# Monitoring Fumarole Gas Chemistry in the Theistareykir Geothermal FIeld, NE Iceland

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#### **ABSTRACT**

The chemical and isotopic composition of fumarole steam is reported for the Theistareykir geothermal field in NE Iceland. The samples were collected annually from selected fumaroles in the years 2012 to 2018 as part of a baseline monitoring of surface activity. The first 45 MWe unit of the Theistareykir geothermal power plant was commissioned in December 2017, and the second 45 MWe unit came online in April 2018. In addition to annual samples for analyses of major gas components and stable water isotope analyses, samples were collected for the determination of stable gas isotopes (He, Ne, Ar, C, N) in 2014.

The gas composition suggests considerable variability within the field, both in terms of estimated reservoir temperatures as determined by gas geothermometry and fluid origin as determined from stable hydrogen and oxygen isotopes. Estimated temperatures range from 250 to 320°C. The Theistareykir fluids are much more depleted in deuterium than local groundwater. The gas isotopes suggest superficial origin of  $N_2$  and Ar, but  $\delta^{13}C$  is typical for the Icelandic mantle plume. The  ${}^3He/{}^4He$  ratio is higher in Theistareykir than in nearby Krafla and Námafjall – and somewhat higher than what is expected for MORB.

During the monitoring period, little changes have been observed in gas composition and overall surface activity of the Theistareykir field, although geothermal manifestations constantly form and disappear. The monitoring period covers a few years of drilling and well testing, but only half a year of production from the Theistareykir field and will provide a valuable base line for future monitoring of the field. Since production started from the field, the hydrogen and oxygen isotopes of steam from one fumarole have changed substantially, suggesting that the fumarole may now draw steam from a different aquifer. This is most likely a result of production-induced pressure changes in the reservoir.

## 1. INTRODUCTION

The Theistareykir geothermal field is situated in the Theistareykir fissure swarm, which is the westernmost of the five NNE striking left-stepping en échelon volcanic systems that constitute the Northern volcanic zone (NVZ) of Iceland (Figure 1, left). The high temperature geothermal activity is believed to be connected to recent magma intrusions. The most recent volcanic activity in the area occurred some 2500 years ago. Exploration drilling in Theistareykir started in 2002, and since then 19 exploration, appraisal and production wells have been drilled, resulting in the commissioning of the Theistareykir power plant in December 2017. The installed capacity of the power plant is 90 MWe.

As part of a baseline study for monitoring of the environmental effects of geothermal utilisation in the field, changes in surface activity and fumarole gas chemistry have been monitored annually. This includes visual inspection of surface activity, thermal imaging, soil gas flux measurements and monitoring of fumarole steam composition. This contribution will focus on the fumarole gas chemistry, including gas concentrations, stable isotopes of hydrogen and oxygen, which were determined annually from 2012 to 2018. Additionally, one set of samples for determination of stable gas isotopes (He, Ne, Ar, C in CO<sub>2</sub>, N in N<sub>2</sub>) was collected in 2014 and will be discussed here.

# 2. SAMPLE COLLECTION AND ANALYSIS

Samples of fumarole steam and steam condensate were collected annually from 2012 to 2018, in August each year. Sampling locations are shown in the right panel of Figure 1. Fumaroles were selected based on their location (trying to cover all the field), temperature (boiling) and flowrate (as high as possible). The samples were collected by inverting a plastic funnel over the fumarole and packing it with clay in order to reduce the probability of atmospheric contamination. The other end of the plastic funnel was connected to a 1" titanium tube fitted with silicone tubing on the other end. Samples for gas composition and gas isotope analysis were collected and condensed into evacuated double-port glass flasks (two flasks for each sample) containing 10 M NaOH, except sample fractions for the determination of  $\delta^{13}$ C, for which dry gas was collected into rubber-stoppered glass phials (vacutainers). Samples for analyses of Cl and stable water isotopes ( $\delta D$ ,  $\delta^{18}$ O) were condensed in a stainless steel cooling spiral and collected to 30 mL plastic phials. The gas concentrations in the samples were analysed by gas chromatography and titrimetry ÍSOR laboratories in Reykjavík, as were concentrations of Cl in condensed steam, which were analysed by ion chromatography. The stable water isotopes were analysed by mass spectrometry at the Institure of Earth Sciences, University of Iceland. The stable gas isotopes were determined by mass measured at the INGV laboratories in Naples and Palermo. The  $^{36}$ Ar, N<sub>2</sub> amount and the N<sub>2</sub> isotope composition was determined on the same aliquot of sample.

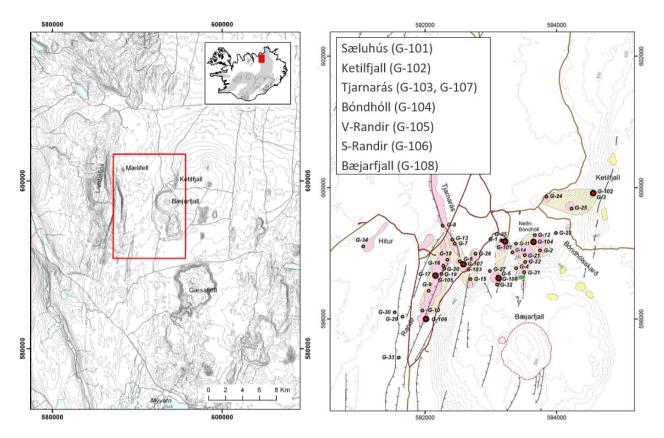


Figure 1: Location of the Theistareykir geothermal area (left). Location of sampled fumaroles in the Theistareykir area (right). The red dots indicate fumaroles sampled in 2012-2018, whereas approximate locations of previous sampling points are shown with yellow dots. Intense geothermal alteration is indicated by pink, slight alteration by yellow and steaming ground by brown areas.

## 3. DISCUSSION

## 3.1 Fumarole Gas

The measured concentrations of the main gas components are plotted in Figure 2 against time. Concentrations are given as mmol gas per kg steam. The <sup>2</sup>H (D) and <sup>18</sup>O content of steam is also shown in the figure. Older values are taken from Ármannsson (2004). The figure shows that although substantial changes in gas composition occurred in the 1990s, gas concentrations in 2012-2018 are generally similar to those measured during early exploration in the 1980s.

From the figure it is apparent that a few samples have been significantly affected by steam condensation, most likely in the subsurface. This is certainly the case for the sample collected from Ketilfjall in 2014 and also for the samples collected from V-randir (fumarole pG-105) in 2014 and 2015. That particular fumarole has since developed into a boiling mudpool and has not been sampled for the last three years. The absence of  $O_2$  and the generally low concentrations of  $N_2$  and Ar suggest that atmospheric contamination is not a severe problem during sampling, but increases in  $N_2$  and Ar concentrations parallel with increases in other gas concentrations again suggest condensation, e.g. in Tjarnarás in 2013.

The gas geothermometers calibrated by Arnórsson et al. (1998) were used to estimate the reservoir temperature for each sample. The average result for each location is given in Table 1. The averages are calculated based on the CO<sub>2</sub>, H<sub>2</sub>S, H<sub>2</sub>, CO<sub>2</sub>/N<sub>2</sub>, H<sub>2</sub>S/Ar and H<sub>2</sub>/Ar geothermometers using data from all years except when samples have been affected by condensation, as discussed above. Generally, the geothermometers based on different gases agree quite well, although some systematic discrepancies are observed. In particular the H<sub>2</sub> geothermometer tends to give unrealistically high values for samples from Ketilfjall and is therefore omitted.

Table 1. Average geothermometry results for each location, calculated using the geothermometers of Arnórsson et al. (1998).

Location	Tave (°C)
Ketilfjall	$307 \pm 15$
Bóndhóll	$300\pm19$
Sæluhús	$297\pm12$
Tjarnarás	$299 \pm 6$
Bæjarfjall	$288 \pm 20$
V-Randir	$255\pm1$
S-Randir	$282\pm2$

When geothermometry tempeartures are compared with the geographical distribution of the fumaroles, it is apparent that the highest temperatures (> 300°C) are observed in the easternmost part of the field (Ketilfjall), with temperatures around 300°C in the central field (Bóndhóll, Ketilfjall, Tjarnarás) and lower temperatures (< 290°C) in Bæjarfjall and Randir to the west.

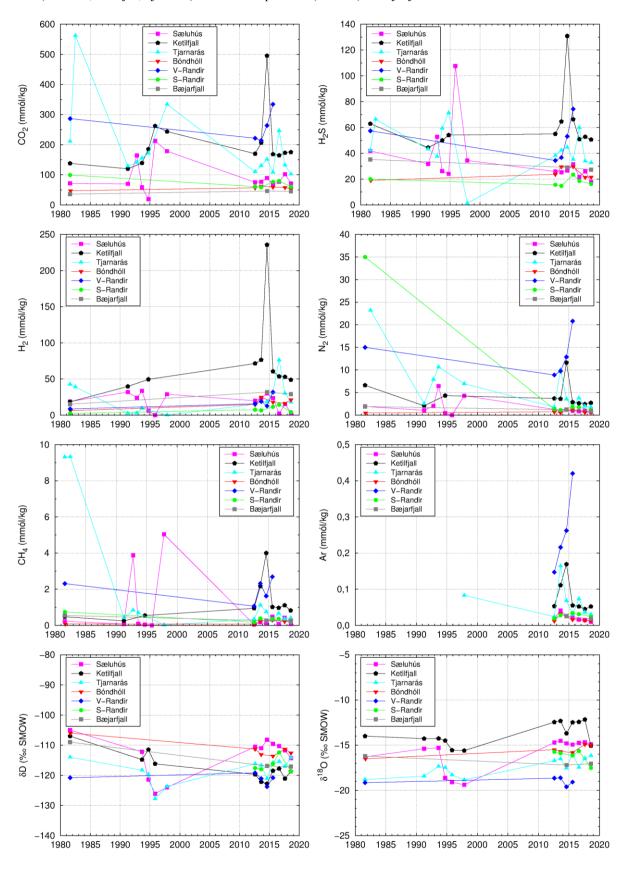


Figure 2: Concentration of major gases (mmol/kg) and stable isotope content of steam samples from fumaroles in Theistareykir, 2012-2018. Location of the sampling points is shown in Fig. 1 (right).

#### 3.2 Stable Water Isotopes

The stable isotope content of condensed steam is plotted in Figure 3, along with the global meteoric water line, local groundwater, the reservoir fluid of two production wells (PG-01 and PG-07) and isotope values of steam formed by depressurisation boiling of those fluids (red and green dotted lines), condensation lines for primary steam from PG-01 at 100°C and 200°C (black dashed lines) and steam formed by evaporation of heated groundwater at 100°C (blue dashed line). The boiling, evaporation and condensation lines are calculated using the equations of Horita and Wesolowski (1994).

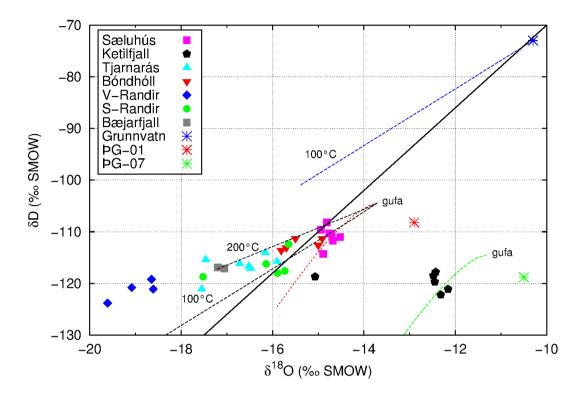


Figure 3: Stable water isotope values for the samples from Peistareykir, 2012-2018. Also plotted are the global meteoric water line (Craig, 1961) local groundwater and isotope values for the reservoir fluid of nearby production wells. The dotted lines show the evolution of steam formed by boiling of the reservoir fluid of each well from the reservoir temperature to 100°C. The dashed lines show condensation at 100°C and 200°C.

The deuterium values span the range -108 to -124‰, but some of the lowest values are from samples that have been affected by condensation, e.g. from V-Randir and Tjarnarás – samples that also show unusually low  $\delta^{18}O$ . The most enriched samples are from the centre of the field (Sæluhús and Bóndhóll) but the most depleted samples are from Ketilfjall. The Ketilfjall samples also show a much larger oxygen isotope shift than the other samples (about 4‰). The relationship between the different sample points suggests that the steam in Ketilfjall may be formed by the boiling of reservoir fluid from nearby well PG-07 – which has an isotope composition different from all other production wells in Peistareykir (Sveinbjörnsdóttir et al., 2013) – whereas the steam in the central Bóndhóll and Sæluhús may originate from the same reservoir as the fluids produced from well PG-01. The samples collected from the western part of the field; Tjarnarás, Bæjarfjall, V- and S-Randir may also originate from the same water system as the PG-01 fluids, but they seem to have been subject to substantial condensation in the subsurface. This is in agreement with the interpretation of Darling and Ármannsson (1989).

Based on the fumarole isotope data, the reservoir fluid is expected to have  $\delta D$  in the range -120 to -100‰, which is much more depleted than local groundwater ( $\delta D \approx$  -73‰; see Figure 3). This suggests that the parent water of the geothermal fluid flows from as far as Vatnajökull to the south. Furthermore, as the proposed reservoir fluid is more depleted in deuterium than any modern-day precipitation (Årnason, 1976), its recharge is likely to contain a significant fraction of palaeowater from a colder regime – probably at least from the last glaciation (Sveinbjörnsdóttir et al., 2013).

## 3.3 Stable Gas Isotopes

Results for the gas isotope analyses are given in Table 1. The values in the upper part of the table are results of measurements whereas the values in the lower half are derived values. The Rc values are <sup>3</sup>He/<sup>4</sup>He ratios corrected for atmospheric He contributions (Poreda et al, 1992).

The geothermal gas in Theistareykir is characterised by a  $^{40}$ Ar/ $^{36}$ Ar ratio similar to that of air (294–296), a  $N_2$ / $^{36}$ Ar ratio of about 14,000 to 16,000 indicating that  $N_2$  and Ar are to a large extent surface-derived.  $\delta^{15}$ N in  $N_2$  is negative but variable (-2.0 to -0.5%, relative to air), which is somewhat higher than depleted MORB mantle (-5±2%; Marty and Dauphas, 2003) but within the range measured for subglacial basalts in the northern rift zone (-2.7 to 4.7%; Halldórsson et al., 2016). The  $\delta^{13}$ C values measured in CO<sub>2</sub> are approximately -2.5% (relative to V-PDB), which is in excellent agreement with the estimated pre-eruptive basaltic melts below Iceland (Barry et al., 2014). The measured  $^{3}$ He/ $^{4}$ He ratio is 10.4-10.8 Ra, which is somewhat higher then the MORB ratio of 7-9 Ra. Similar values were obtained for a Theistareykir fumarole by Poreda et al. (1992), and for a Theistareykir mud pool by Füri et al.

(2010), although the latter also measured values within the MORB range. The He/Ne ratio suggests that there is little atmospheric contribution, and therefore the corrected <sup>3</sup>He/<sup>4</sup>He ratios are only slightly higher (10.7-11.0 Ra).

Krafla and Námafjall show lower  ${}^3\text{He}/{}^4\text{He}$  ratios, about 9.8 Ra after correction for slight air contamination of the latter sample. The samples from Krafla and Námafjall also have somewhat lower  $N_2/{}^{36}\text{Ar}$  ratios, 13,800 and 12,400 respectively. The  ${}^{40}\text{Ar}/{}^{36}\text{Ar}$  ratio is 296 for Námafjall but 301 for Krafla. Both show slightly negative  $\delta^{15}N$ ; -1.43 and -0.32‰ for Krafla and Námafjall, respectively. The samples from Krafla and Námafjall are more depleted in  ${}^{13}\text{C}$  than the Theistareykir samples, with  $\delta^{13}\text{C}$  of -4.0 and -4.5‰, respectively.

Table 2: Results of gas isotope analyses and noble gas concentrations for samples collected in Theistareykir, Krafla and Námafjall in 2014. Rc denotes the <sup>3</sup>He/<sup>4</sup>He ratio after correction for atmospheric He.

Location	Sæluhús	Ketilfjall	Tjarnarás	Krafla	Námafjall
He (ppmV)	14.7	23.0	115	221	76.6
Ne (ppmV)	1.34	1.07	3.97	1.26	8.13
<sup>3</sup> He/ <sup>4</sup> He (Ra)	10.41	10.84	10.64	9.75	9.48
$^{40}$ Ar/ $^{36}$ Ar	294.7	296.1	293.8	301.2	295.6
$N_2/^{36}Ar$	14016	15714	14303	13863	12365
δ <sup>15</sup> N (‰ATM)	-1.968	-0.593	-0.487	-1.427	-0.319
δ <sup>13</sup> C (‰PDB)	-2.57	-2.45	-2.79	-3.98	-4.51
He (μmol/kg)	1.16	1.95	2.05	2.83	0.232
Ne (μmol/kg)	0.105	0.0904	0.0707	0.0161	0.0246
Rc (Ra)	10.69	10.98	10.75	9.76	9.78
$CO_2/^3He$ (x $10^9$ )	2.88	1.64	0.409	0.304	0.439

Figure 4 shows the corrected  ${}^{3}\text{He}/{}^{4}\text{He}$  ratios plotted against  $\delta^{15}N$  (left) and the  $CO_{2}/{}^{3}\text{He}$  ratio (right), for the samples from Peistareykir, Krafla and Námafjall, along with ranges for expected endmembers; air, crust, MORB and the lower mantle. The  $CO_{2}/{}^{3}\text{He}$  ratios are generally within the MORB range (Marty and Jambon, 1987).

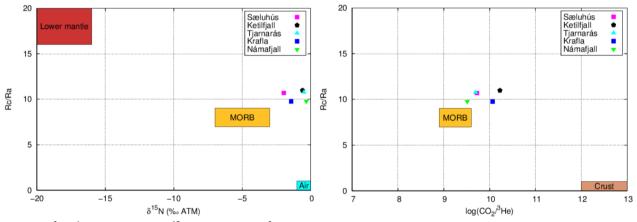


Figure 4:  ${}^{3}$ He/ ${}^{4}$ He ratio against  $\delta^{15}$ N (left) and CO<sub>2</sub>/ ${}^{3}$ He ratio (right), for the samples from Þeistareykir (Sæluhús, Ketilfjall, Tjarnarás), Krafla and Námafjall. Also shown are ranges for expected endmembers.

### 4. CONCLUSIONS

The gas composition suggests considerable variability within the Theistareykir field, both in terms of estimated reservoir temperatures as determined by gas geothermometry and fluid origin as determined from stable hydrogen and oxygen isotopes. Estimated temperatures range from 250 to 320°C, with increasing temperature towards the east, i.e. from Randir to Ketilfjall. The geothermal fluids in Theistareykir are much more depleted in deuterium than local groundwater and present-day precipitation, suggesting that the recharge fluids contain a palaeowater component. The gas isotopes suggest superficial origin of  $N_2$  and Ar, but  $\delta^{13}C$  values are in excellent agreement with Icelandic pre-eruptive basalt melts. The  $^3He/^4He$  ratios measured in Theistareykir are higher than in nearby Krafla and Námafjall – and all are somewhat higher than what is expected for MORB.

During the monitoring period, little changes have been observed in gas composition and overall surface activity of the Theistareykir field, although geothermal manifestations regularly form, change and disappear. The latter part of the monitoring period covers some years of drilling and well testing and half a year of production from the Theistareykir field, whereas limited operations were ongoing during the first half of the period, which will therefore provide a valuable baseline for future monitoring of the field. Since production started from the field, the hydrogen and oxygen isotopes of steam from one fumarole have changed substantially, suggesting that the fumarole now draws steam from a different aquifer. This is most likely a result of pressure change in the reservoir due to production.

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