Deep Meteoric Water Circulation in Earth's Crust

Jennifer C. McIntosh[1,2,*,†] and Grant Ferguson[1,2,†]

1. Hydrology and Atmospheric Sciences, University of Arizona, Tucson, AZ 85721 USA
2. Civil, Geological and Environmental Engineering, University of Saskatchewan, Saskatoon, SK CA

**corresponding author: jenmc@email.arizona.edu; 520-626-2282*

†These authors contributed equally to this work.

Key Points

- Maximum circulation depths of meteoric waters vary considerably from <1 to 5km across North America
- The deepest meteoric water circulation occurs in mountainous terrains in western North America
- Topographic gradients and fluid density are primary controls on the extent of meteoric water circulation, rather than permeability

Abstract

Deep meteoric waters comprise a key component of the hydrologic cycle, transferring water, energy, and life between the earth's surface and deeper crustal environments, yet little is known about the nature and extent of meteoric water circulation. Using water stable isotopes, we show that the maximum circulation depths of meteoric waters across North America vary considerably from <1 to 5 km, with the deepest circulation in western North America in areas of greater topographic relief. Shallower circulation occurs in sedimentary and shield-type environments with subdued topography and shallow brines. The amount of topographic relief available to drive regional groundwater flow and flush saline fluids is the primary control on the extent of meteoric water circulation, rather than permeability. The presence of an active flow system in the upper few km of the Earth's crust and stagnant brines trapped by negative buoyancy offers a new framework for understanding deep groundwater systems.

Plain Language Summary

Deep circulation of waters, coming from precipitation, connects the Earth's surface with deeper subsurface environments, transferring water, energy and life critical for key processes, such as deep mineral weathering and release of nutrients, and geothermal energy systems. Deeper, more saline groundwater is typically only weakly connected to the rest of the hydrologic cycle. The penetration depth of precipitation-derived waters and the bottom of the more active hydrologic cycle is relatively unknown. This study shows the depth of meteoric water circulation varies considerably across North America as a function of topography and fluid density, rather

than permeability. Study results help constrain locations of deeper meteoric water penetration and potential hydrologic connections to the earth's surface, which has important implications for the extent of water resources and transport and long-term storage of anthropogenic contaminants in the subsurface.

Index Terms and Keywords

1829-Groundwater hydrology

1836-Hydrological cycles and budgets (1218, 1655)

1832-Groundwater transport

1402-Critical Zone

1. Introduction

The extent and controls on deep groundwater circulation are poorly understood, creating challenges for groundwater resource assessment (Gleeson et al., 2016; Richey et al., 2015), waste isolation (Cherry et al., 2014; Ferguson, McIntosh, Perrone, et al., 2018), integration of groundwater into catchment hydrology (Condon et al., 2020; Frisbee et al., 2017) and Critical Zone science (Küsel et al., 2016), and the distribution and evolution of life in the subsurface (Lollar et al., 2019; Warr et al., 2018). Permeability exerts an important control on the rate of groundwater circulation (and groundwater age) and there have been a number of attempts to assess the variations in permeability with depth (Achtziger-Zupančič et al., 2017; Ingebritsen & Manning, 1999; Stober & Bucher, 2007). Permeability generally decreases with depth and residence times increase, however there is no conclusive evidence that groundwater circulation would cease due to the low permeabilities found at depth (Ingebritsen et al., 2006).

There have been comparatively few studies that have examined the extent of meteoric groundwater circulation through compiling geochemical and isotopic evidence. An examination of the origin of waters in sedimentary basins in North America suggested that topography and fluid density control the extent of meteoric water circulation rather than permeability (Ferguson, McIntosh, Grasby, et al., 2018). That study demonstrated that there is insufficient topography to flush dense brines from the deepest extents of many basins, despite sufficient permeability. These results were in agreement with δ^2H and $\delta^{18}O$ values that fell beneath the GMWL or a range of other geochemical measures, such as low Cl:Br, that indicated that there was residual paleo-evaporated seawater present in the basin. Here, we build on those findings to assess the depth to which flushing by meteoric water would be possible in different geologic terrains at the continental scale using water stable isotopes (δ^2H and $\delta^{18}O$). We show that the maximum circulation depth varies considerably over a range of geological environments across North America and this appears to be associated with the amount of topographic relief available to overcome the negative buoyancy associated with the density of saline fluids at depth.

2. Distribution of Meteoric Waters

Meteoric waters typically have δ^2H and $\delta^{18}O$ values that fall near the global meteoric water line (GMWL) (Craig, 1961), and this can be used to delineate groundwaters that originate as precipitation and have not been significantly modified by water-rock reactions or mixing with non-meteoric fluids (Ferguson, McIntosh, Grasby, et al., 2018) (Figure 1). Non-meteoric waters that deviate from the GMWL can be identified in terms of deuterium excess (D excess) relative to the GMWL (Dansgaard, 1964):

$$Relative\ D\ excess\ = \delta^2 H - 8\ x\ \delta^{18} O \qquad [1]$$

Recognizing that shifts away from the GMWL can occur due to changes in $\delta^{18}O$, this can also be expressed as oxygen depletion (^{18}O depletion) (Kloppmann et al., 2002):

$$Relative\ ^{18}O\ depletion\ = \delta^2 H/_8 - \ \delta^{18} O \qquad [2]$$

At the local scale, meteoric waters plot along local meteoric water lines that have slightly different slopes and intercepts from the GMWL, depending on local climatic conditions. These local deviations may over- or underestimate the relative D excess and ^{18}O depletion (Kloppmann et al, 2002), and alter the maximum circulation depths approximated in this study.

Deeper groundwaters that originated as evaporated seawater (e.g., sedimentary basin brines) typically have δ^2H and $\delta^{18}O$ values that plot beneath the GMWL (i.e. negative D excess and negative ^{18}O depletion values) (Kharaka & Hanor, 2003). High-temperature (geothermal) waters plot to the right of the GMWL, enriched in ^{18}O from high temperature isotope exchange with minerals; also displaying negative apparent D excess values (Truesdell & Hulston, 1980). Deep saline waters in cratonic (shield-type) environments often plot to the left of or above the GMWL due to low temperature water-rock interactions at low water to rock ratios that have modified either seawater, hydrothermal fluids, and/or meteoric water over long time periods (Fritz & Frape, 1982; Warr et al., 2020). Fluids that have interacted with CO_2 can also plot to the left of the GMWL or to the right (Karolytė et al., 2017).

Here we examine δ^2H and $\delta^{18}O$ data from water and energy wells and mine inflows to determine the maximum depth of meteoric water circulation. We supplement these data with estimated circulation depths for thermal springs where δ^2H and $\delta^{18}O$ values of discharged waters fall along the GMWL.

3. Methods

3.1. Databases and Mapping

The primary databases used to compile δ^2H and $\delta^{18}O$ data from wells for this study were the USGS Produced Waters database (Blondes et al., 2016) and data compiled for the Canadian Shield (Stotler et al., 2012). These data were supplemented by data from additional studies (Clark et al., 1998; Mariner & Janik, 1995; McIntosh et al., 2002, 2008, 2010; Osburn et al., 2019; Zhang et al., 2009). These datasets were culled to consider only those samples that provided a well location and depth. Additional data from the USGS NAWQA dataset (USGS, 2020) were also used to understand the distribution of meteoric water with depth in this study, but were not considered during mapping because of the shallow depth of most water supply wells and associated groundwater quality monitoring.

3.2. Estimating Meteoric Water Circulation Depths

Meteoric waters are typically defined as waters with δ^2H and $\delta^{18}O$ values falling near the GWML. However, meteoric waters vary in their distance from the meteoric water line due to a range of processes, such as partial evaporation and convective air mass mixing that create local meteoric water lines or paleorecharge under different climatic conditions (Jasechko, 2019). Tolerances for where meteoric waters fall around the GMWL are not typically defined quantitatively. Here, we consider waters with D excess values falling between -10 and 30‰ (20‰ variability in δ^2H or 2.5‰ variability in $\delta^{18}O$ around GMWL) as meteoric waters.

To supplement δ^2H and $\delta^{18}O$ data from wells, we used studies that have estimated maximum temperatures from aqueous geothermometry on samples collected from thermal springs discharging meteoric water (Davisson et al., 1994; Frisbee et al., 2017; Grasby et al., 2016; Grasby & Hutcheon, 2001; Mayo & Loucks, 1995; Pepin et al., 2015). Those studies used local geothermal gradients to estimate the circulation depth required to obtain those maximum temperatures.

The maximum depth of circulation was estimated by determining the maximum depth of water samples with a D excess value falling between -10 and 30‰ or estimated circulation depth of a thermal spring with a meteoric water isotope signature based on a 2 degree by 2 degree grid across North America. Over much of North America, especially outside of oil and gas producing regions, the availability of deep samples is limited, and our mapped results are likely to underestimate the depth to which meteoric water is present. In addition, the approach used here underestimates meteoric water circulation depth by not considering deeper meteoric waters that have been isotopically-altered through low or high temperature water-rock reactions, or through isotopic exchange with CO_2. Where no samples deeper than 500 m were available, the grid spaces were left blank during mapping.

3.3. Assessment of Topography and Driving Force Ratio

We assess the possibility that the distribution of meteoric waters is controlled by the amount of topography available to drive regional groundwater flow and the negative buoyancy of dense, saline fluids at depth. The relative importance of these two factors is described by the driving force ratio (*DFR*), which is defined as follows (Bachu, 1995):

$$DFR = \left(\frac{\Delta\rho}{\rho_o}\frac{|\nabla E|}{|\nabla h|_o}\right) \quad [3]$$

where ρ is the fluid density, ρ_o is a reference density (commonly assumed to be 1,000 kg/m^3), $|\nabla h|_o$ is the magnitude of the hydraulic gradient based on a reference density and $|\nabla E|$ is the magnitude of the average structural gradient of the groundwater flow system (i.e. the slope that the water must travel along to exit the groundwater system). This approach was originally intended to assess errors arising from using potentiometric maps based on reference densities, but has been extended to examine where dense brines would be trapped by negative buoyancy in sedimentary basins (Ferguson, McIntosh, Grasby, et al., 2018). In this case, the condition necessary for waters to stagnate can be described as:

$$\nabla h_o = \frac{\nabla\rho}{\rho_o}\nabla E \quad [4]$$

For systems where the water table closely follows the topography and the highest hydraulic head (h_o) in the flow system overlies the deepest point of the flow system, [3] can be approximated as:

$$h_o = \frac{\rho - \rho_o}{\rho_o} z_{max} \qquad [5]$$

Where z_{max} is the maximum circulation depth. Where h_o is insufficient to overcome the density contrast, dense waters below z_{max} are isolated from the overlying topographically-driven flow system and will not discharge to surface water bodies.

Here, we use topographic drops as a proxy for the maximum hydraulic head change. Maximum topographic drops were calculated from the USGS GETOPO 30 digital elevation model (USGS, 1997) on a 2 degree x 2 degree grid across North America using QGIS. We chose a gridded approach because there are many areas of North America where there are no obvious boundaries for deep groundwater flow systems in many cases. Permeability contrasts associated with geological contacts have been used to constrain these in some studies (Ferguson, McIntosh, Grasby, et al., 2018), but this approach is problematic outside of sedimentary basins. Watershed-based approaches are also problematic because deep groundwater flow often transfers water between watersheds (Fan, 2019). These topographic drops are then compared to the sample depths in each grid block.

4. Maximum Circulation Depth

The maximum circulation depth of meteoric waters in North America ranges from less than 1 km in eastern North America to approximately 5 km in the west (Figure 2). Deeper circulation depths occur in areas of greater topographic relief and the greatest circulation depths are associated with thermal springs. The shallowest circulation depths are associated with oil/gas produced waters in sedimentary basins and mines in crystalline bedrock.

Lithology does not appear to exert a strong control on circulation depth. The circulation depths in the Canadian Shield are similar to many sedimentary basins in midcontinent North America, despite the large differences in permeability (Figure 3). The extent of meteoric water circulation in the Canadian Shield roughly coincides with the depth where bulk permeability approaches the matrix permeability at ~1 km (Achtziger-Zupančič et al., 2017). While permeability and meteoric water circulation appear to coincide in the Canadian Shield, examination of other environments suggests that permeability might not be the primary controlling factor.

In sedimentary basins, decreases in permeability with depth do not explain the extent of meteoric water circulation. Over much of central North America, circulation depths are less than 2 km. Relatively high permeability (>10^{-16} m^2) sandstone and carbonate aquifers are present at the bottom of many sedimentary basins (> 2 km depth) (Figure 3). Yet these basal aquifer systems often contain non-meteoric waters, derived from paleo-evaporated seawater (Bein & Dutton, 1993; Ferguson et al., 2007; Stueber & Walter, 1991). Conventional oil and gas production and saltwater disposal are common in these deep strata (Ferguson, 2015; Scanlon et al., 2019; Zhang et al., 2016), indicating that appreciable groundwater flow rates are possible where hydraulic gradients are sufficiently high. We hypothesize an alternate mechanism for trapping saline fluids at depth in sedimentary and crystalline environments - due to negative buoyancy.

The deepest circulation of meteoric groundwater is found in thermal springs in mountainous areas of western North America. The median circulation depth of the 38 springs compiled here was 2.6 km and this approach is thought to underestimate circulation depth due to geochemical re-equilibration of waters as they interact with the rock mass as they rise toward the discharge area, and due to mixing of waters from different depths (Ferguson et al., 2009). The results presented here are similar to those found in the Alps (Diamond et al., 2018). Very little is known about the permeability distribution of these systems from direct measurements, but numerical modelling indicates that country rock values on the order of 10^{-16} m^2 are required to supply a sufficient amount of water to a fault to support the formation of thermal springs (Forster & Smith, 1989).

The link between topography and circulation depth indicates that the forces driving circulation may exert a stronger control than permeability on how deep meteoric water penetrates into the Earth's crust. Deep groundwaters that do not fall on the GMWL typically have salinities that are several times that of seawater (Fritz & Frape, 1982; Kharaka & Hanor, 2003). These highly saline waters appear to be ubiquitous at depth in both sedimentary environments (Kharaka & Hanor, 2003) and in crystalline bedrock (Stotler et al., 2012; Warr et al., 2018) and are also thought to be present in the lower crust (Manning, 2018). Due to their high salinities (TDS~300 g/L), these waters have densities that approach 1,200 kg/m^3 (Adams & Bachu, 2002). For regional groundwater flow systems where the water table coincides with the ground surface, the topographic drop to depth ratio must exceed 0.2 for a 1,200 kg/m^3 density brine to allow for it to be flushed by meteoric water (equation 4). Actual topographic drop to depth ratios required for flushing would have a critical value greater than 0.2, as regional hydraulic gradients are less than topographic gradients.

Meteoric waters with D excess values between -10 and 30‰ and ^{18}O depletion values from -1.25 to 3.75‰, corresponding to the GMWL, tend to have large topographic drops relative to their depths. Where topographic drop to depth ratios of less than 0.2 and trapping due to negative buoyancy is expected, D excess values tend to be less than -10‰ and ^{18}O depletion values tend to be less than -1.25‰ (Figure 4). The most negative D excess and ^{18}O depletion values (i.e., most saline basin brines) are associated with topographic drop to depth ratios less than ~1, although a variety of D excess and ^{18}O depletion values are found at these ratios. Relative ^{18}O depletion and apparent D excess values exceeding +3.75‰ and +30‰, respectively (i.e., shield-type brines) tend to be associated with topographic drop to depth ratios of ~1.

Many of the samples with relative D excess and ^{18}O depletion values outside of the range expected from meteoric waters have topographic drop to depth ratios greater than the critical value (0.2) or higher than is required to displace a brine with a density of 1,200 kg/m^3 (see equation 3) (Figure 4). Many of these samples likely have a component of meteoric water that is actively circulating or are residual brines that are currently being flushed by regional groundwater flow. Flushing may take extended periods of time, especially in low permeability lithologies such as shale and salts. Other samples that plot in this region could be trapped by negative buoyancy due to the overestimation of the hydraulic gradient by using topography as a proxy (e.g., in areas with deep water tables that are a subdued reflection of surface topography) or by underestimating the structural gradient (e.g., instances where the highest hydraulic head values does not overlie the maximum circulation depth) (Ferguson, McIntosh, Grasby, et al., 2018). The most ancient shield-type brines may be trapped in isolated fractures that are not hydrologically connected to active circulation systems (Warr et al., 2018).

5. Conclusions - Rethinking the Extent of the Deep Hydrological Cycle

Many previous studies have assumed that groundwater resources extended to 1 or 2 km globally (Gleeson et al., 2016; Nace, 1969; Richey et al., 2015). The remarkable spatial variability of circulation depth suggests that previous estimates of the volume (Gleeson et al., 2016; Nace, 1969; Richey et al., 2015) and residence times of groundwater at global scales (Befus et al., 2017) are likely incorrect and misleading if translated to the regional scale.

Topography and variations of fluid density with depth exert a strong control on the extent of the meteoric water circulation in the crust. This represents a paradigm shift in hydrogeology, which has focused on permeability decreases as a primary constraint on the circulation of groundwater at depth (Ingebritsen & Gleeson, 2017). Global assessments of bulk permeability have suggested that groundwater flow is possible over most of the brittle crust, which extends to a depth of ~10 km (Ingebritsen & Manning, 1999). Our results indicate that circulation of meteoric water outside of orogenic belts is largely restricted to the upper 1 to 2 km, regardless of permeability and is influenced by topography and negative buoyancy. The inability of meteoric water to circulate to depths exceeding more than ~1 to 2 km over large areas of continents is consistent with observations of very old, saline waters at these depths in both cratons (Holland et al., 2013; Lippmann et al., 2003; Warr et al., 2018) and sedimentary basins (Castro et al., 1998; Zhou & Ballentine, 2006). It is also consistent with penetration depths of meteoric waters that have recently been in contact with the atmosphere based on the presence of tritium (Gleeson et al., 2016) and radiocarbon (Jasechko et al., 2017).

These results showing the importance of topographic gradients and fluid density elicit a change in how we characterize hydrogeologic systems. We have few tools other than sampling deep wells, boreholes or mines to characterize groundwater salinity and residence times at depth. In particular, deep wells are few and far between in mountainous regions (Markovich et al., 2019) – areas with the deepest meteoric water circulation – and beyond ~1 km in crystalline shield-type environments. The need for geophysical or other techniques to address the extent of meteoric groundwater in the Earth's crust represents a major challenge for the geosciences.

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(2012), Clark et al. (1998), Mariner and Janik (1995), McIntosh et al. (2002, 2008, 2010), Osburn et al. (2019), Zhang et al. (2009), and USGS (2020).

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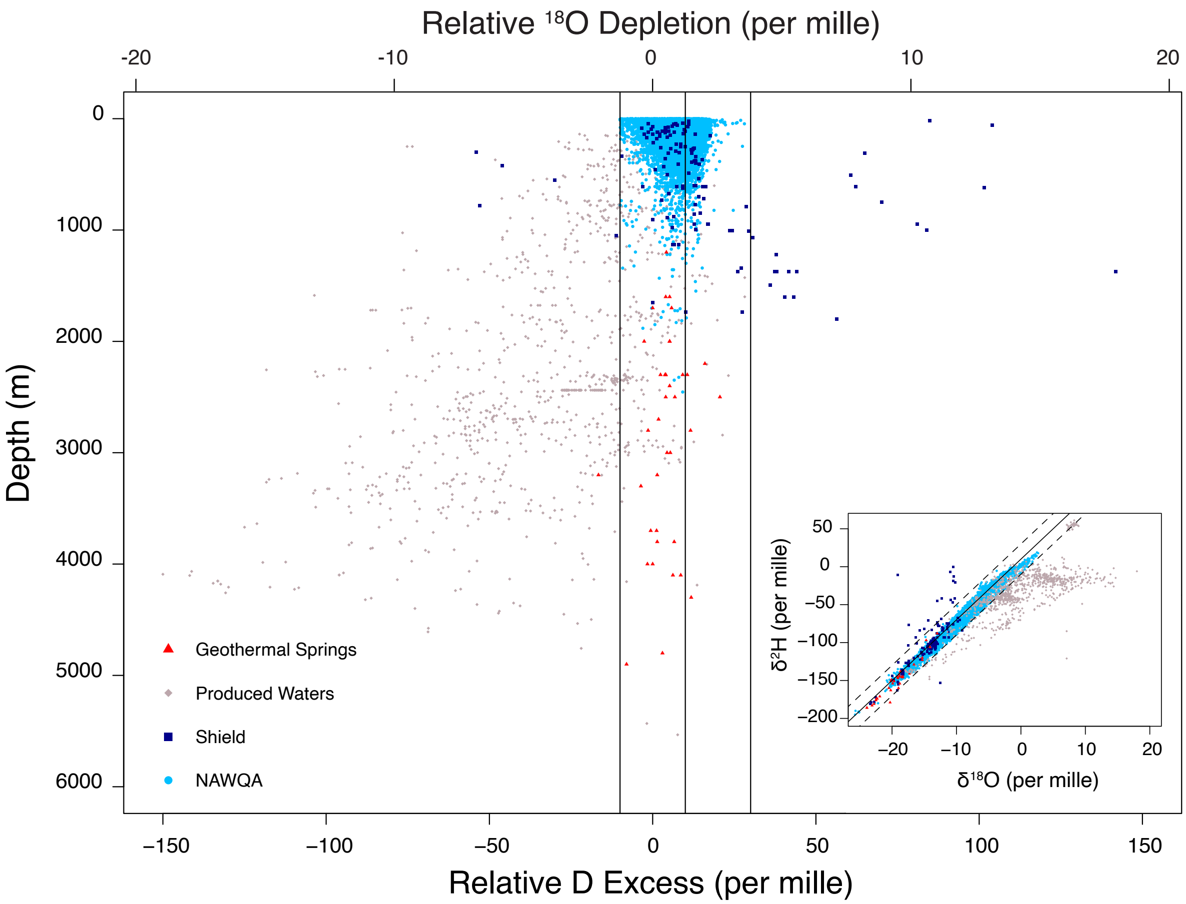

Figure 1: Depth of meteoric waters based on water stable isotopes. The relative deuterium excess and ^{18}O depletion generally decreases with depth showing a transition from waters with a meteoric origin (relative D excess between -10 and 30‰ or relative ^{18}O depletion from -1.25 to 3.75‰) to more negative values associated with sedimentary basin brines. Waters from several hundreds of m deep in the Canadian Shield tend to plot with more positive relative ^{18}O depletion and apparent D excess values. There is a great degree of variability with depth, reflecting different circulation depths of meteoric water and variability in the D excess values in deep brines. Inset shows the relationship between δ^2H and $\delta^{18}O$ in relation to the GMWL and lines plotted at D excess values +/- 20‰ of the GMWL. Values from the NAWQA database, tend to fall on the GMWL (Craig, 1961) as do most geothermal springs in North America. Produced

waters from sedimentary basins often reflect mixing of meteoric waters with paleo-evaporated seawater source and plot to the right of the GMWL. Waters from depths of several 100s of m in the Canadian Shield plot often to the left of the GMWL. See methods section for data references.

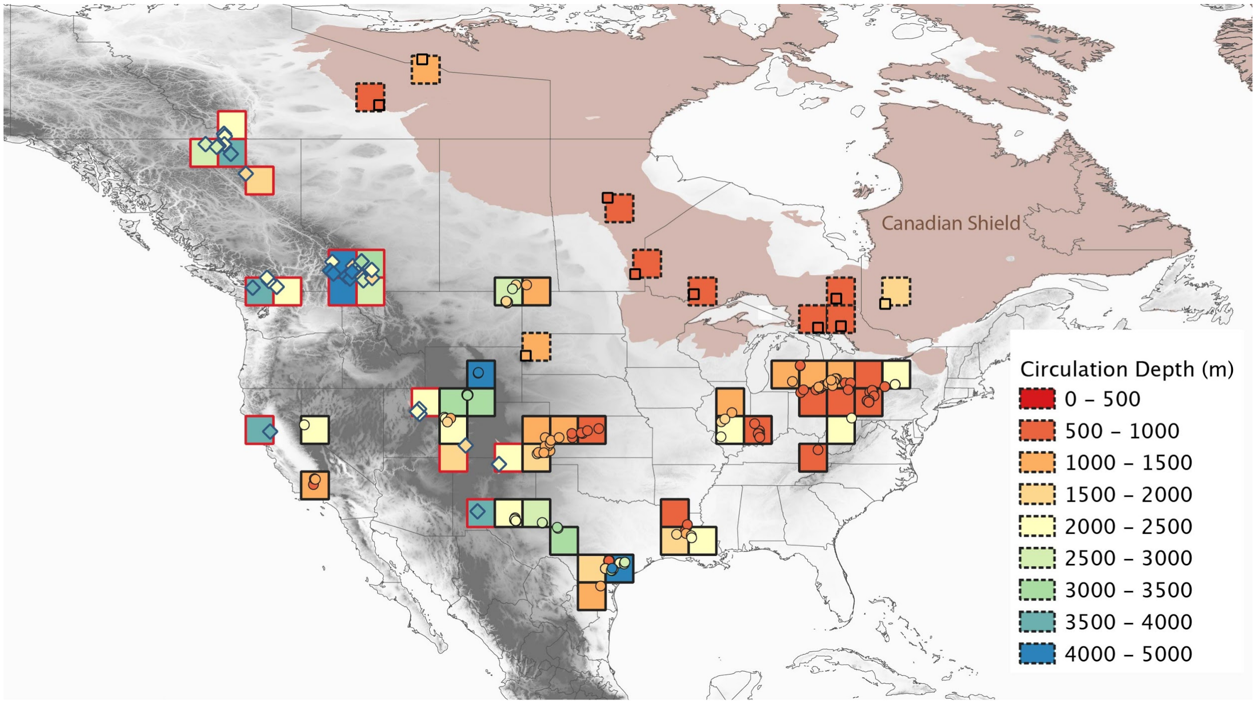

Figure 2: Meteoric water circulation depth across North America. Depth of circulation as estimated from deepest sample with D excess value within 20‰ of the GMWL in a 2 degree by 2 degree grid. Squares with solid black outlines are from the USGS produced waters database (Blondes et al., 2016) plus additional references in Methods, red outlines from estimates based on geothermometry from springs (see Methods for references) and dashed outlines samples from mines or other projects in Precambrian rock (see Methods for references).

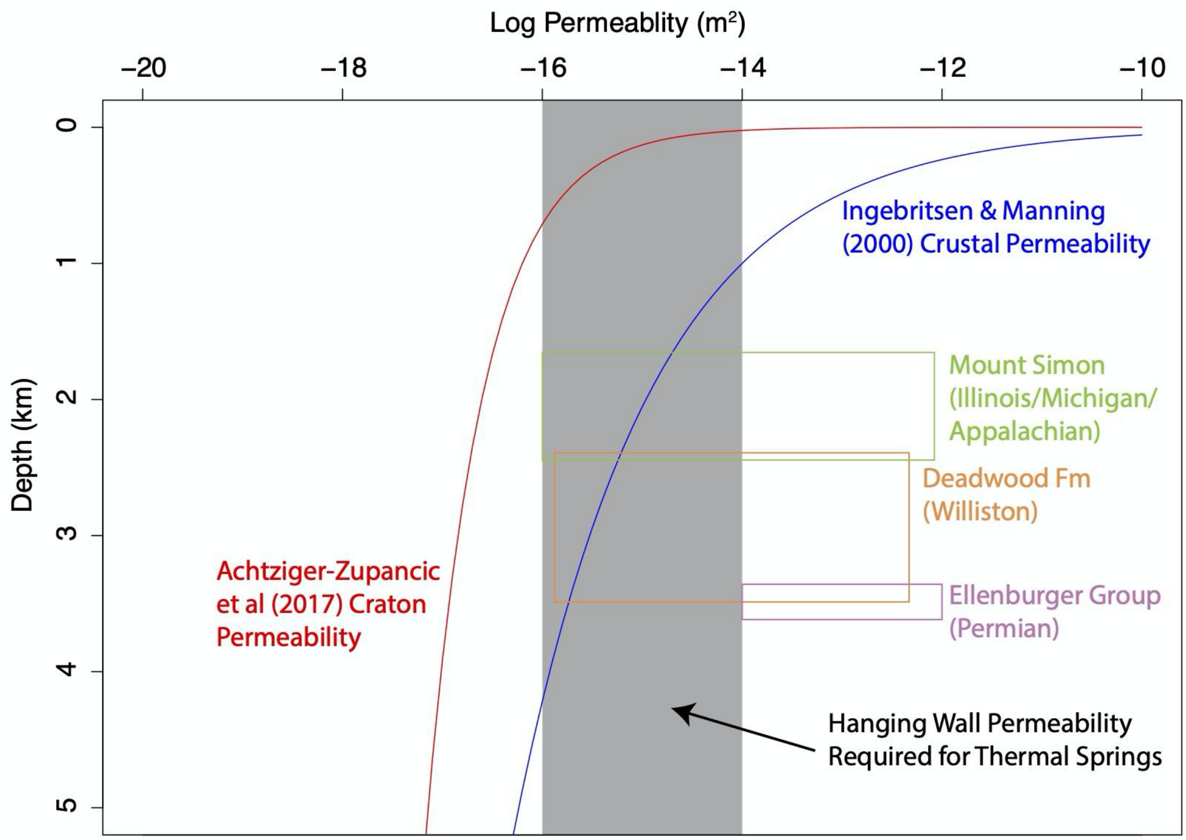

Figure 3: Available permeability for fluid circulation. Global databases (Achtziger-Zupančič et al., 2017; Ingebritsen & Manning, 1999) show a tendency for permeability to decrease with depth but elevated permeabilities exist in sedimentary basins (Medina et al., 2011; Phillips, 2019; Y. Zhang et al., 2016), yet non-meteoric, paleo-evaporated seawaters persist. Numerical models of thermal springs indicate elevated permeability is present to depths of several km in orogenic belts (Forster & Smith, 1989).

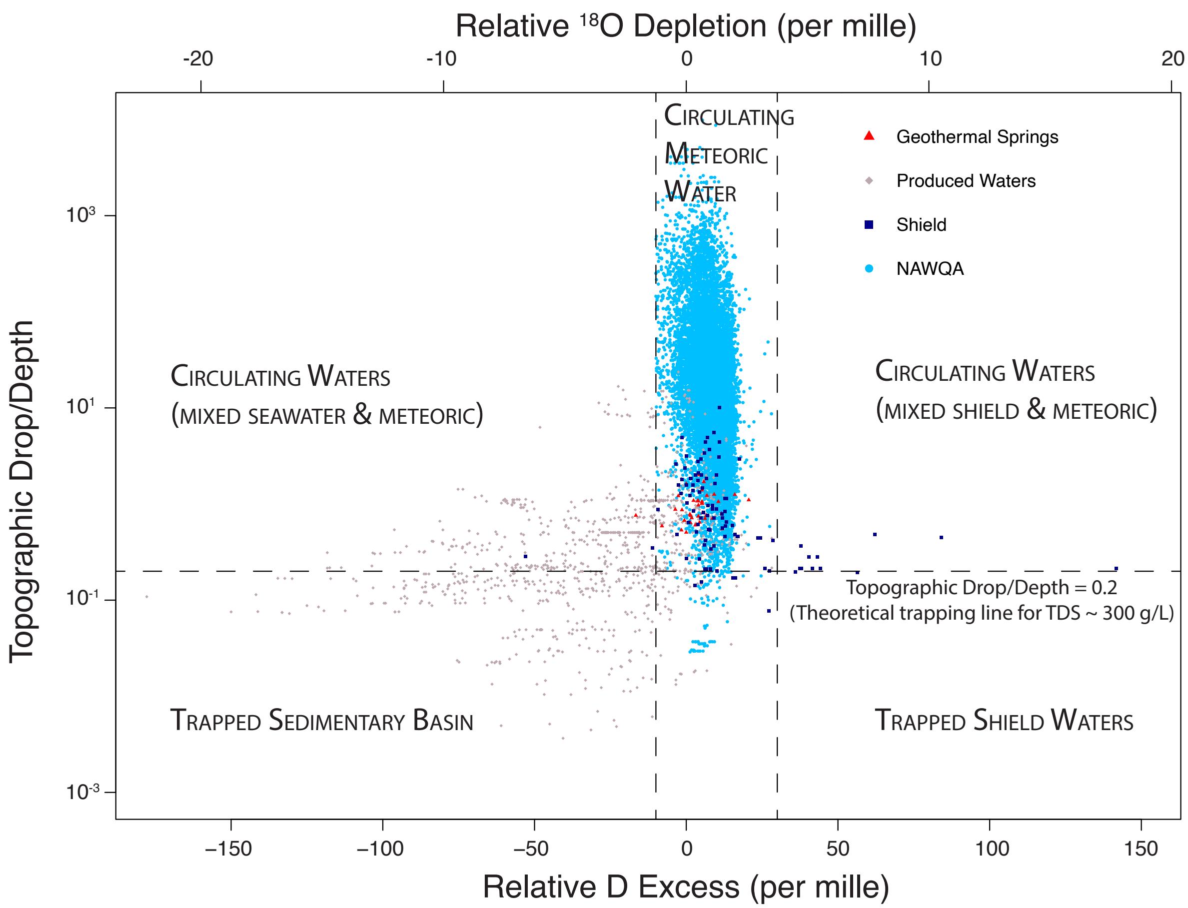

Figure 4: Prediction of circulating vs stagnant fluids based on topographic gradients. Relative D excess between -10 and 30‰ (relative ^{18}O depletion from -1.25 to 3.75‰) values indicating meteoric waters mainly occur where topographic drop to depth ratios exceed 0.2, which is the theoretical critical values required for a brine with a density of 1,200 kg/m^3 to be trapped by negative buoyancy. Negative D excess indicative of non-meteoric, paleo-evaporated seawater derived brines tend to plot at low topographic drop to depth ratios. Strongly positive ^{18}O depletion (and apparent D excess) values found in shield environments tend to plot at topographic drop to depth ratios near the critical value for trapping by negative buoyancy.