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Gas Isotope Characterisation of the Reykjanes Geothermal Field, Iceland

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ABSTRACT

We present gas concentrations and stable gas isotope results for samples collected from production wells and a fumarole in the Reykjanes geothermal field in SW-Iceland in 2007, 2009 and 2013. The geothermal gas in Reykjanes is characterised by a low $^{40}Ar/^{36}Ar$ ratio, a $N_2/^{36}Ar$ ratio of about 14500 and a negative but variable measured $\delta^{15}N$ in N_2, suggesting that N_2 and Ar are to a large extent surface-derived. The $\delta^{13}C$ values measured in CO_2 are close to the estimated magmatic ^{13}C endmember for Icelandic geothermal fluids (-3.8‰). The He concentrations and measured $^3He/^4He$ (R) ratio have decreased significantly since the commission of the 100 MWe Reykjanes Power Plant in 2006, especially in the SW-part of the field where production has been most extensive. When corrections have been made for atmospheric contributions to He and N_2, the isotopic signature of the deeper component suggests an origin deep in the mantle, with R from 15 to 17 Ra and a $\delta^{15}N$ down to -17‰. The corrected R and $\delta^{15}N$ show a stronger deep mantle fingerprint for the samples collected in 2013 than the older samples.

1. INTRODUCTION

The Reykjanes geothermal system is located on the SW-tip of the Reykjanes Peninsula in SW-Iceland (Figure 1). The reservoir temperature below 1 km depth ranges from about 270 to 310°C and the fluid is hydrothermally modified seawater with some addition of magmatic gases (Arnórsson, 1978). The areal extent of surface manifestations is of the order of 2 km^2 and extensive drilling seems to indicate that the productive part of the geothermal system is not significantly larger. The surface manifestations at Reykjanes include steam vents, mud pits and warm ground. The intensity of the surface activity is known to vary over time; it increases abruptly as a result of seismic activity and then decreases slowly over time until the next seismic events occur.

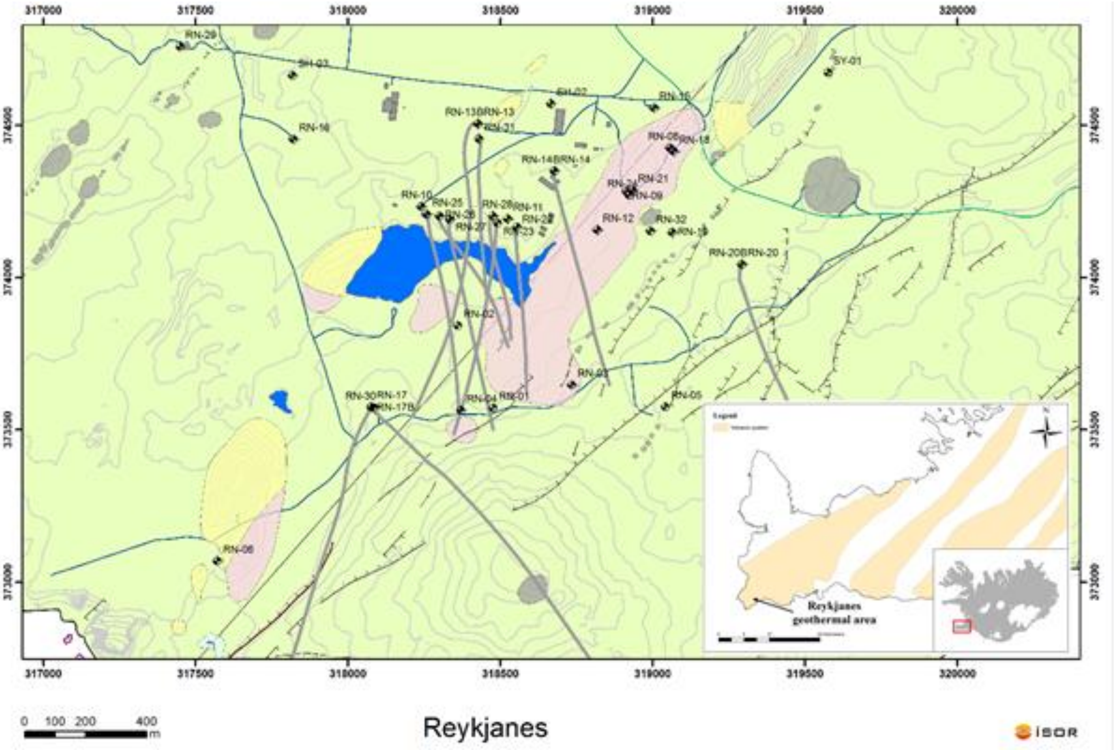

Figure 1. Well head locations and well paths in the Reykjanes geothermal field. Recent alteration on the surface is indicated by pink, and yellow indicates fossil alteration. Location of the Reykjanes field is shown on the insert maps in the lower right hand corner.

The history of geothermal production from the Reykjanes system dates back to 1970 when the first proper production well, well number 8, was brought into production. During the period from 1970 to 2006 at least one well was in production with the exception of the period from 1975 to 1978, generally with a production of 40 to 80 kg/s. In 2006 the 100 MWe Reykjanes Power Plant was commissioned. The consequent increase in production to about 750 kg/s in 2007 resulted in a dramatic pressure drop in the reservoir amounting to about 35 bar in the first three years, but since 2009, production has been about 500 to 600 kg/s so pressure drawdown has not been as rapid as in the first years. The total pressure drop from 2006 to 2013 was about 40 bar (Gylfadóttir, 2014). The pressure drop and associated drawdown has resulted in formation of a steam cap over the liquid dominated part of the reservoir, allowing the production of dry steam from shallow wells. The pressure drawdown has also caused increased boiling in the system resulting both in higher enthalpy of the well discharge and a significant increase in the surface geothermal activity (Fridriksson et al., 2010). On the other hand, the δD and $\delta^{18}O$ of the calculated reservoir fluid have had fairly stable values of about -20‰ and -0.5‰ (against Standard Mean Oceanic Water; SMOW), respectively, since production started from the field, indicating no influx of cold groundwater or seawater. The effects of progressive boiling of the geothermal fluids have been observed in samples collected from the production wells, in particular the ones producing from the SW-part of the field. Among the observed changes due to progressive boiling are increasing concentrations of non-reactive non-volatile components such as Cl and decreasing concentrations of non-reactive volatile components such as N_2. These changes and other observations regarding the effects of production on the chemical characteristics of the Reykjanes fluid are discussed in further detail by Óskarsson et al. (2015).

Alongside regular sample collection for geochemical production monitoring of the Reykjanes field in October 2013, vapour samples were collected for the analysis of isotopes in the gas phase, namely ^3He, ^4He, ^{36}Ar, ^{40}Ar, ^{13}C in CO_2 and ^{15}N in N_2, as well as total concentrations of He and Ne. The concentrations of CO_2, H_2S, H_2, CH_4, N_2, O_2 and Ar in the vapour phase were determined as a part of the regular geochemical monitoring, as well as major and trace components in the liquid phase and stable isotope ratios of H and O in liquid and condensate. The objective of this communication is to present the results of the stable gas isotope analyses of the abovementioned samples.

Some earlier sampling for gas isotope analyses has been carried out in Reykjanes (Sano et al., 1985; Poreda et al., 1992; Füri et al., 2010), yielding $^3He/^4He$ ratios (R) as high as 14 times the atmospheric value (Ra), which is considerably higher than the established value for mid-oceanic ridge basalts (MORB; 7-9 Ra), and $\delta^{13}C$ of about -4‰ (against Pee Dee Belemnite; PDB) which is taken to represent the magmatic endmember for high-CO_2 geothermal fluids in Iceland (Marty et al, 1991). In addition to the published results already mentioned, the authors collected samples from Reykjanes well 11 in the spring of 2007 and from wells 19, 21 and 23 as well as a fumarole in the autumn of 2009. The results of these analyses have not yet been published but will be presented here for reference.

2. SAMPLE COLLECTION, ANALYSIS AND RESULTS

Samples were collected from ten production wells in Reykjanes. A portable Webre separator was connected to the well heads and used to collect liquid and vapour separately. Samples for 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 analysis of ^{13}C, for which dry gas was collected into rubber-stoppered glass phials (vacutainers). The gas and isotope content of the samples was analysed by mass spectrometry, gas chromatography and titrimetry at the INGV laboratories in Naples and Palermo and ÍSOR laboratories in Reykjavík. The ^{36}Ar, N_2 amount and the N_2 isotope composition was determined on the same aliquot of sample allowing a good evaluation of air and deep component percentage.

Results for the gas isotope analyses are given in Table 1. Two gas bottles, for samples from wells 19 and 27, were broken during transport to Palermo so there is no nitrogen or argon isotope data for those samples. The bottle for the measurement of He and Ne in the sample from well 12 was contaminated by air (about 15%), which gives a too low $^3He/^4He$ ratio, and the bottle for the measurement of N and Ar isotopes in the sample from well 26 had a rather low pressure of head-space gas, thus reducing the resolution of gas chromatography and magnifying errors in the calculated gas ratios. This should be kept in mind when looking at the results.

Table 1. Results of gas isotope analyses for samples from Reykjanes, 2007, 2009 and 2013.

Well	R/Ra	R/Ra corr	$\delta^{13}C$ ‰PDB	$^{40}Ar/^{36}Ar$	$N_2/^{36}Ar$	$\delta^{15}N$ ‰ATM	$\delta^{15}N$ corr ‰ATM	N_2 atm %
RN-11	13.61	13.73	-	-	15902	-3.01	-9.49	60.3
RN-23	13.68	13.74	-	301	15621	-2.47	-8.11	43.3
Fumarole	13.67	13.74	-	310	14787	-1.54	-5.81	26.7
RN-21	13.53	15.44	-	297	14465	-4.83	-19.40	57.5
RN-19	11.73	12.02	-	300	14268	-4.65	-19.49	44.8
RN-18	12.65	15.60	-3.04	296	14276	-3.36	-14.04	33.4
RN-24	14.74	15.67	-3.02	305	14551	-3.43	-13.53	34.3
RN-12	12.61*	15.96	-2.88	313	14953	-2.29	-8.39	35.1
RN-14b	11.88	16.48	-4.44	307	14504	-4.05	-16.15	31.0
RN-13b	10.09	15.28	-3.00	296	14530	-2.37	-9.37	39.8
RN-26	7.34	12.54	-4.28	298*	13740*	-4.69*	-22.39*	13.5
RN-27	8.23	14.20	-3.17	-	-	-	-	-
RN-11	7.72	16.99	-3.86	301	14192	-2.48	-10.59	43.6
RN-23	9.31	16.79	-3.35	295	14213	-4.02	-17.04	27.9
RN-19	8.75	15.58	-3.37	-	-	-	-	-

As the sampling pressures are markedly lower in 2013 than in previous years, it is difficult to directly compare measured concentrations in the vapour phase, and therefore the gas concentrations in the deep liquid are calculated for each sample and presented in Table 2. The deep liquid is calculated by combining the vapour and liquid fractions according to the vapour fraction at collection. The vapour fraction on the other hand depends on sample pressure and the reservoir temperature for each well, which is determined from temperature logs and geothermometry. The established reservoir temperature of the Reykjanes wells ranges between 270 and 310°C.

Table 2. Gas concentrations in calculated deep liquid of Reykjanes wells. Also given is the sampling pressure and reservoir temperature for each well.

Well	P_s bar-g	T_{res} °C	CO_2 mg/kg	H_2S mg/kg	N_2 mg/kg	O_2 mg/kg	Ar mg/kg	H_2 mg/kg	CH_4 mg/kg	He µg/kg	Ne µg/kg
RN-11	43.8	295	1420	42.0	12.3	<0.01	0.34	0.09	0.15	82.7	12.3
RN-23	32.5	290	915	22.0	10.1	<0.01	0.25	0.33	0.11	56.4	7.44
RN-21	25.5	285	882	31.2	10.0	<0.01	0.23	0.24	0.06	15.2	31.8
RN-19	24.2	275	881	33.0	7.81	<0.01	0.17	0.39	0.03	2.95	1.20
RN-18	24.0	285	1040	29.1	9.22	<0.01	0.26	0.27	0.10	1.63	5.23
RN-24	24.0	285	1360	32.3	11.6	<0.01	0.34	0.93	0.13	3.20	3.20
RN-12	27.0	295	2220	65.5	11.7	<0.01	0.33	0.10	0.19	3.02	10.7
RN-14b	25.3	300	1370	48.0	10.5	<0.01	0.31	0.49	0.09	1.68	7.92
RN-13b	27.0	295	1230	58.3	18.5	<0.01	0.53	0.72	0.23	1.31	7.53
RN-26	25.5	295	741	41.4	5.73	<0.01	0.15	0.30	0.05	0.54	3.89
RN-27	26.0	270	658	32.7	6.78	<0.01	0.19	0.50	0.08	0.55	3.97
RN-11	32.3	295	1590	55.7	13.8	<0.01	0.40	0.12	0.23	1.23	11.3
RN-23	25.0	290	850	36.0	9.31	<0.01	0.27	0.29	0.09	0.77	5.82
RN-19	22.7	275	878	25.6	7.65	<0.01	0.23	0.49	0.05	0.74	5.50

3. DISCUSSION

3.1 Reservoir changes

The most striking results are that the He concentrations have decreased considerably since 2007 (the deep liquid concentration in well 11 measured 82.7 µg/kg in 2007 but only 1.23 µg/kg in 2013) and that the measured $^3He/^4He$ ratio (R) has decreased significantly in well 19 and in the wells that produce from SW-part of the reservoir. The measured R ratio was about 13.6 Ra for well 11 in 2007, wells 21 and 23 and the fumarole in 2009, but the development towards lower R and decreasing He concentrations seems to have already begun in well 19 in 2009, where the ratio measured 11.7 Ra and the He concentration in deep liquid was 2.95 µg/kg. The established R ratio for MORB is (8±1) Ra but it is expected to be > 20 Ra for the deep mantle (Poreda et al., 1992), so the current results suggest that helium in the discharge is to a lesser extent of deep magmatic origin in 2013 than in previous years.

In 1981, Poreda et al. (1992) collected samples from a well and a fumarole in Reykjanes and measured R ratios of 14.3 Ra and 14.4 Ra. In the samples from 2013, R is lower than 10 Ra in wells 11, 19, 23, 26 and 27, approximately 10 Ra in well 13b and higher in the other wells, although only well 24 has a value higher than 13 Ra. Füri et al. (2010) report R ratios of about 10 Ra for samples from a Reykjanes fumarole in 2007. It is not clear whether it is the fumarole sampled by ÍSOR and INGV in 2009.

It appears that there are two different explanations for this lowering of the R ratio; in well 19 it is probably caused by the reinjection into well 20b as the injected liquid is largely degassed and the dissolved gases that remain are to some extent in equilibrium with gas in the atmosphere. Tracer injection into well 20b in 2013 has confirmed the clear connection between the two wells (Matthíasdóttir et al., 2015). In the SW-part of the Reykjanes field, it is likely that the deep magmatic input is not sufficient to sustain the current level of production and therefore recharge to the field is to a greater extent from the shallower parts of the reservoir. This is illustrated in the left panel of Figure 2, where the markers are positioned at the largest feed-zones for each well. It should be noted that no R values are lower than the MORB values, so it is unclear whether the values represent mixing of the deep-mantle component with a MORB-type or an atmospheric component – or both. The right panel of Figure 2 depicts the measured R ratio in well samples from 2007, 2009 and 2013 against the calculated deep liquid concentrations of He. It is apparent that both the R ratio and He concentrations decrease with time, first the latter and then the former.

The $^{40}Ar/^{36}Ar$ and $N_2/^{36}Ar$ ratios (see Table 1) appear to have quite constant values of about 300 and 14500, respectively in 2013 (excluding the sample from well 26 which had very low gas concentrations). The $^{40}Ar/^{36}Ar$ ratio was also measured in samples collected in 2009, with similar results, which are typical for air-saturated seawater (295.5; Ozima, 1994) and much lower than established values for the lower (5000±1000) and upper (≥40000) mantle (Marty and Dauphas, 2003; Burnard et al., 1997). The $N_2/^{36}Ar$ ratio had a somewhat higher average value of 14800 in 2009 and was measured at 15900 in well 11 in 2007. This is consistent with the development observed during geochemical monitoring of the field, namely that the measured N_2/Ar ratio in the deep liquid decreased by about 15% from 2006 to 2011, but has remained fairly constant since (Óskarsson et al., 2015). Although decreasing with time, the $N_2/^{36}Ar$ ratio remains higher than the established value for ASW, which is 10864.

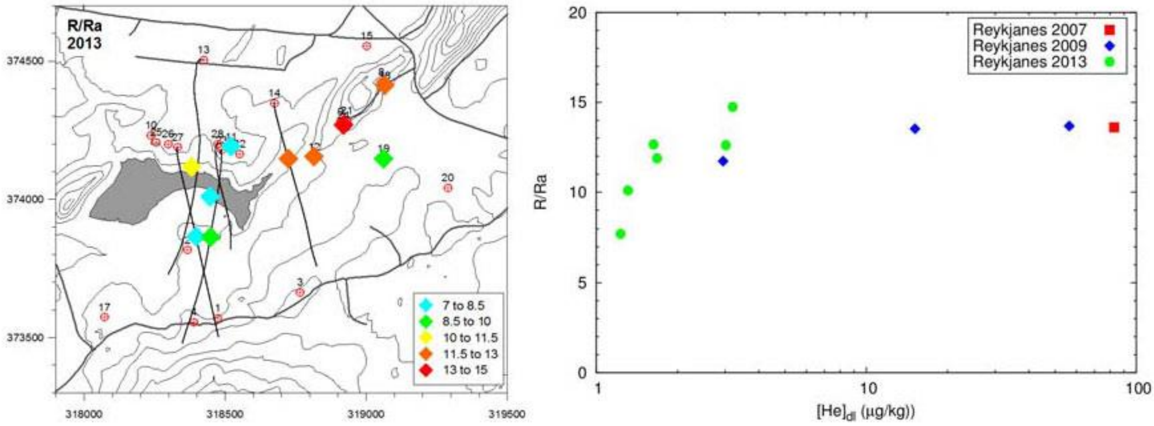

Figure 2. Left: The spatial distribution of the R/Ra ratio in the Reykjanes field in 2013. Well numbers are given next to the well head symbols. For vertical wells, the coloured diamonds are drawn at the well head location, but at the major feed zone for directional wells, for which well tracks are also shown. Right: The measured $^3He/^4He$ ratio (R/Ra) plotted against He concentration in the deep liquid. The He concentration decreases with time, as does the R-ratio.

$\delta^{15}N$ in the samples ranges between -2.3 and -4.7‰ indicating magmatic input, as the atmospheric value is 0‰ by definition. The isotope values for the mantle endmember are not as well defined, but are in any case negative. Marty and Humbert (1997) suggest a $\delta^{15}N$ of -5‰ for the MORB mantle, whereas Mohapatra and Murty (2004) argue that the mantle value is on the order of -15 to -20‰. No significant changes are seen in the measured $\delta^{15}N$ since 2009 or 2007 although the average isotope signature is slightly closer to that of atmosphere. The left panel of Figure 3 depicts the measured $^3He/^4He$ ratio of the samples plotted against the measured $\delta^{15}N$. Also shown are the values expected for lower mantle magma, MORB and air. The measured R values in 2013 are lower in some wells than in earlier years, but still higher than MORB values for most samples. On the other hand, the nitrogen isotope signature falls within the same range as before; somewhere between MORB and air.

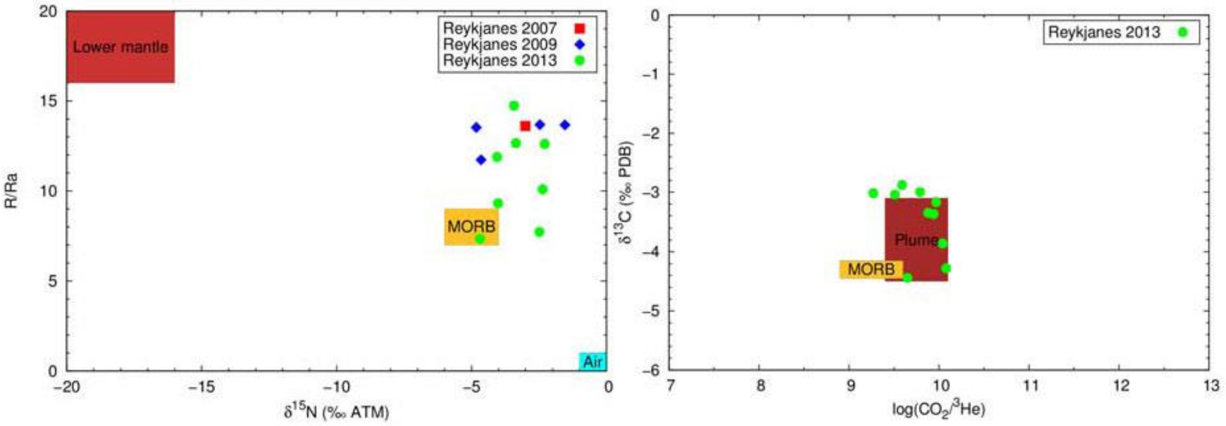

Figure 3. Left: The measured $^3He/^4He$ ratio of the samples plotted against the measured $\delta^{15}N$. Right: The $\delta^{13}C$ of the 2013 samples plotted against the measured $CO_2/^3He$ ratio.

The $\delta^{13}C$ values for the 2013 samples range from -2.9 to -4.4‰ (vs. PDB), with an average value of -3.4‰ which is close to the value reported by Poreda et al. (1992) for a sample from a fumarole in 1981. However, Poreda et al. (1992) did not report $\delta^{13}C$ in CO_2 for the well sample collected at the same time. No samples for $\delta^{13}C$ were collected in 2007 or 2009. Most of the $\delta^{13}C$ values found in 2013 fall within the range -3.8 ± 0.7‰ suggested by as the magmatic ^{13}C endmember for Icelandic geothermal fluids with high CO_2 concentrations (Marty et al, 1991; Poreda et al., 1992). These values are higher than the primary isotopic signature for mantle carbon, estimated at -5 to -8‰, compared to which the outgassed CO_2 is expected to be somewhat enriched (Javoy et al., 1986). The $\delta^{13}C$ values are plotted against the $CO_2/^3He$ ratio in the right panel of Figure 3, along with the ranges for MORB (Craig et al., 1980; Marty and Jambon, 1987) and the estimated values for the Icelandic mantle plume (Poreda et al, 1992).

3.2 The "deep" component

Helium in geothermal fluids is considered to be the product of mixing between atmospheric and magmatic components. The atmospheric part may be subtracted from the total concentrations of ^3He and ^4He by using the He/Ne ratio, as the concentration of Ne is negligible in the deep fluids (Sano et al., 1985). This manoeuvre yields corrected concentrations with R values of up to 17 Ra, see Table 1. With the exception of well 26, these values for samples from 2013 are noticeably higher than the deep-component values for samples from 2009 and 2007, which spanned the interval 12.0 to 15.4 Ra. The effect of the correction is also increasing, as the He/Ne ratio is much lower in 2013 (0.5 to 5.0) than in 2009 (2.4 to 38) and 2007 (34). Therefore, the difference between the measured R and the corrected R is much greater for samples from 2013 than in the 2007 and 2009 samples, where the difference is less than 1 Ra in all but one case (that of well 21).

The corrected R values for all samples are much higher than the values expected for upper mantle (the value for MORB 8±1 Ra) and thus suggest that the "deep" component is at least partly derived from the lower mantle for which R values as high as 20 Ra are expected according to measurements on ocean island basalts (Mohapatra et al., 2009).

In much the same way, the atmospheric and mantle contributions to the N_2 deep liquid concentrations may be calculated using the $N_2/^{36}Ar$ ratio, as the mantle component has negligible ^{36}Ar. The results of these calculations are given in Table 2 and show that the atmosphere-derived part of N_2 in 2013 amounts to 28 to 44% of total N_2 (disregarding the results for well 26). The atmospheric part was estimated at 43 to 58% of the total N_2 in the well samples from 2009 and about 60% in the sample from well 11 in 2007. The amount of mantle-derived N_2 does on the other hand appear to be similar in the deep liquid of all wells (except 26) at about 2.1 to 4.5 mg/kg. This concentration of mantle nitrogen in the deep liquid was 1.8 to 3.0 mg/kg in the well samples from 2009 and 4.0 mg/kg in 2007. Therefore, it appears that the amount of mantle-derived N_2 is rather decreasing in the deep liquid, but the concentration of atmospheric N_2 is declining even faster. This general trend towards lower N_2 concentrations in the Reykjanes field has been noted by Óskarsson et al. (2015).

The division of N_2 into atmospheric and mantle contributions also allow a similar mixing model to be applied to $\delta^{15}N$ in order to determine the $\delta^{15}N$ of the mantle component, as $\delta^{15}N = 0$ for the atmospheric component. These atmosphere-corrected values are much lower, ranging from about -8 to -17‰ (excluding well 26, see Table 1) and falling between the values expected for the lower mantle (as low as -20‰; Inguaggiato et al., 2009; Mohapatra et al., 2009) and the MORB values ($\delta^{15}N \approx -5‰$; Fischer et al., 2005). The corrected $\delta^{15}N$ values calculated in 2009 were -19.4‰ for wells 19 and 21 and -8.1‰ for well 23, and well 11 had a corrected $\delta^{15}N$ of -9.5‰ in 2007. The left panel of Figure 4 shows the $^3He/^4He$ ratio (R/Ra) of the mantle component plotted against the $\delta^{15}N$ isotope shift, also of the mantle component. Also shown are the ranges expected for air, MORB and lower mantle. These "corrected" isotopic signatures suggest that the origin of the He and N_2 in the Reykjanes system is deeper than MORB.

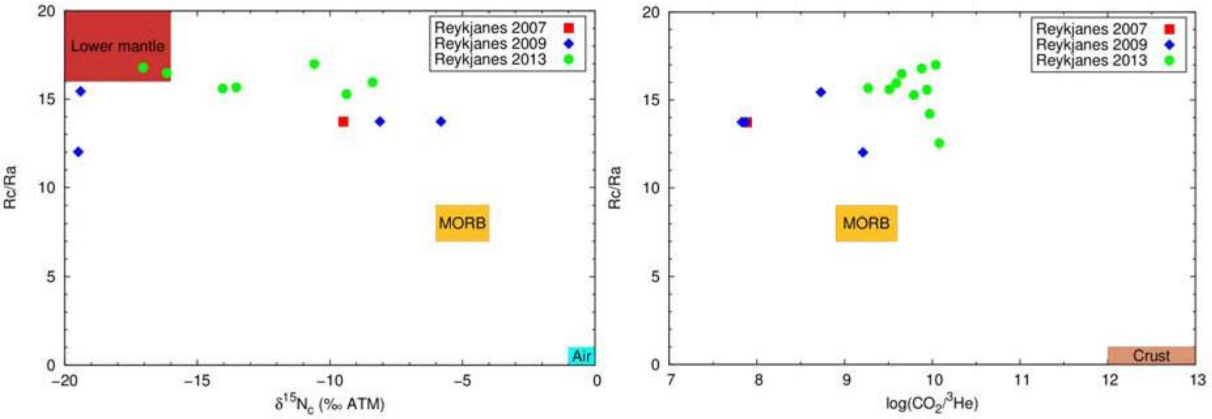

Figure 4. Left: The $^3He/^4He$ ratio of the mantle component of the samples plotted against the $\delta^{15}N$ of the mantle component. Both indicate a source deeper than MORB. Right: The $^3He/^4He$ ratio of the mantle component of the samples plotted against the measured $CO_2/^3He$ ratio. The latter is close to MORB, but the former indicates a deeper source.

The $CO_2/^3He$ ratio in the samples spans the range from 1.9×10^9 to 1.2×10^{10} with an average of 7×10^9. This ratio is higher in the SW-part of the field, where the concentrations of ^3He are lower. The average value is close to the value reported by Poreda et al. (1992) for well 7 in 1981, namely 6.1×10^9, and the lower values are similar to the value for well 19 in 2009, but the $CO_2/^3He$ ratio was considerably lower (about 10^8) for well 11 in 2007 and for wells 21 and 23 and fumarole 1 in 2009. As the CO_2 concentrations in the wells are quite stable (Óskarsson et al., 2015), these changes are mainly indicative of declining amounts of ^3He in the gas. This is in accord with the conclusion of Óskarsson et al. (2015) that substantial progressive boiling took place in the SW part of the Reykjanes field in the years 2006 to 2011. The right panel of Figure 4 shows the $^3He/^4He$ ratio of the mantle component plotted against the measured $CO_2/^3He$ ratio. Also shown are the values expected for MORB and oceanic crust (Craig et al., 1980; Marty and Jambon, 1987).

4. CONCLUSIONS

The data presented above show that there have been considerable changes in the gas isotope composition of the Reykjanes fluid since the initiation of the 100 MWe Reykjanes power plant in 2006. The overall decrease in He concentrations and the lowering of the $^3He/^4He$ ratio indicate that production is now to a greater extent from shallower fluids, which have a He isotope composition akin to air-saturated seawater. This is corroborated by the Ar isotopic ratio, which is, and has been, very close to that of air-saturated seawater, and the N_2/Ar ratio which has been declining in recent years (Óskarsson et al., 2015), and is now close to the value for air-saturated water. This trend is clearest by far in the wells that produce from the SW part of the field, where production has been most extensive since 2006. The same trend is observed in well 19, probably as a consequence of reinjection into nearby well 20b. The ^{13}C isotopic signature of the gas falls within the range expected for gas derived from the Icelandic mantle plume.

When the isotopic composition has been used to tell apart the shallower and deeper portions of the sampled gas, it appears that the "deeper" portion has an isotopic He and N signature suggesting a source in the lower mantle – or at any case deeper than MORB. Moreover, the changes in the isotope content with time seem to be moving towards a deeper source, although the generally declining gas concentrations reduce the resolution of the data. The boiling observed in the SW part of the Reykjanes field appears to affect the amount of atmosphere-derived N_2 to a greater extent than mantle-derived N_2, which has showed less decline.

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