for those polygons<sup>79</sup>. We use the residence times, depths and porosities from various noble gas studies (Supplementary Data 1). Where porosities were not available in those studies, we use a porosity of 1% based on a number of previous studies that have reviewed porosity in Precambrian rocks  $^{2,3,17,56,58}$ .

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## **Data Availability**

Noble gas and porosity data compiled in this study are available in Supplementary Data 1 and can be accessed at 10.6084/m9.figshare.23713047.

## **Competing Interests Statement**

All authors declare no competing interests.

## **Author Contributions**

GF and JCM conceived of this study and led the writing of this paper. Methodology was developed by GF, who also performed calculations. Calculations were performed GF and data was compiled by GF and OW. GF, JCM, OW and BSL contributed to writing and editing.

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 $Table\ 1: Permeability-depth\ relationships\ derived\ from\ previous\ studies.$ 

Environment	Permeability-Depth	Method	Maximum
	Relationship		Depth (km)
Prograde	log k = -14 – 3.2 log z	Geothermal and	28.4
metamorphic <sup>15,20</sup>		metamorphic analysis	
Tectonically active	log k = -11.5 – 3.2 log z	Metamorphic and seismic	38
continental crust		analysis	
(dynamic) <sup>80</sup>			
Upper crust <sup>81</sup>	$log k = -25.4 + 13.9(1+z)^{-0.25}$	Various	5
Crystalline rock	log k = -16.26 – 1.53 log z	Hydraulic testing	1.0
(various			(measurements
environments) <sup>14</sup>			below 1.0 km not
			used in
			regression)
Stable shields and	log k = -16.12 – 1.35 log z	Hydraulic testing	1.0
platforms <sup>14</sup>			
Canadian Shield	log k = -21 +(5.55/[1+	Hydraulic testing	1.3 (22
(equivalent porous	$(z/0.151)^{4.2}]^{0.1919}$		measurements
medium)			below 1 km)

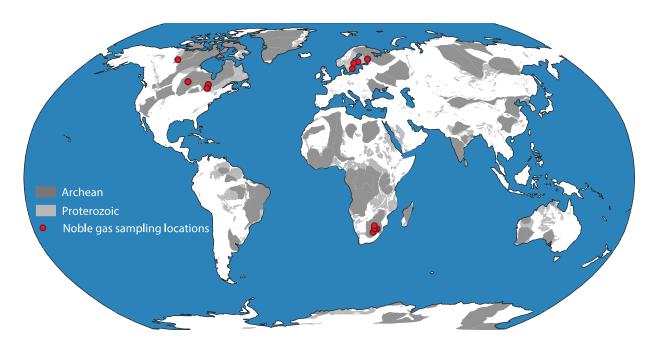


Figure 1: Global distribution of Archean cratons (exposed and buried) $^{82}$  and Proterozoic rock $^{83}$  showing locations of noble gas-derived residence time data used to estimate permeabilities in this study $^{2,4,31,36-43}$ .

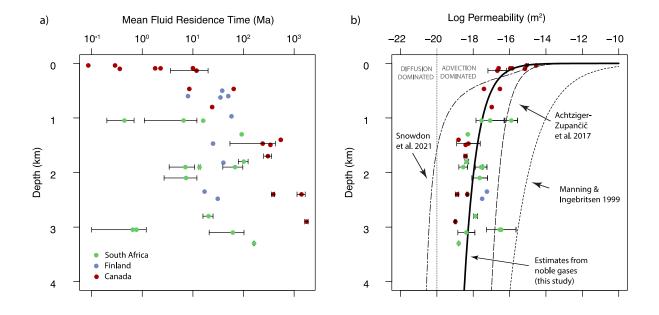


Figure 2. Residence times estimated from noble gases provide constraints on the distribution of permeability with depth. a) Fracture water residence times estimated from noble gas analyses from Warr et al.<sup>5</sup> and references therein show an increase with depth. b) Permeability estimates from groundwater residence times are lower than those expected from Ingebritsen and Manning's <sup>20</sup> permeability-depth relationship and those for stable provinces found by Achtziger-Zupančič et al. <sup>14</sup> but higher than the relationship found by Snowdon et al <sup>16</sup>. Error bars in both figures are based on the minimum and maximum residence times where provided by previous studies.

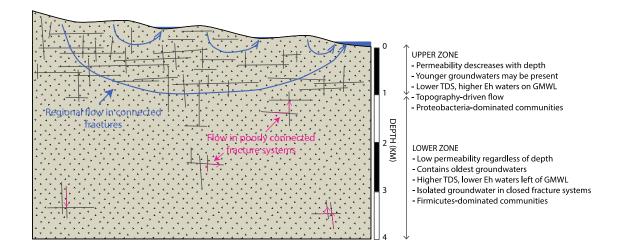


Figure 3: The permeability estimates in this study are consistent with the conceptual model of an upper zone characterized by decreasing permeability with depth that contains lower TDS, higher Eh waters with stable water isotope ( $\delta^2 H$  and  $\delta^{18} O$ ) values that plot near the Global Meteoric Water Line (GMWL); and a lower zone characterized by low permeability without a strong relationship with depth that contains higher TDS, lower Eh waters that plot to the left of the GMWL . The upper zone hosts protobacteria-dominated communities, while the lower zone tends to contain Firmicutes-dominated communities  $^{10,33}$ .