

Fluid and metal origins at Theistareykir Geothermal Field, Iceland

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ABSTRACT

The ultimate source of metals in magmatic-hydrothermal systems is a matter of debate. Volatile trace metals and noble gases are here used as a potential way of tracing sources of metals in geothermal systems and differentiating between a magmatic source and derivation from the leaching of country rocks. We are currently studying the hydrothermal system of Theistareykir (Iceland), set on the mid-Atlantic oceanic ridge. A new geothermal field has been developed with comprehensive access to geological materials and fluids, thanks to 18 geothermal wells drilled and numerous fumaroles and mud volcanoes in the surrounding areas. Volatile trace metals have been analyzed in rocks and fluids from the deep wells, surface fumaroles and mud pots. The current objective is to use noble gases to constrain the geothermal reservoir's behavior. The second objective is to compare volatile trace elements from the deep geothermal wells with noble gases from the same wells. Since noble gases are highly volatile, we expect to see some trends between the two families of elements. The first results from noble gases show that the magma beneath Theistareykir has a dominant influence of the mantle, as sampled at the mid-ocean ridges, and a lesser influence of the mantle plume. The $^3\text{He}/^4\text{He}$ mean ratio is 11.45Ra (with Ra the atmospheric ratio of 1.384×10^{-6}), when a typical MORB is $8 \pm 1\text{Ra}$. Finally, preliminary results show that volatile metals can be linked to their source using noble gases. As an example, thallium shows a positive correlation with ^3He , which is an indicator of magma degassing zones in the Theistareykir area.

1. INTRODUCTION

The respective contribution of magma degassing and rock hydrothermal weathering to the metal content of an active magmatic-hydrothermal system is still debated. Fluids released by magma are relatively enriched in metals compared to their host magma (Hedenquist et al., 1993), sequestering the metals and injecting them into the hydrothermal system. However, it is unclear if sufficient mass of metals can be liberated in this process to generate an ore deposit (Yang and Scott, 2006). On the other hand, mass balance between altered rocks and their fresh counterparts commonly shows extensive metal remobilization, which has been related to the elevated temperatures of the hydrothermal fluids and their high ligand concentrations (Williams-Jones and Migdisov, 2014, Seward et al., 2014). Rock leaching thus appears as an efficient mechanism for reconcentrating metals in the hydrothermal fluids. The ligands are dominantly derived from magma degassing, and proponents of host-rock leaching as the dominant metal source therefore still have to invoke a degassing contribution (Hedenquist and Lowenstern, 1994).

To tackle this question, researchers have used a large spectrum of methods from thermal modelling, to O and H isotopes (Craig, 1963; Giggenbach, 1992), metal isotopes, volcanic gas compositions (Nadeau et al., 2016), fluids inclusion (Samson et al., 2008), laboratory experiments and geochemical modelling (Williams-Jones and Heinrich, 2005), to noble gases (Burnard and Polya, 2004).

Noble gases, and especially helium, have long been studied both in active magmatic-hydrothermal systems to decipher the origin of fluids and melts involved. Noble gases are potential tracers for constraining the metal content of ore forming-fluids by defining the contributions of purely magmatic fluids versus fluids impacted by leaching of host rocks (crustal fluids) using the helium isotopic ratio $^3\text{He}/^4\text{He}$ or the argon isotopic ratio $^{40}\text{Ar}/^{36}\text{Ar}$. Noble gases elemental ratios Ne/Ar , Kr/Ar and Xe/Ar are very helpful to constrain solubility-driven processes such as boiling and steam condensation which are crucial processes in understanding magmatic-hydrothermal systems' dynamic (Fisher, 1997; Burnard, 2001).

In this study, we focus on the origin of fluids in the magmatic-hydrothermal system of Theistareykir using noble gases as tracers and briefly showing their potential in tracing metal paths in the reservoir.

2. STUDY AREA

2.1 Geological setting and surface emissions

Theistareykir geothermal field is located in the northernmost segment of the NVZ and it is the westernmost of the five *en echelon* volcanic systems of the NVZ. The high temperature geothermal activity is connected to recent magma intrusions, with most recent volcanic activity occurring some 3000-2500 years ago (MacLennan et al., 2002, Óskarsson et al., 2013). At the surface, four lava units occur in the area, most related to the activity of the Theistareykjarbunga shield volcano: Skildingahraun (>14.5 ka) lava shield; Stórávíti, a post-glacial lava shield of 30 km^3 erupted ca. 10.5 ka ago (MacLennan et al., 2002); Borgarhraun (10-8 ka) formed of picritic basalt and the younger Theistareykjahraun lava shield (2400 yrs) (Saemundsson, 2007) (Fig. 1).

Tectonically, Theistareykir developed at the intersection of the N-S oriented Northern Rift Zone (corresponding also to the NVZ) and the WNW-ESE-oriented transform zone called the Tjörnes Fracture Zone (Khodayar et al., 2018) (Fig. 1). The Theistareykir fissure swarm is 7-8 km wide and 70-80 km long (Saemundsson, 2007). Until recently, it was assumed that only N-S structures created permeability in the reservoir, but relation between deep-seated gas manifestations and tectonic feature suggested that the main WNW, ENE, NW and N-S fault segments create permeability (Khodayar et al., 2018).

The geothermal area of the Theistareykir is 25-30 km² wide, but surface manifestations are confined to the eastern part of the fissure swarm, northwest and north of Mt. Bæjarfjall, with a total surface of 11 km² (Óskarsson et al., 2015). The geothermal manifestations comprise warm soil, mud-pools, fumaroles and solfataras as well as steam areas. Mud pots are exposing a wide range of colors related to hydrothermal clay alteration (white, grey, blueish, reddish, greenish) and are bubbling for the most part. The temperatures of the fumaroles are all around 99°C, but reconstructed gas temperatures at depth are between 257 and 310°C (Óskarsson et al., 2015). The Theistareykir thermal area has been historically divided into five N-S oriented sub-areas from East to West (Theistareykjahraun (E), Tjarnarás (D), Theistareykjagundur (C), Bóndhólsskard (B) and Ketilfjall (A)) on the basis the estimated reservoir temperatures by gas geothermometry, with results ranging from 230 to 315°C (Grönvold and Karlsdóttir, 1975, Gíslason et al. 1984, Ármannsson et al., 1986). From these five sub-areas, Tjarnarás (D), Theistareykjagundur (C) and Ketilfjall (A) seemed to be the most promising for drilling.

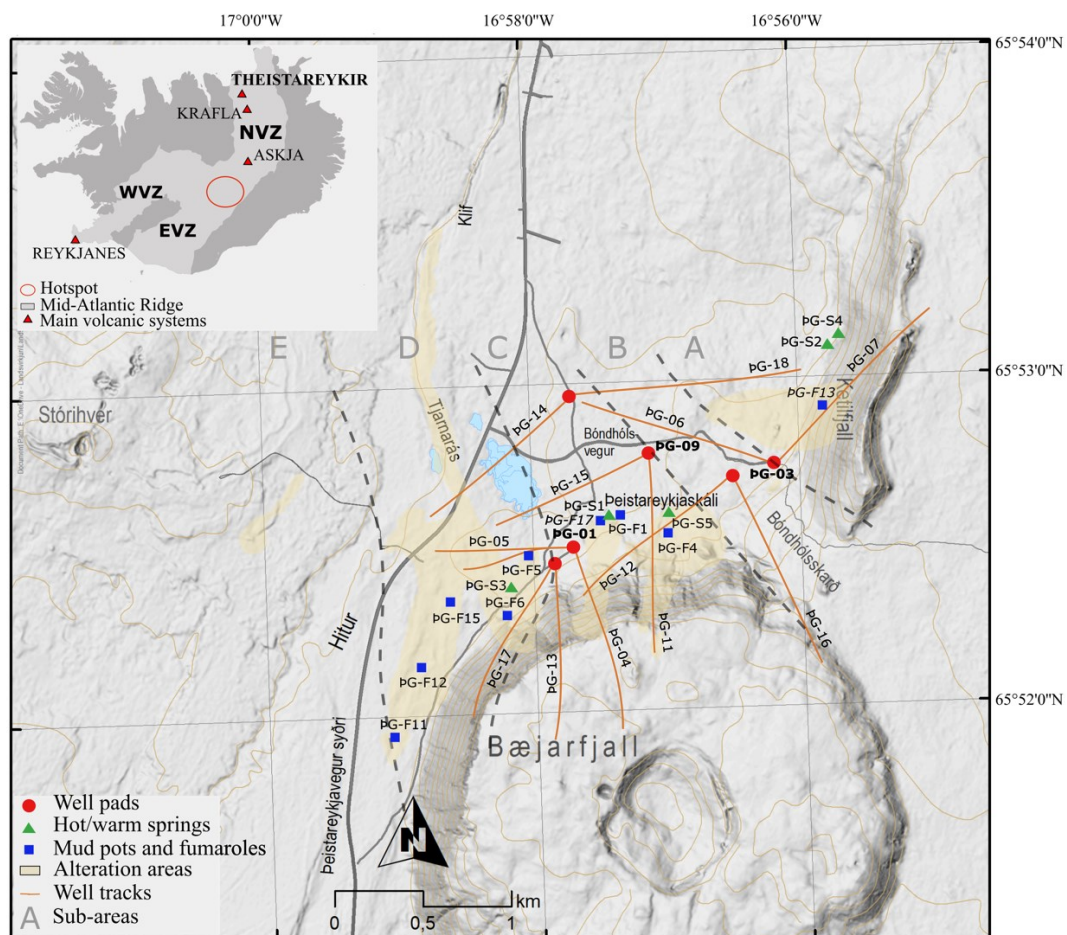


Figure 1: Simplified map of the Theistareykir geothermal field with major alteration zones and positions of the sampled sites (wells, mud pots, and fumaroles) indicated. Numbers on the X- and Y-axes indicate the latitudinal and longitudinal geographical coordinates respectively on the World Geodetic System 1984 (WGS84). NVZ = Northern Volcanic Zone; WVZ = Western Volcanic Zone; EVZ = Eastern Volcanic Zone.

The heat source of the field is related to the most recent activity in the area and would be supply by an E-W-oriented magmatic intrusion (Óskarsson et al., 2015). The meteoric recharge of the Theistareykir geothermal system was long thought to be controlled by a modern water flow coming from the highlands around the area (Ármannsson et al. 1986; Darling et al., 1989). But recent studies based on the stable isotope composition of water suggest that the recharge of the system is older, possibly recharged during a colder climate than the actual (Sveinbjörnsdóttir et al., 2013, 2015).

The Theistareykir geothermal field started its development with a first set of nine wells drilled between 2002 and 2011 at depths ranging between 1723 m to 2799 m meters b.s.l. At the moment, a total 18 geothermal wells have been drilled. Geothermometers indicate reservoir temperatures between 270 and 300°C. The capacity of the power plant started with a unit of 45MW (Gudmundsson et al., 2008) and recently a second unit has been added to bring the total outcome power to 90 MW.

3. SAMPLING AND ANALYTICAL METHODS

Samples of geothermal fluids were collected during summers 2017, 2018 and 2019. Among the surface manifestations, a total of eleven mud pots and two fumaroles (named ÞG-FX) were sampled for noble gases; five warm springs (named ÞG-SX) were collected for major ions and trace elements abundances and for water isotopes ($\delta^{18}\text{O}$, $\delta^2\text{H}$). A total of nine geothermal wells (named ÞG-X) were sampled for noble gas isotopes, water stable isotopes and for major ions and trace element abundances.

The water phase was collected from boiling hot springs. Mud pots were not sampled for water, but only for gases because the mud is extremely dense, and filtration is impossible on site. At the geothermal well site, the water and gas phases were collected using a portable steam/fluid separator installed just at the exit of the wellhead, being the field device of Webre separators. Collected water was poured in a 30 ml HDPE bottle filled to the top and closed avoiding air bubbles for the analysis of water isotopes. The gas phase was collected using a standard refrigeration-grade 3/8" (14 cm³ internal volume) copper tube. The copper tube was directly installed at the gas exit of the portable fluid/steam separator using all metal NPT connections. After letting the gas flow for several minutes, the tubes were sealed using stainless steel pinch-off clamps closed using electric drill to minimize air contamination (Pinti et al., 2017). Gases were collected from the mud pots using an armed-PVC tubing connected at one end to a plastic funnel. The funnel was placed directly above the bubbling mud pot with the mud acting as a seal to prevent air contamination. At the other end of the PVC tubing, the tube itself was placed in a bottle filled with water to check for gas flow and to avoid air contamination. Water has also been filtered and collected in 30 ml HDPE acid pre-washed bottles with addition of 0.5% HNO_3 .

Water isotopes were analyzed at the GEOTOP facilities at the University of Québec in Montréal by Isotope Ratio Infrared Spectrometer with two lasers for the simultaneous measurements of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ by Off-Axis Integrated Cavity Output Spectroscopy. The uncertainties on the measurements are better than 0.07‰ for the measured $^{18}\text{O}/^{16}\text{O}$ ratio and 0.28‰ for the measured $^2\text{H}/^1\text{H}$ ratio. The total analytical uncertainty (1σ) is better than $\pm 0.1\text{‰}$ for $\delta^{18}\text{O}$, $\pm 1.0\text{‰}$ for $\delta^2\text{H}$, including that of the standards. Noble gas analyses of fumarole, mud pots and wells were carried out at the Noble Gas Laboratory of the University of Michigan, except for well ÞG-17 and mud pot replicates ÞG-F12 and 15 which noble gas analyses were carried out at the Noble Gas Laboratory of the GEOTOP. At the University of Michigan gas samples – connected to a stainless-steel purification line – were expanded in a known volume and gas pressure reduced by computer-controlled sequential pumping until reaching a value acceptable for analyses. The reduced gas sample was successively dried on a molecular sieve trap and active gases are removed using three Ti getters at 600°C for three minutes each. The He, Ne, Ar, Kr, and Xe were quantitatively extracted using a computer-controlled cryo-separator at temperatures of 49 K, 84 K, 210 K, 245 K, and 290 K respectively, and sequentially allowed to enter a Thermo Scientific® Helix SFT mass spectrometer for He and Ne isotope analyses, and a Thermo Scientific® ARGUS VI mass spectrometer for the Ar, Kr, and Xe isotope analyses. All noble gas isotopes were measured using a Faraday detector, except for ^3He , which was measured using an electron multiplier in ion counting mode. Prior of each analysis, a blank run is done following the same procedure as the sample. Typical blanks are 0.04 to 0.15% of the measured sample value, respectively. Quantitative analyses were obtained by calibrating the two mass spectrometers with a known aliquot of standard air. Typical standard reproducibility for ^4He , ^{20}Ne and ^{36}Ar are 0.4–0.5% while for $^{20}\text{Ne}/^{22}\text{Ne}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios the reproducibility is 0.07% for both. Calculated standard errors for concentrations range from 1.3 to 2.2% of the measured values.

Trace metals and chemo-physical parameters from the well's fluids have been analyzed at GEOTOP at McGill University by ICP-MS and AAS as well as by the Krafla Laboratory in Iceland. At McGill University, the majority of trace metals have been analyzed by a Thermo Scientific iCAP Q Inductively Coupled Plasma Mass Spectrometer (ICP-MS). Arsenic has been analyzed using a Perkin Elmer AAnalyst 800 atomic absorption spectrometer.

4. ISOTOPIC SIGNATURES OF FLUIDS

4.1 Water isotopes

Water stable isotopes $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values measured in geothermal wells directly at the steam/liquid separator ranges from -12.85 ‰ to -8.62 ‰ and from -127.01 ‰ to -105.18 ‰, respectively. The stable isotopic water composition has been measured in the residual water phase after separation at the well-head. This composition needs to be recalculated for the so-called “total discharge” (TD), i.e., the composition of the water+steam mixture at the reservoir conditions (Arnórsson and Stéfansson, 2005).

After correction, the “total discharge” $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values ranges between -13.58 ‰ to -9.02 ‰ and from -127.36 ‰ to -106.85 ‰, respectively. The $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values measured in fumaroles and mud pots by ÍSOR (Óskarsson, 2019) range from -19.61 ‰ to -12.16 ‰ and from -123.8 ‰ to -108.2 ‰, respectively (Fig. 2). Based on these results and previous studies (Sveinbjörnsdóttir et al. 2015) of the same field, the general Theistareykir water isotopic composition show fluids that are particularly depleted both in ^{18}O and ^2H than the other volcanic areas in Iceland and compared to local precipitation. Previous studies also showed distinct differences for the subfields within the Theistareykir geothermal field, suggesting different fluid origins and/or paths (Sveinbjörnsdóttir et al. 2015).

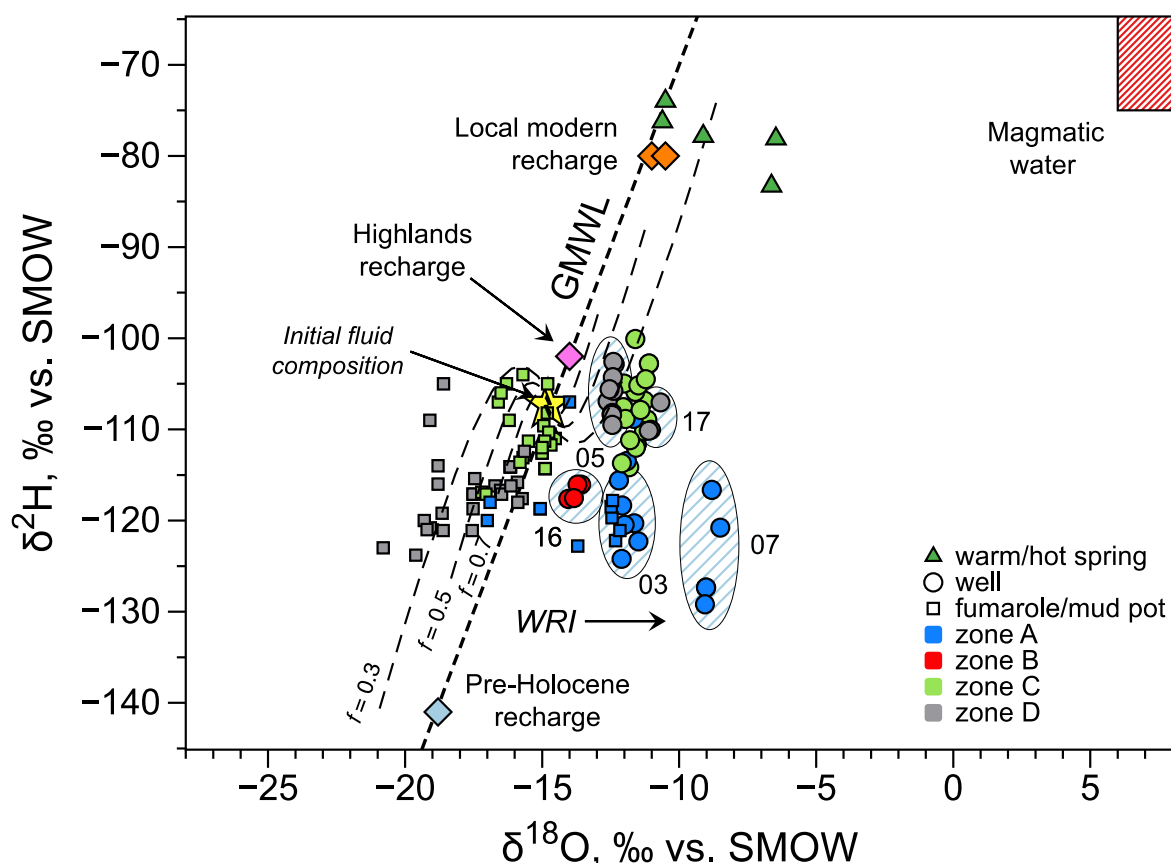


Figure 2: Plot of $\delta^2\text{H}$ vs. $\delta^{18}\text{O}$ corrected for total discharge (TD) of the Theistareykir production fluids, mud pots, and fumaroles sampled over a 20-year period (data from this study and archived ÍSOR data). The global meteoric water line (GMWL; dashed line) was calculated following Craig (1961). Pre-Holocene recharge composition is from data of Sveinbjörnsdóttir et al. (2013), and the regional highlands recharge is from Stéfansson et al. (2017). Primary magmatic water composition is from Sheppard and Epstein (1970). The dashed curves represent the evolution of $\delta^2\text{H}_{\text{TD}}$ vs. $\delta^{18}\text{O}_{\text{TD}}$ in the vapor and residual liquid after phase segregation, starting from an initial composition (yellow star), and causing isotopic fractionation. Water isotopic compositions thus evolve along these lines according to their final equilibrium temperature. The f values correspond to the fraction of residual liquid. Blue dashed areas group stable isotopic compositions measured over the years for wells 03, 05, 07, 16, and 17. Zone A: Ketilfjall, Zone B: Bóndhóllsskard, Zone C: Theistareykjagrundir, Zone D: Tjarnarás.

The fluids of Theistareykir are mainly composed of meteoric water with a little portion of magmatic fluids degassing from the magmatic intrusion (Gudmundsson, 2008) as also indicated by helium isotopes (see next section). The origin of water seemed first non-local as suggested by the depleted ^2H isotope values but was then interpreted as pointing out the presence of past precipitated water under colder climate, probably from the last glaciation (Sveinbjörnsdóttir et al., 2013), since the value of -127‰ was way under the most depleted values found in Iceland in the present. ^{18}O values seem to reflect a high rock leaching heated water circulating in the area with an oxygen shift up to 6.5‰ (Óskarsson et al., 2015).

4.2 Helium isotopes

The measured $^3\text{He}/^4\text{He}$ ratio is reported as “R” and normalized to that of the atmosphere ($R_a = 1.384 \times 10^{-6}$). Air-normalized helium isotopic ratios, R/R_a , calculated from the wells range from 9.43 ± 0.2 for well ÞG-13 to 11.44 ± 0.30 for well ÞG-07. The same results for the fumaroles range from 1.78 ± 0.01 for fumarole ThG-F11 to 10.41 ± 0.09 for fumarole ÞG-F13. These values indicate a clear mantle origin for helium (Fig. 3). Figure 3 shows the $^3\text{He}/^4\text{He}$ ratios measured in fumaroles and geothermal wells against the $^4\text{He}/^{20}\text{Ne}$ ratio at Theistareykir.

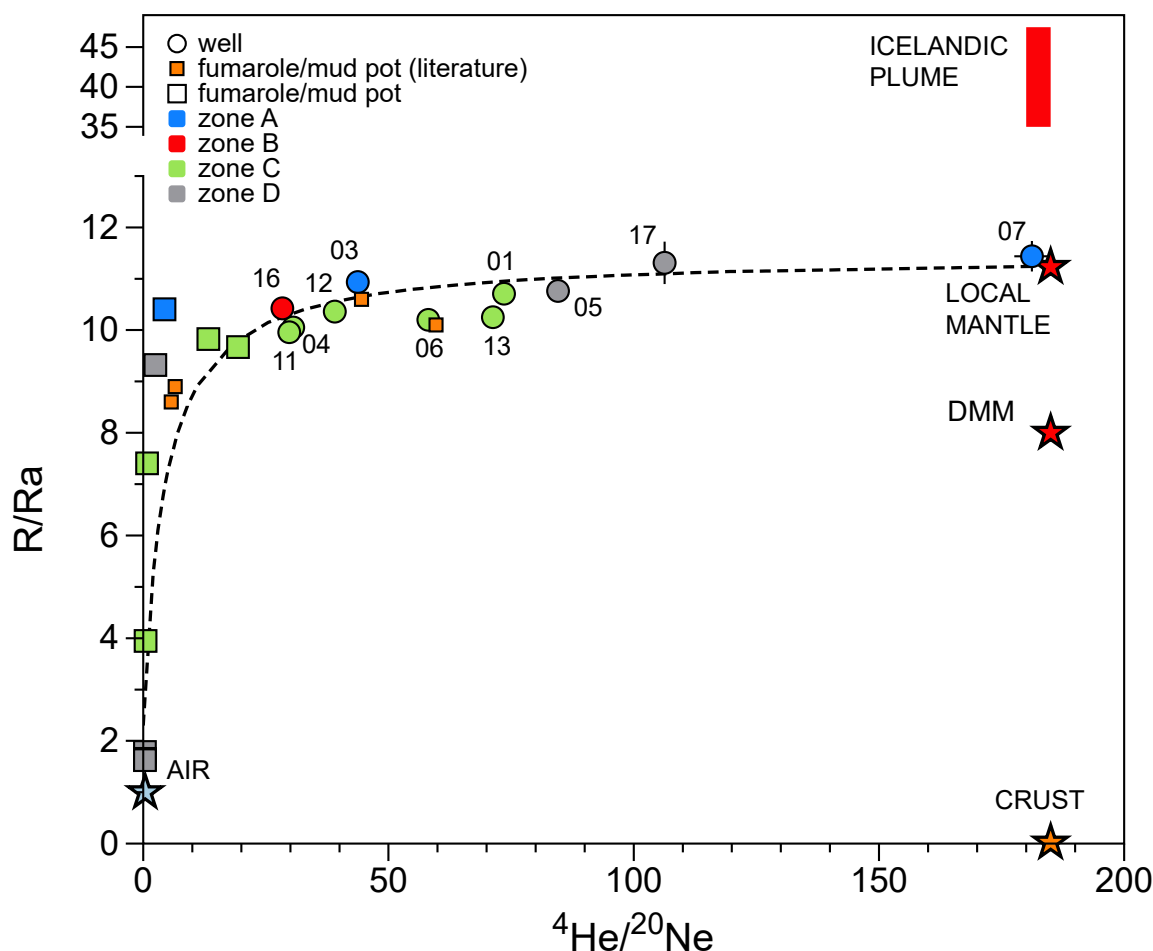


Figure 3: The measured $^3\text{He}/^4\text{He}$ ratio (R), normalized to that of the atmosphere (Ra), versus $^4\text{He}/^{20}\text{Ne}$ ratios. Extrapolated mixing hyperbola obtained by data inversion suggests mixing between an atmospheric helium component (ASW or air) and a local mantle helium component with a R/Ra of 11.45, resulting from the mixing of a depleted MORB-type mantle (DMM) and a plume-type mantle. The values from literature are from Hilton et al., (1990), Poreda et al., (1992), and Füri et al. (2010).

In this figure, the endmember labelled MORB represents the convective mantle with R/Ra of 8 ± 1 (Allègre et al., 1995), while a maximum R/Ra of 42.9 – as measured in alkaline lavas of Iceland by Breddam and Kurz (2000) – is assumed representing the Icelandic hotspot, which is presently centered ca. 250 km south of Theistareykir (Ritsema and Allen, 2003). Labelled CRUST endmember represents the $^3\text{He}/^4\text{He}$ ratio in crustal-derived fluids (R/Ra of 0.02). Finally, the AIR labelled endmember represents local meteoric water that has dissolved atmospheric noble gases (ASW or Air Saturated Water). Samples from Theistareykir show a mixing between the local mantle and the air, as obtained by an inverse regression hyperbola (Albarède et al., 1995; Pinti et al., 2019). The local mantle (MANTLE label in Fig. 3) has a R/Ra value of 11.45 which can be assumed to be the mixing of 8.7 to 12.7% Icelandic plume helium (R/Ra of 35–47.5; Breddam and Kurz, 2001; Harðardóttir et al., 2018) with 87.2–91.3% of helium from the mid-ocean ridge.

4.3 Volatile trace metals

All trace metals have been analyzed but these results of this study are focused on the most volatile ones: arsenic (As), cadmium (Cd), thallium (Tl), mercury (Hg), antimony (Sb) and bismuth (Bi). They have been detected in the fluids collected from the wells but their concentrations are very low ranging from 0.93 to 25.11 ppb for As, 0.0014 to 0.0072 ppb for Cd, 0.031 to 0.088 ppb for Tl, 0.1051 to 0.3022 ppb for Hg, 0.0013 to 1.033 ppb for Sb and 0 to 0.036 ppb for Bi.

5. FLUID ORIGINS, RESEVOIR DYNAMIC AND VOLATILE METALS' BEHAVIOUR

The origin of water at the Theistareykir geothermal field is globally quite homogeneous despite small variations within the field (Sveinbjörnsdóttir et al. 2015). Meteoric water seems to be the dominant component of the fluids (Fig. 2; Gudmundsson, 2008), mixed with a small proportion of fluids enriched in deep volatiles from the magmatic source underneath the area, as shown by helium isotopes (Fig. 3). Regarding the dynamic of the reservoir, the results indicate that for now, the field is not affected by either intense boiling induced by the production of the wells nor by condensation of fluids coming from the injectate. In that regard, we can consider that the reservoir is not influenced by the human production and can be studied as a natural system to understand sources and behaviors of fluids and trace metals.

The shift in ^{18}O from the water isotopic characterization show that the rocks composing the reservoir are undertaking some leaching (Fig. 5), enriching the water with the heavier oxygen isotope of mass 18 (Giggenbach, 1992). This conclusion seems to be supported by the decrease of the R/Ra. This decrease is due to addition of radiogenic ^4He , the decay product of ^{238}U , ^{235}U , and ^{232}Th contained in rocks. The produced ^4He is transferred from rocks into groundwater mainly by diffusion and α -recoil (Torgersen et al., 1985). Then, the longer the reservoir rock is in contact with the geothermal fluids, the more it is enriched in radiogenic ^4He . This process is first enhanced due to the heating intrusion but also by considering that the water (pre-Holocene; Sveinbjörnsdóttir et al. 2015) has probably already undertaken interaction with rocks for a long period of time. On another hand, at least two wells (pG-07 and pG-17) seem to reflect a magma degassing process with an enrichment in both R/Ra and ^{18}O , showing that both rock leaching and magma degassing are definitely occurring in the area.

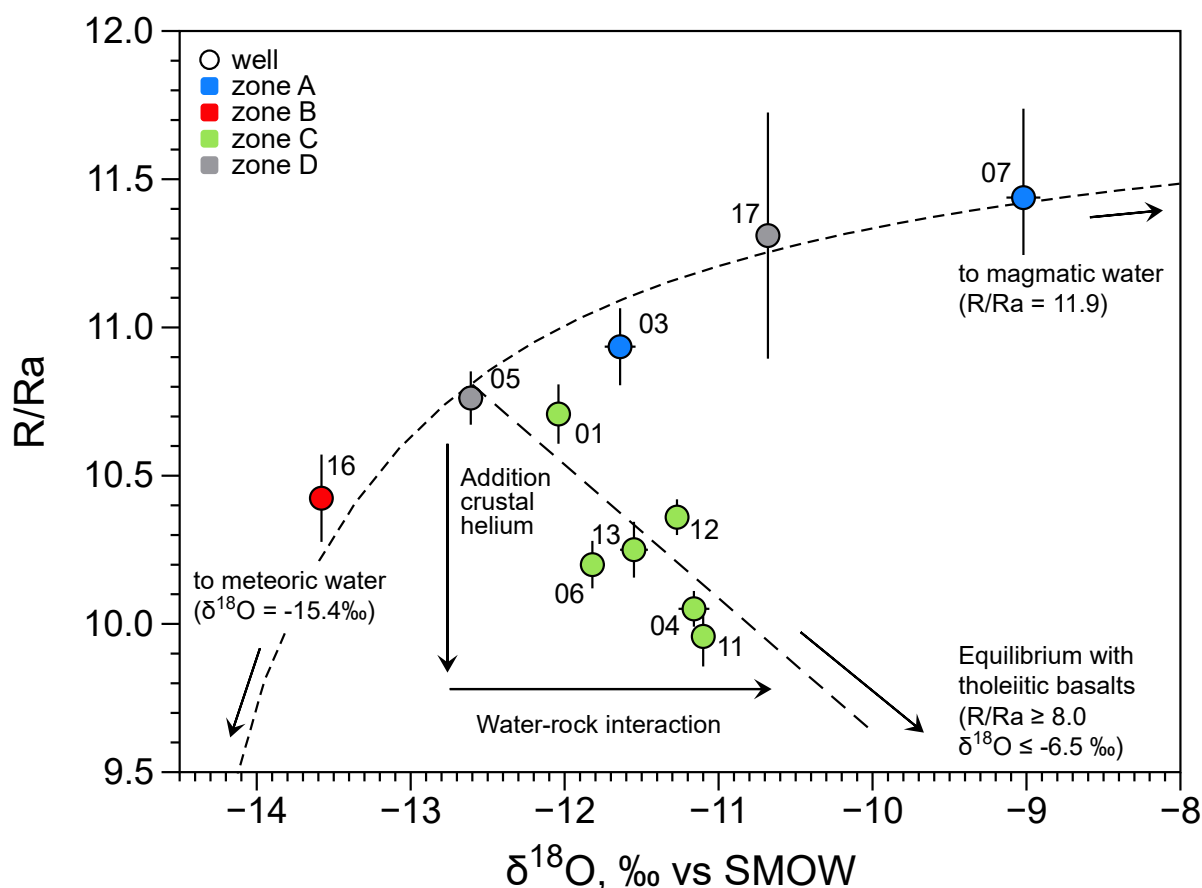


Figure 5: Helium isotopic composition (R/Ra) vs. the $\delta^{18}\text{O}$ values of Theistareykir well fluids. Dashed lines show mixing between meteoric fluid (a mixture of local recharge, more distant regional recharge, and pre-Holocene recharge with a mean $\delta^{18}\text{O}$ of -15.4 ‰) and magmatic fluid (with a theoretical initial composition of R/Ra > 11.9 and $\delta^{18}\text{O} \approx +4.7$ ‰ (Macpherson et al., 2005), and the effect of water-rock interactions with the addition of radiogenic ^4He to fluids and heavier $\delta^{18}\text{O}$ (with R/Ra ≈ 8.0 and $\delta^{18}\text{O} \approx -6.5$ ‰) respectively.

One of the goals of this study is to try to decipher the origin of volatile metals in the geothermal field, knowing the sources of fluids and the dynamic of the system. The preliminary results from the study show that for example, the thallium source is directly correlated to ^3He (Fig. 6). This would suggest that the source of Tl in the reservoir's fluids originates mainly from the degassing of the underlying intrusion.

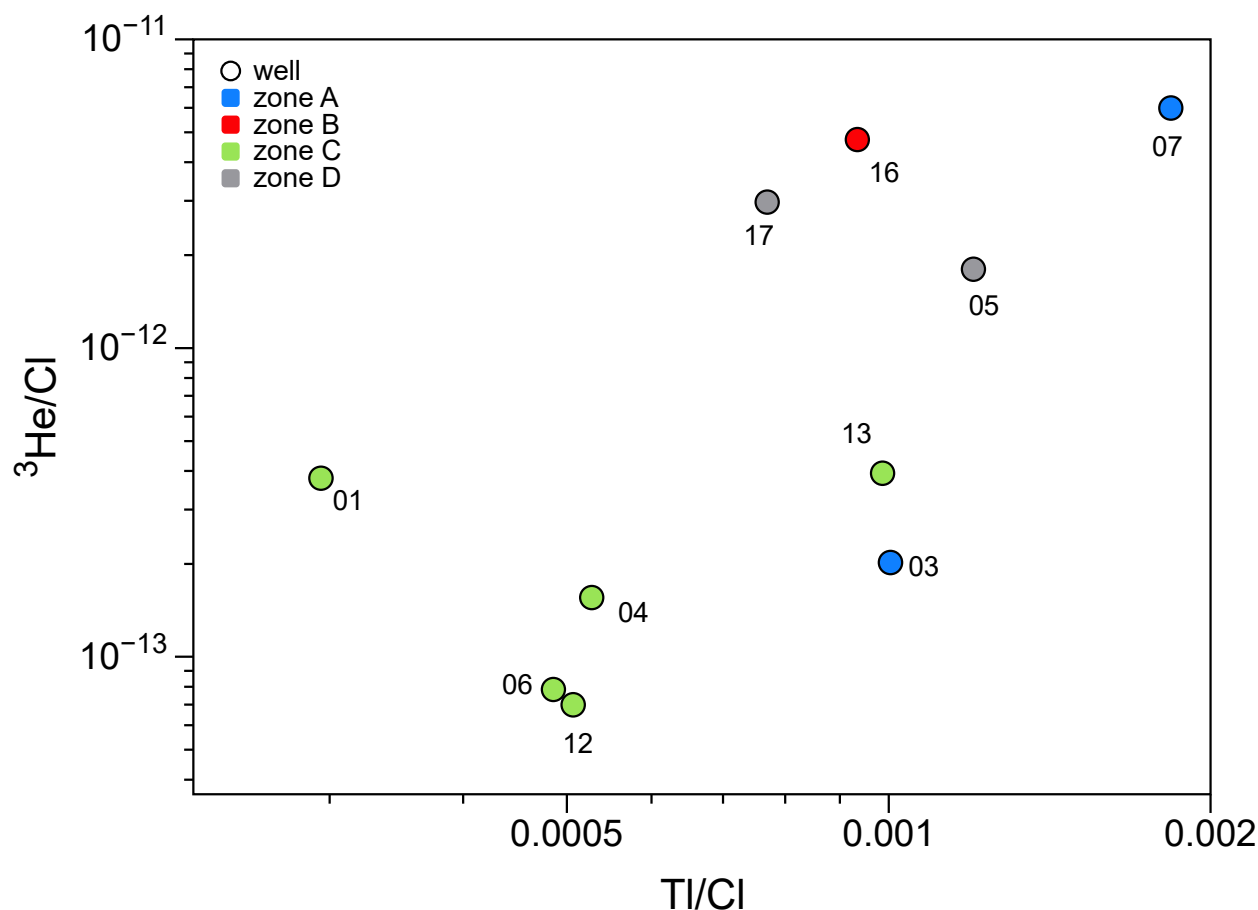


Figure 6: Isotopic values of helium ($^3\text{He}/\text{Cl}$ ratio) against thallium (Tl/Cl) concentration in the deep fluids from the wells discharge.

Then characterizing the source of metals in the system becomes critical to quantify the proportions of each source in the total balance. To do so, experiments are currently carried out to evaluate the amount of volatile metals released by the intrusion fluids given the initial composition of the magma. On the other hand, a sampling campaign is also done to sample the deep fluids of the geothermal system to evaluate the composition of the fluids in equilibrium with the surrounding rocks, before the boiling zone. This information will help us quantify the amount of metals released by the rocks during interaction with the geothermal fluids.

6. CONCLUSIONS

The great range of samples available in the study area as well as the access to the samples make this study unique to understand elements' behaviors to a smaller scale and with more precision. This work will be of primary interest to better constrain and understand trace metals and noble gases' behavior in active magmatic-hydrothermal systems. These preliminary results clearly show the input of both magma degassing and rock leaching to the total composition of the reservoir fluids. The study of noble gases and volatile metals thus seems promising in quantifying these inputs.

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