

## Iceland Deep Drilling Project (IDDP): Stable Isotope Evidence of Fluid Evolution in Icelandic Geothermal Systems

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### ABSTRACT

The Reykjanes and Krafla geothermal systems, located within the active rift zone of Iceland, are both sites that will be drilled to 4-5 km by the Iceland Deep Drilling Project (IDDP). To effectively characterize geochemical and hydrologic processes occurring at these depths, it is essential to establish the source, composition and evolution of geothermal fluids. We use oxygen and hydrogen stable isotopes in hydrothermal minerals to resolve the fluid history in these IDDP geothermal systems. Here we report the results from existing drillholes to depths of  $\leq 3$  km.

The stable isotope composition of hydrothermal epidote in the Reykjanes geothermal system demonstrates a complex history of fluid source and fluid-rock interaction since at least the Pleistocene. The chlorine concentration of modern Reykjanes geothermal fluids indicate that they are hydrothermally modified seawater. However, measured hydrogen isotope values of these fluids are as low as -23‰.  $\delta D$  values of hydrothermal epidote from wells RN-8, -9, -10 and -17 collectively range from -60 to -78‰, and  $\delta^{18}O_{EPIDOTE}$  in these wells are between -3.0 and 2.3‰. The analyzed epidotes are not in stable isotopic equilibrium with present-day geothermal fluids, but retain an isotopic signature of glacially derived fluids occurring early in the evolution of the geothermal system. Estimates of the water-rock ratio and modal abundance of hydrous alteration minerals in the geothermal system suggest that there is sufficient relict (Ice Age) hydrogen in the altered basaltic host rock to diffusively exchange with modern geothermal fluids and lower the fluid hydrogen isotope composition by as much as 20‰.

Fluid elemental and isotope chemistry studies of geothermal fluids from Krafla present evidence of a local, meteoric fluid source. Additionally, oxygen isotope compositions of present-day geothermal fluids are not significantly more positive than local meteoric water, indicating either limited fluid/rock interaction or an extremely high water to rock ratio. Preliminary hydrogen isotope values of epidote in the Krafla geothermal system are between -116 and -125‰ in wells K-20, K-34, and K-26, and are highly variable between wells. Oxygen isotope compositions of epidote in these wells are between -9.6 and -13.0‰. The variability observed spatially and with depth in the Krafla system is likely due to complex subsurface hydrology and multiple potential fluid sources, including a

significant input of magmatic fluids. Additional analysis of hydrothermal alteration minerals within the Krafla system is necessary to fully resolve the fluid evolution within this geothermal field.

### 1. INTRODUCTION

The active volcanic zone of Iceland is host to more than twenty high-temperature geothermal systems. These systems collectively provide 484MW of geothermal energy (Arnórsson et al., 2008) and represent an important natural resource in Iceland (Arnórsson, 1995). They also offer an opportunity to study, in situ, geochemical interaction between basaltic rocks (magmatic products of the Iceland mantle plume) and aqueous electrolyte solutions at elevated temperatures. Chemical mass transfer in these magma-hydrothermal environments is of fundamental importance because it affects the evolution of ocean chemistry resulting from hydrothermal alteration at spreading ridges (Muehlenbachs and Clayton, 1981) and is a basic process controlling the formation of massive sulfide deposits (e.g. Weissberg et al., 1979).

The Iceland Deep Drilling Project, which intends to increase both the economic and scientific utility of these geothermal systems, will be drilling wells to a depth of 4-5 km within the Reykjanes and Krafla geothermal regions (Figure 1; Elders and Fridleifsson, 2010). Before we can fully understand the geochemical and hydrologic processes occurring at depth within these regions and thus maximize the benefit of this exploration, it is essential to establish the source, composition and evolution of geothermal fluids. Previous authors (e.g. Árnason, 1977; Darling and Ármannsson, 1989; Arnórsson, 1978 and 1995) used the major element chemistry and stable isotope composition of geothermal fluids to determine their source. This study undertakes a more critical look at fluid evolution in the Reykjanes and Krafla geothermal systems, using the stable isotope systematics of hydrothermal epidote. We aim to establish a more complex fluid history in both systems than is apparent from the chemical and isotopic characteristics of the geothermal fluids alone.

### 2. BACKGROUND

#### 2.1 Reykjanes

The Reykjanes geothermal system is located on the southwestern tip of the Reykjanes Peninsula, on the landward extension of the Mid-Atlantic Ridge (Figure 1a). Like other high-temperature systems in Iceland, it is composed of highly fractured basalt lavas and hyaloclastites

that have been intruded by shallow dikes and sills. These intrusions become more frequent with increasing depth (Kristmannsdóttir, 1983; Franzson et al., 2002). Fractures and faulting in the region is due to extension along a NE-

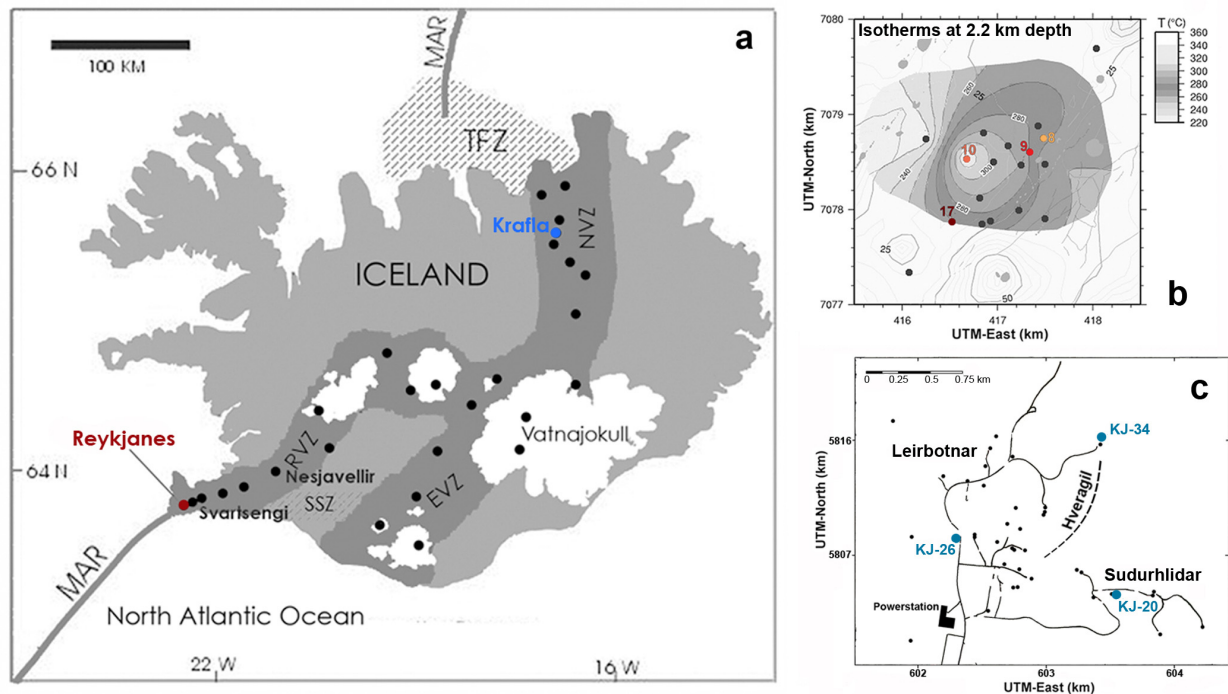
SW trending graben zone, marking the sub-aerial continuation of the Reykjanes Ridge (see Arnórsson, 1995 for summary). The host rocks of Reykjanes have extensively reacted with hydrothermal fluids, forming common secondary minerals that include zeolites, mixed-layer clays, chlorite, epidote and actinolite. These minerals display a distinct zoning with depth, and are used to classify hydrothermal alteration into temperature- and depth- related zones (e.g. Franzson et al., 2002; Fridleifsson et al., 2005). Reykjanes is a two-phase system and the highest recorded downhole temperature in the system is about 320 °C (Figure 1b).

The Reykjanes geothermal system is considered to be dominated by seawater-derived fluids due to the high chloride content and total dissolved solids observed in this region relative to other high-temperature fields in Iceland, which have a meteoric source (e.g. Krafla and Nesjavellir), or a mixed meteoric-seawater source (e.g. Svartsengi), as shown in Figure 2a (Arnórsson, 1995). A seawater-derived fluid source is also likely given the system's proximity to the coast, and the highly permeable and heavily fractured host-rocks, which allow for an influx of seawater into the hydrothermal system (Arnórsson, 1978; 1995). However, hydrogen isotope values of Reykjanes fluids are as low as -23.1‰ (Figure 2b). This is inconsistent with an exclusively seawater-sourced geothermal fluid, but is comparable to the isotopic composition of Svartsengi fluids (~ -24‰), which are considered to be a mixture of ~70% seawater and 30% meteoric water.

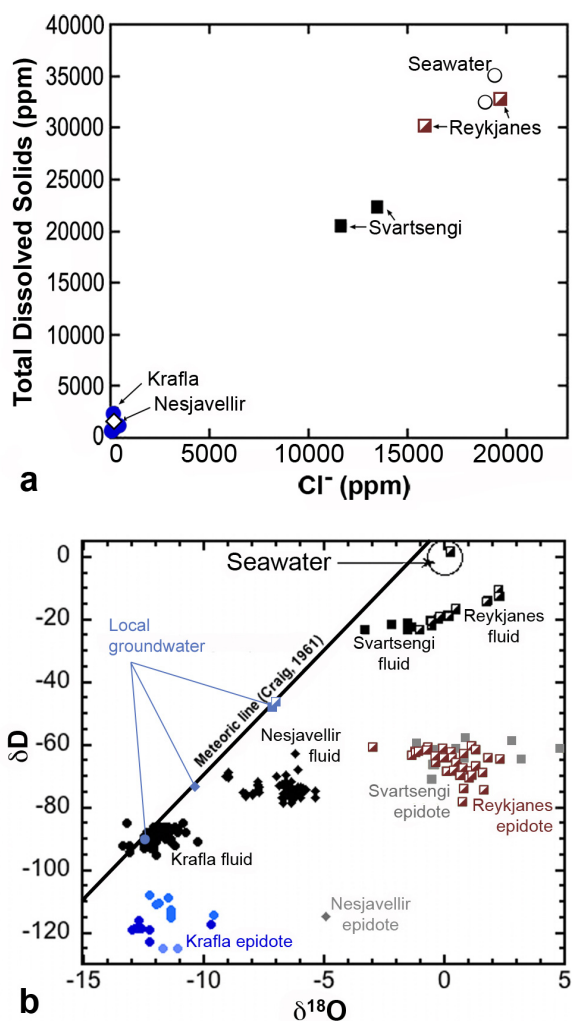
## 2.2 Krafla

The Krafla geothermal system is located within the active Krafla caldera of the Northern Volcanic Zone (Figure 1a). The system is divided into three distinct well fields due to a highly variable distribution of temperature with depth (Arnórsson, 1995). Of the two most productive well fields (shown in Figure 1c), Sudurhlidar shows a classical two-phase system in which temperatures follow the boiling point curve with depth. In contrast, the Leirbotnar well field shows a temperature-depth profile that remains constant at ~205 °C for the upper kilometer, and then follows the boiling point curve below that depth. This pattern is the result of an aquiclude that occurs at approximately 1000-1500 m depth, and separates an Upper and Lower Zone of fluid circulation in this field (Ármansson et al., 1987; Darling and Ármansson, 1989). The aquiclude apparently pinches out beneath Hveragil, a topographic low that outlines a NE-SW trending fault swarm and is associated with a row of explosive craters. The primary upflow zone of the Krafla system is located along this region. The high temperatures of a third, smaller well field, located to the south and not shown in Figure 1c, are attributed to the ascent of hot fluids along the Krafla caldera rim.

The complexity of Krafla geothermal fluids is not apparent in the fluid elemental and isotope chemistry shown in Figures 2a and b. Both present evidence of a meteoric fluid source that has an isotopic composition similar to local precipitation (Darling and Ármansson, 1989). However, a closer look at the chemistry and physical properties of individual wells reveals several anomalies.



**Figure 1:** a. Geologic map of Iceland. Filled circles = high-temperature geothermal fields. MAR = Mid-Atlantic Ridge; RVZ = Reykjanes volcanic zone; EVZ = Eastern volcanic zone; NVZ = Northern volcanic zone; TFZ = Tjörnes fault zone; SSZ = South Iceland Seismic Zone. b. Reykjanes geothermal field. Filled circles represent wells; temperature isotherms of wells at 2200 m depth are designated by shading, isotherm contour interval = 20 °C. c. Krafla geothermal field. Adapted from Pope et al. (2009), and courtesy of Iceland Deep Drilling Project



**Figure 2: a. Fluid chemistry of Icelandic geothermal fields. b. Isotopic composition of Icelandic geothermal fluids, compared to the meteoric water line, seawater, local groundwater and the isotopic composition of hydrothermal epidote from those geothermal systems. Adapted from Pope et al. (2009)**

First, in the Upper Zone of Leirbotnar temperatures are lower than expected if fluids were a product of boiling from the Lower Zone. Instead, there must be mixing with a cold groundwater recharge from the north. Second, Krafla Mountain, located to the northeast of Hveragil, forms a hydrologic barrier between the Sudurhlidar well field and high-elevation regions to the north that recharge the Leirbotnar field. Recharge to Sudurhlidar is more likely coming from a more distant southern region, but it is not clear where from or how groundwater is crossing fault barriers along the southern caldera rim (Darling and Ármannsson, 1989). Finally, the high CO<sub>2</sub> measured in Krafla steam indicates that as much as 15% of the hydrothermal fluids in the system are of magmatic origin (Árnórsson, 1995).

### 3. METHODS

The stability of epidote in hydrothermal systems is a sensitive function of temperature, permeability and fluid compositions (Árnason and Bird, 1992). Thus, we can use the hydrogen and oxygen isotope composition of

hydrothermal epidote sampled from the drill cuttings of active geothermal wells as a record of the average isotope composition of the fluids from which they precipitated. Using experimental temperature-dependent oxygen and hydrogen isotope fractionation curves we can then characterize the isotopic properties of the geothermal fluids, given temperatures approximated from the boiling point curve with depth.

Epidote grains were hand separated from drillhole cuttings from four wells in Reykjanes and three wells in Krafla from depths between 900 and 3000 m. Oxygen and hydrogen isotope analyses of the epidotes were done at the Biogeochemical Stable Isotope Laboratory at Stanford University, following the methods of Sharp (1990) and Sharp et al. (2001). Isotope compositions of the samples were corrected relative to National Bureau of Standards samples as well as laboratory standards, and are correct within  $\pm 1.4\text{‰}$  for hydrogen and  $\pm 0.5\text{‰}$  for oxygen of accepted values. Results of these analyses are presented in the standard delta notation as parts per thousand (‰), relative to the V-SMOW standard.

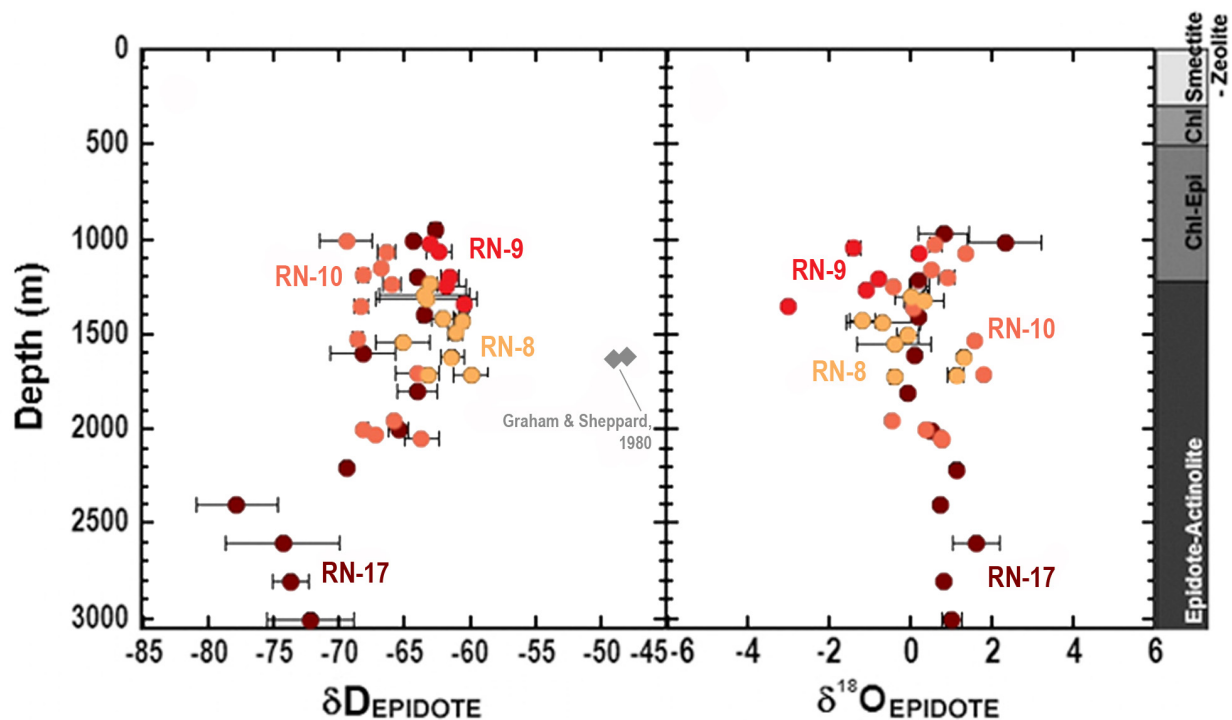
## 4. RESULTS AND DISCUSSION

### 4.1 Reykjanes Geothermal System

Hydrogen and oxygen isotope analysis of geothermal epidote from Reykjanes are shown in Figure 3 as a function of depth. δD values of geothermal epidote from wells RN-8, -9, -10 and -17 range from -60 to -63‰, -61 to -63‰, -64 to -70‰ and -63 to -78‰, respectively. δ<sup>18</sup>O values of epidotes are between -1.2 and 1.3‰, -3.0 and 0.2‰, -0.5 and 1.8‰, and finally between -0.1 and 2.3‰ (Pope et al., 2009). Each well has epidotes with negative δ<sup>18</sup>O values, which requires some meteoric or glacial component to the fluid-source. The variability of the δ<sup>18</sup>O data is likely due to heterogeneities in the extent of water/rock interaction throughout the system, which will control the amount of isotope exchange that occurs between the hydrothermal fluids and altered host rock (Criss and Taylor, 1986).

$1000\ln\alpha_{\text{EPIDOTE-H}_2\text{O}}$  was determined for δD and δ<sup>18</sup>O using measured epidote isotope values and published fluid isotope values (Pope et al., 2009), and is plotted relative to published fractionation curves in Figures 4a and b. Temperatures for fractionation between Reykjanes epidotes and fluids are approximated from the boiling point curve with depth. Hydrogen isotope fractionation between Reykjanes epidotes and geothermal fluid is lower than equilibrium values estimated by the Chacko et al. (1999) published fractionation curve. In contrast, oxygen fractionation correlates closely with the epidote-water fractionation curve based on the combined experimental data from Matsuhisa et al (1979), Matthews et al. (1983) and Kohn and Valley (1998). The δD of the fluids that would be in equilibrium with Reykjanes epidotes (~ -35‰) suggests a significant component of meteoric- or glacially-derived fluids not observed in the modern system.

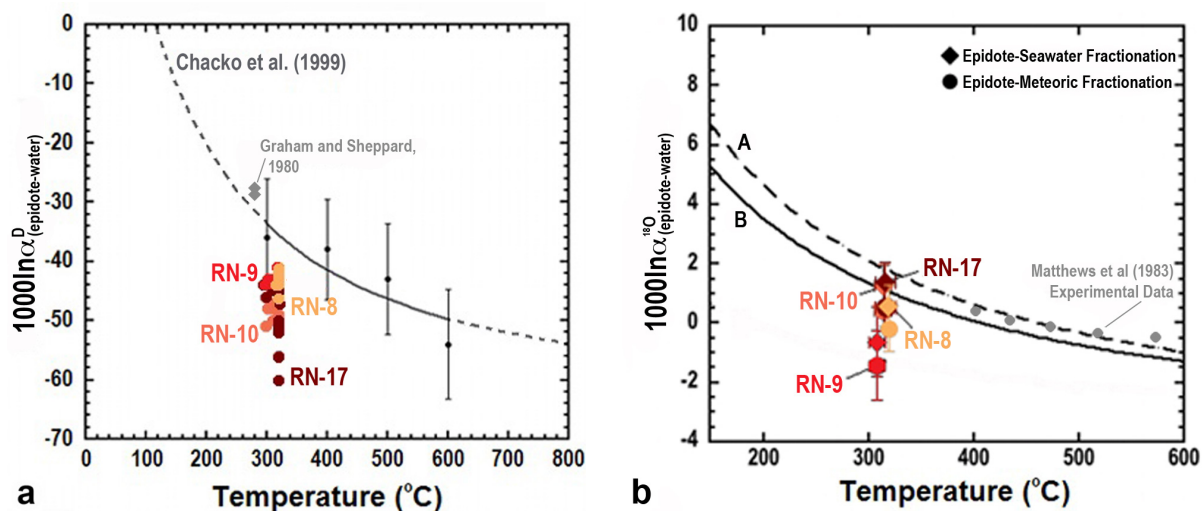
Evidence from stratigraphic and fluid inclusion analyses suggests that the Reykjanes geothermal system originated in a subglacial environment (e.g. Franzson et al., 2002). Hence, alteration minerals would initially have formed in isotopic equilibrium with glacially derived geothermal fluids. Deglaciation of the Reykjanes peninsula was followed by incursion of seawater in the hydrothermal system. Hence, the relict alteration minerals reacted with modern seawater-derived hydrothermal solutions.



**Figure 3: Hydrogen and oxygen isotope composition of Reykjanes geothermal epidote for wells RN-8, RN-9, RN-10 and RN-17 as a function of depth, shown relative to approximate mineral-zone boundaries. Published  $\delta D$  values for well RN-8 by Graham and Sheppard, 1980 are shown for comparison. Adapted from Pope et al. (2009)**

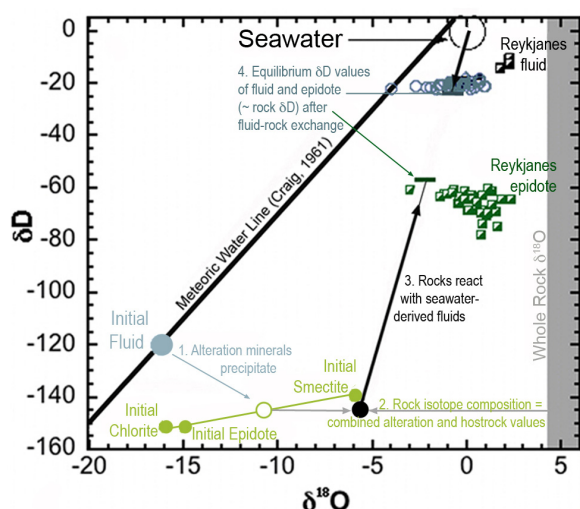
Figure 5 is a graphical representation of this process, in which we approximate the evolution of the Reykjanes system using a simple mass-balance model (Pope et al., 2009). Early in the evolution of the system, hydrothermal epidote, chlorite and smectite form in isotopic equilibrium with initial glacially derived fluids. The values shown in Figure 5 (Step 1) are calculated based on published fractionation curves and are used to approximate the whole rock  $\delta D$  value of the early system (Step 2; black filled circle). The line between this initial rock composition and

seawater (the modern fluid source) represents exchange between these two endmembers (Step 3). When the fluid-rock isotope exchange has followed this reaction path to the point where the  $\delta D$  value of the rock is equal to our measured  $\delta D_{\text{EPIDOTE}}$ , the corresponding hydrogen isotope composition of the hydrothermal fluid is  $\sim -20\text{‰}$  ( $\delta D_{\text{FLUID}}$  – blue open circles), which is on average within 1‰ of observed geothermal fluid values. The altered rock and hydrothermal fluids will continue to exchange until a new equilibrium is reached, shown as Step 4 in Figure 5.



**Figure 4: a. Hydrogen isotope fractionation between average measured Reykjanes geothermal fluid ( $\delta D \sim -20.8\text{‰}$ ) and geothermal epidote from wells RN-8, -9, -10 and -17 as a function of temperature, compared to temperature-fractionation curve by Chacko et al. (1999). b. Oxygen isotope fractionation between epidote and geothermal fluid (circles), and between epidote and seawater (diamonds) in the same wells. Error bars indicate  $1\sigma$  from average composition and temperature. Compared with temperature-isotope fractionation data by Matthews et al. (1983) and curves A (Zoisite-Water from Matthews et al., 1983, Matsuhisa et al., 1979) and B (Epidote-Water from Matthews et al., 1983, Matsuhisa et al., 1979; Kohn and Valley, 1998). Adapted from Pope et al. (2009)**

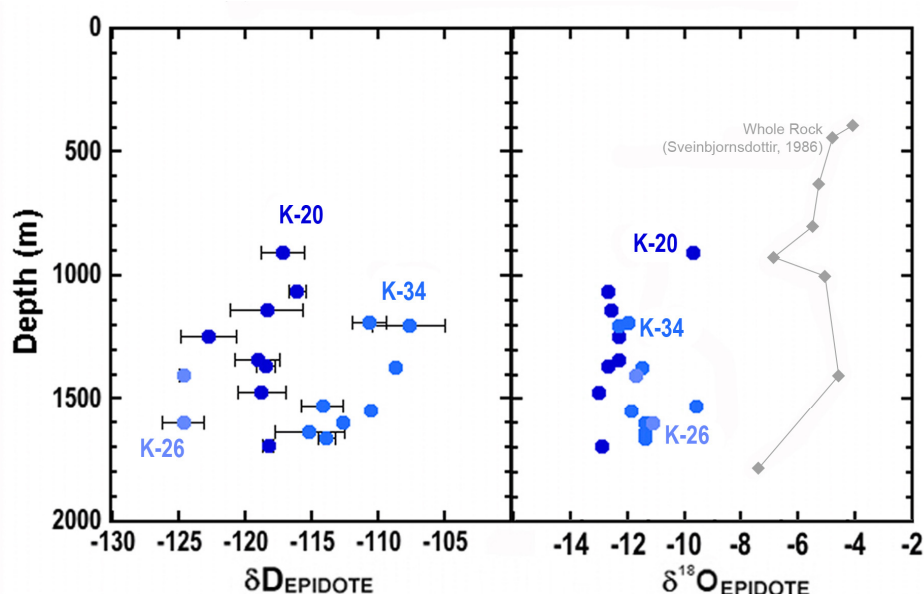




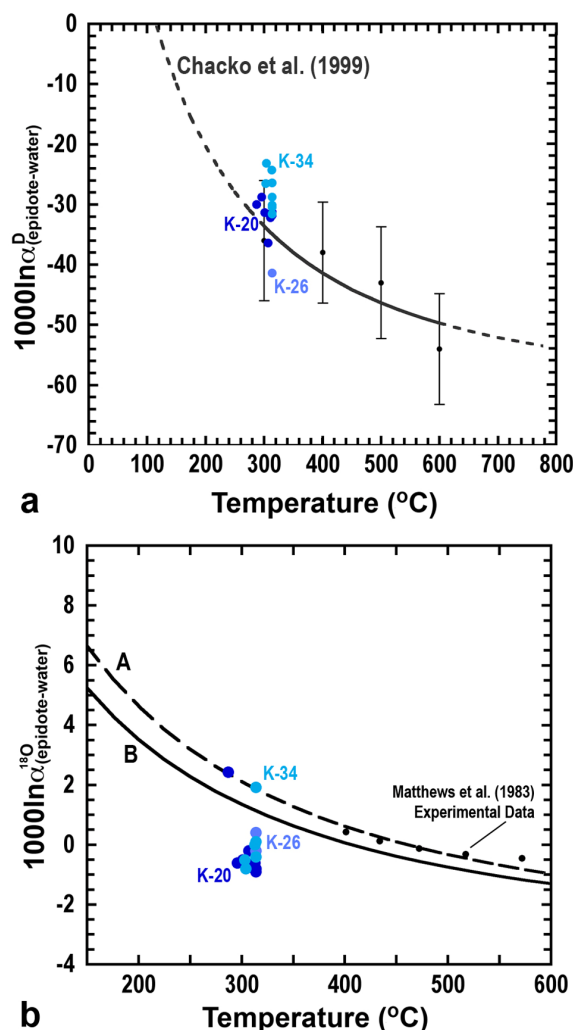
**Figure 5:** Closed system model of the hydrogen and oxygen isotope evolution of alteration minerals and geothermal fluid in Reykjanes. Initial epidote, chlorite and smectite isotope values (filled green circles) are derived from glacially-derived fluids (filled blue circle), and are calculated based on published fractionation curves. Black squares are measured Reykjanes geothermal fluids and green squares are geothermal epidote. The line between seawater (modern initial fluid, open black circle) and the weighted average isotope value of the rock (initial rock composition, filled black circle) represents isotopic exchange between these two endmembers. Blue open circles represent the resulting fluid composition ( $\delta D_{\text{FLUID}}$ ) when  $\delta D_{\text{ROCK}}$  is equal to modern epidote values. The arrows on the exchange line denote the extent of exchange that has occurred in both the initial alteration mineral composition and the initial geothermal fluid composition. Adapted from Pope et al. (2009)

## 4.2 Krafla Geothermal system

Preliminary epidote hydrogen and oxygen isotope analysis was concentrated in three wells in Krafla; two in Leirbotnar (K-26 and -34), and one in Sudurhlidar (K-20), near the primary upflow zone of the entire system. Data is shown as a function of depth in Figure 6. Hydrogen isotope values are between -116 and -123‰ in well K-20, -108 and -115‰ in well K-34, and two analyses in well K-26 both have a  $\delta D_{\text{EPIDOTE}}$  value of -125‰. Oxygen isotope compositions of epidote in these wells are -9.7 to -13.0‰ (K-20), -9.6 to -12.3‰ (K-34) and -11.1 to -11.7‰ (K-26). Although the  $\delta^{18}\text{O}$  value of hydrothermal epidote does not vary significantly between wells,  $\delta D$  values have a range greater than 15‰. The intermediate values of this range are from well KJ-20, located near Hveragil, along which the primary upflow zone of the system occurs. Fractionation between measured epidote and published geothermal fluid compositions are plotted relative to hydrogen and oxygen equilibrium fractionation curves in Figure 7. The hydrogen and oxygen isotope compositions of the fluids were averaged values of measurements taken at or nearby the wells in which we analyzed the epidote (from Sveinbjörnsdóttir et al., 1986; Darling and Ármannsson, 1989). Thus the values plotted in Figure 7 represent fractionation between hydrothermal epidote and fluids within the immediate vicinity. It appears that well K-20 has epidote with a hydrogen isotope composition in equilibrium with local fluids ( $\delta D \sim -90\text{‰}$ ; Figure 7a) within the error the Chacko et al. (1999) fractionation curve. Of the two Leirbotnar wells, epidote in well K-26 is slightly too high to be in equilibrium with local fluids ( $\delta D \sim -88\text{‰}$ ), whereas epidote in well K-34 is considerably too low. The oxygen isotope composition of Krafla epidotes indicates that with the exception of two outliers they are not in equilibrium with hydrothermal fluids in any wells (Figure 7b). Darling and Ármannsson (1989) noted a strong heterogeneity in  $\delta^{18}\text{O}$  of Krafla fluids, and attributed it to varying degrees of exchange between fluids and the hostrock of the system. It is possible that the disequilibrium observed in Krafla epidotes is reflecting this process.



**Figure 6:** Hydrogen and oxygen isotope composition of Krafla geothermal epidote for wells K-20, K-26 and K-34 as a function of depth. Published whole rock  $\delta^{18}\text{O}$  values for well K-7 by Sveinbjörnsdóttir et al. (1986)



**Figure 7: a. Hydrogen isotope fractionation between Krafla geothermal fluid and geothermal epidote from wells K-20, K-26 and K-34 as a function of temperature, compared to temperature-fractionation curve by Chacko et al. (1999). b. Oxygen isotope fractionation between epidote and Krafla geothermal fluid. Compared with temperature-isotope fractionation data by Matthews et al. (1983) and curves A (Zoisite-Water from Matthews et al., 1983; Matsuhisa et al., 1979) and B (Epidote-Water from Matthews et al., 1983, Matsuhisa et al., 1979; Kohn and Valley, 1998)**

As a whole, the observed variations in stable isotope properties of epidote from Krafla likely reflect the hydrologic features that characterize the geothermal system. This includes multiple hydrothermal fluid sources, mixing between upper and lower aquifers and between northern and southern geothermal fields, the likely input of magmatic fluids, and the effects of boiling (Arnórsson, 1995; Sveinbjörnsdóttir et al., 1986; Darling and Ármannsson, 1989). Further analysis of alteration minerals in several wells throughout the Krafla system will improve our understanding of how these factors influence the evolution of hydrothermal fluids. This is because there is evidence that in spite of the extensive alteration of the Krafla host-rock that is shown by the low whole rock  $\delta^{18}\text{O}$  data of Sveinbjörnsdóttir et al. (1986) in Figure 6, there seems to be little isotopic alteration of the hydrothermal fluids due to interaction with the rock. This is shown in Figure 2b, where the Krafla fluids are not enriched in  $\delta^{18}\text{O}$

relative to local meteoric water, suggesting that the water in this system has reacted very little with the surrounding hostrock. Hence, the stable isotope composition of alteration minerals in this system should only reflect the composition of modern geothermal fluids.

## CONCLUSIONS

The Reykjanes and Krafla geothermal systems represent two end members of Icelandic high-temperature geothermal systems. The Reykjanes geothermal system is a seawater-dominated system that has been active since the Pleistocene, and has undergone extensive hydrothermal alteration; enough so that modern fluids are chemically modified through diffusional exchange with relict hydrous alteration minerals. In contrast, the Krafla geothermal system occurs in a region of high volcanic activity, where magmatic fluids and local precipitation comprise the fluid source, and it displays a high water to rock ratio. Krafla is therefore an ideal location for studying how alteration minerals isotopically record hydrologic and geochemical heterogeneities in three dimensions within high-temperature geothermal systems.

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