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Enclosed is the manuscript: *Gas emissions and crustal deformation from the Krýsuvík high temperature geothermal system, Iceland.* Authors are: myself, Evgenia Ilynskaya, Sigrún Hreinsdóttir, Baldur Bergsson, Karolina Michakczewska, Alessandro Aiuppa and Auður Agla Óladóttir. We would like to submit the manuscript as an article for Journal of Volcanology and Geothermal Research. The manuscript consists of a main text, 9 color figures (requested black and white in print verison) and 5 tables.

I am the corresponding author for this article and may be contacted (see details below) regarding the revision process. I confirm that this work has not been published, nor is under consideration for publication elsewhere.

This article focuses on gas emissions and crustal deformation in Krýsuvík high-temperature geothermal system. Based on that, we suggest that the following individuals are highly qualified to act as Reviewers for this manuscript:

1. Giovanni Chiodini, INGV Bologna, giovanno.chiodini@ingv.it
2. P. A. Hernández, Institudo Volcanológico de Canarias (INVOLCAN), Spain, phdez@iter.es
3. Tasmin Mather, University of Oxford, tasmin.mather@earth.ox.ac.uk
4. Luigi Marini, Via Antonio Fratti, 253 I-55043, Viareggio, Italy, luigomarini@rocketmail.com
5. Clive Oppenheimer, Department of Geography, University of Cambridge, Downing Place, Cambridge CB2 3EN, UK, clive.oppenheimer@geog.cam.ac.uk
6. Rossella Di Napoli, University of Palermo, Italy, rossella.dinapoli@unipa.it

Sylvía Rakel Gudjónsdóttir, Sjávargata 11, 225 Gardabaer, Iceland.– Tel. +3548478611 — e-mail:srg@isor.is

Gas emissions and crustal deformation from the Krýsuvík high temperature geothermal system, Iceland.

Sylvía Rakel Gudjónsdóttir (1), Evgenia Ilyinskaya (2), Sigrún Hreinsdóttir (3), Baldur Bergsson (4), Karolina Michakczewska (5), Alessandro Aiuppa (6), Audur Agla Óladóttir (1)

(1) Iceland GeoSurvey, Grensásvegur 9, 108 Reykjavík, Iceland
(2) School of Earth and Environment, University of Leeds, Leeds, LS7 4LR, United Kingdom
(3) GNS Science, PO Box 30368, Lower Hutt 5040, New Zealand
(4) Icelandic MetOffice, Bústaðarvegi 7-9, 108 Reykjavík, Iceland
(5) Institude of Earth Sciences, University of Iceland, Sturlugata 7, Askja, 101 Reykjavík, Iceland
(6) Università di Palermo DiSTeM, Palermo, Italy

Abstract

The Krýsuvík volcanic system is located at the oblique spreading Reykjanes Peninsula, SW Iceland. Since early 2009 the region has been undergoing episodes of localized ground uplift and subsidence. In April 2013, near-real time monitoring of gas emissions was started in Krýsuvík using a Multi-component Gas Analyzer System (Multi-GAS), collecting data on gas composition (H_2O, CO_2, SO_2, H_2S). Gas emissions from Krýsuvík geothermal system are examined and correlated with crustal deformation and seismicity. The dataset comprises near-continuous gas composition time series (Multi-GAS), quantification of diffuse CO_2 gas flux, direct samples of dry gas, seismic records, and GPS data.

The gas emissions from the Krýsuvík system are H_2O-dominated, with CO_2 as the most abundant dry gas species, followed by lesser amounts of H_2S. The average subsurface equilibrium temperature was calculated as 278 °C. This is consistent with previous observations made through sporadic spot sampling campaigns. In addition, the semi-continuous Multi-GAS dataset reveals higher variations of gas composition than previously reported by spot sampling.

The diffuse soil CO_2 flux is found to be variable between the three degassing areas in Krýsuvík ranging from 10.9-70.9 T/day, with the highest flux in Hveradalir where the Multi-GAS station is located. The total flux is estimated as 101 T/day.

Comparison between Multi-GAS and geophysical data shows that peaks of H_2O-rich emissions follow events of crustal movements. Coinciding with the H_2O-rich peaks, SO_2 is detected in minor amounts (~0.6 ppmv), allowing for calculations of H_2O/SO_2, CO_2/SO_2 and H_2S/SO_2 ratios. This is the first time SO_2 has been detected in the Krýsuvík area.

The large variations in H_2O/CO_2 and H_2O/H_2S ratios are considered to reflect variable degassing activity in the fumarole. The activity of the fumarole appears less intense during intervals of low or no recorded seismic events. The H_2O/CO_2 and H_2O/H_2S ratios are lower, presumably due to H_2O condensation processes affecting the steam jet before reaching the Multi-GAS inlet tube.

Keywords: Krýsuvík, volcanic gas, volcano monitoring, geothermal gas, crustal deformation, volcanic CO_2 flux

1 Introduction

Monitoring volcanic and geothermal gases, along with seismicity and ground deformation, can lead to better understanding of volcano behaviour, and can provide early warning of volcanic activity. Several studies have focused on quiescent degassing from active volcanic and geothermal systems, detecting peaks of increased gas emissions prior to eruptions (e.g., Young et al., 1998, Aiuppa et al., 2010It has been shown that, under certain conditions, seismicity and ground deformation may help releasing gases into geothermal systems and increase fumarolic emission (e.g., Watson et al., 2000, Italiano et al., 1998, Toutain and Baubron, 1999, Chiodini et al., 2012, 2015, and references therein). Monitoring of soil CO_2 diffuse degassing in geothermal areas has also proven to give reliable information on the mass/energy budget of utilized and non-utilized geothermal systems (Brombach et al., 2001; Chiodini et al., 2001, Fridrikson, 2006, Óladóttir, 2012). In Iceland, there has been limited research on the relationship between degassing from high temperature geothermal areas (where temperature at 1 km depth is greater than 200°C) and seismic energy release and ground deformation (Ref?). The Krýsuvík volcanic system, located on the oblique spreading Reykjanes Peninsula (RP), is characterized by abundant degassing through soil and fumaroles within a high-temperature geothermal area. It has high seismic activity, characterized by swarms of micro-earthquakes as well as main-shock/aftershock sequences (Klein et al., 1977 and references therein, Ward and Björnsson, 1971, Kristjánsdóttir, 2013). In the last decade the region has undergone episodes of uplift and subsidence, with high seismic activity occurring during periods of uplift (Michalczewska et al., 2012).

Here we present semi-continuous, near-real time gas measurements in the Krýsuvík geothermal system. Such measurements are relatively new in Iceland with the first station installed on top of Mt. Hekla volcano in 2012 (Ilyinskaya et al., 2015; Di Napoli et al., 2016). In this study we evaluate the gas composition emitted from the Krýsuvík geothermal system and interpret its origin. This is done through analysis of a semi-continuous time series of gas composition (Multi-GAS sensor system, e.g., Aiuppa 2009), direct steam sampling of fumaroles, and quantification of the diffuse CO_2 flux from the geothermally active areas (accumulation chamber method, e.g., Fridriksson et al., 2006). We correlate the gas time series with geophysical observations from Krýsuvík (ground deformation and seismicity).

1.1 Regional settings

Krýsuvík is one of five active, NE-SW trending, volcanic systems located on the RP (Saemundsson et al., 2010, Hreinsdóttir et al., 2001, Einarsson, 2008) (Figure 1). The system is thought to be in an early stage of evolution, dominated by rift volcanism with no major magma chamber (Arnórsson et al., 1975). Volcanic activity is periodic with roughly 1000 years intervals between eruptive episodes, each eruptive episode lasting for 400-500 years (Saemundsson et al., 2006, Jónsson, 1978). The last volcanic eruption in Krýsuvík took place in the 12th century (Saemundsson and Sigurgeirsson, 2013). Krýsuvík currently hosts a high-temperature geothermal system, the heat source of which is considered to be dyke intrusions (Arnórsson et al., 1975, Arnórsson, 1987). Recent resistivity measurements within the Krýsuvík system indicate a conductive body at approximately 2 to 5 km depth (Didana, 2010, Hersir et al., 2013). This body is located near the central part of the Krýsuvík geothermal area, with an approximate size of 10 km^2, and coincides with the source of the inflation and deflation observed with GPS and InSAR measurements

(Michalezewska et al., 2012, Hersir et al., 2013). The lack of S-wave attenuation in the region has been used as an argument against the presence of large volumes of molten materials, but rather the presence of a gaseous or supercritical fluid (Adelinet et al., 2011).

The Krýsuvík geothermal system is often split into 6 subareas: Sandfell, Trölladyngja, Köldunámur, Seltún, Hveradalir, and Austurengjar (Figure 2). This study is focused on the last three, which have the highest and most continuous geothermal surface activity including hot and altered ground with mud pools, fumaroles, and local solfataras. The main surface activity is confined to the Vesturháls and Sveifluháls hyaloclatite ridges (including Seltún and Hveradalir), and a fault through a boiling dilute mud pool in Austurengjar (Austurengjahver) east of the Sveifluháls ridge (Markússon and Stefánsson, 2011). Resistivity measurements indicate that the geothermal subareas within Krýsuvík, originate from one and the same system of an approximate size of 40-60 km^2 (Gudmundsson et al., 1975, Saemundsson and Sigurgeirsson, 2013) (Figure 2).

2 Methodology and data processing

2.1 Gas compositon analysis using MultiGAS and direct fumarole sampling

A Multi-component Gas Analyzer System station (Multi-GAS, INGV-type, see e.g., Aiuppa 2009, Ilyinskaya et al., 2014) was installed on 26 April 2013 at Hveradalir in Krýsuvík (Figure 3), next to a fumarole in an area of high and persistent surface geothermal activity. Data collection was discontinued over a short period from 27 June – 5 July 2013 when the station was needed for another project. The sampling intake was ~20 cm above ground level (Figure 3), which was necessary to avoid saturation of the CO_2 sensor. The station was powered by a permanent wind turbine and was configured to acquire data in cycles of 200 samples, each being the median of 9 measurements @ 1 Hz (30 minutes per sampling cycle). A time interval of 6 hours between sampling cycles was set. A 3G radio modem was used for telemetry, and data were retrieved remotely using custom-made software (Ratiocalc 2.0, Tamburello, 2015) which allows for automatic creation of gas species scatter plot from the acquired data. The gas molar ratios were calculated from the gradient of best fit regression lines (Aiuppa et al., 2009, 2010) and calculations were restricted to intervals when measured concentrations showed substantial excess relative to ambient air (see e.g., Ilyinskaya, 2014). Overall uncertainty in the derived ratios is ≤20% (Aiuppa et al., 2009).

A total of 8 samples of dry gas were collected in two campaigns with 8-week interval (four samples each time, Figure 2) from fumaroles with a focussed steam flow (four samples in Hveradalir, two in Seltún, and two in Austurengjar). The samples were collected into evacuated double port bottle with 25 ml of a 10M NaOH solution. The ground temperature next to the fumarole was recorded and was in all cases >97°C.

The dry gas samples were analysed for gas composition using standard procedures at the Iceland GeoSurvey (ÍSOR), Reykjavík, Iceland. Headspace gases (N_2, CH_4, Ar, H_2, and

O_2) were analysed for using gas chromatography in a Perkin-Elmer Arnel 4019 gas chromatograph. CO_2 and H_2S were analysed for by titration of the caustic solution.

2.2 Soil temperature and diffuse CO_2 flux through soil

The soil CO_2 flux was measured using a West Systems fluxmeter in the three studied sub-areas of Krýsuvík using the accumulation chamber method (e.g., Fridriksson et al., 2006). The method has proven to be an accurate way to measure soil CO_2 fluxes in volcanic and geothermal areas, where it does not require assumptions or corrections that depend on soil characteristics (Chiodini et al., 1998). The measurements were carried out on approximately 25×25 meter grid, where possible. The total number of measurement points was 435; 217 of which were in Hveradalir, 136 were in Seltún, and the remaining 82 were in Austurengjar. Most of the measurements were taken in late summer or autumn, during dry and calm weather conditions to avoid the influence of external weather factors and water saturation of the soil. The average time of each measurement was 1-2 minutes, depending on the time the rate of CO_2 concentration increase stabilized. To evaluate the total CO_2 emission from the measured areas the Kriging algorithm (Cardellini et al., 2003, and references therein) was used for interpolation. The soil temperature was measured with a handheld digital thermometer with a 15 cm long probe.

2.3 Geophysical data

Seismic and GPS data from Krýsuvík were used to correlate with the gas measurements. The seismic data were provided by the Icelandic Meteorological Office (IMO) from the SIL seismic network.

2.3.1 Seismic data

The seismic data included 217 events in the Krýsuvík region occurring from late April through November 2013. The seismic moment was used to estimate the moment magnitude, M_W (Hanks and Kanamori, 1979):

$$M_w = \frac{2}{3(\log_{10}[(M_O) - 9.1)]} \qquad (1)$$

The largest recorded seismic event for the study period was M_W 2.2 on 11 July 2013. The frequency-magnitude distribution (Gutenberg and Richter, 1944) for the Krýsuvík catalogue gives a magnitude of completeness, M_W 0.75 and the slope or b-value of 1.6. All events M_W<0.75 were discarded from further analysis, leaving a total of 172 events.

The global average b-value is around 1 but ranges locally from 0.5-2 depending on factors like the type of tectonic environment and stress. Keiding et al., (2009) used data spanning 1997 to 2006 to evaluate the b-value for the Krýsuvík region, giving a considerably lower value of 0.9. The dataset used here (172 events) is considerably smaller which could bias the estimate. However, high b-values are often observed in volcanic and geothermal regions and assumed to be related to heterogeneous crust as well as local stress perturbations and fluids (Wyss, 1973, Schorlemmer et al., 2005).

The daily cumulative seismic moment was estimated showing several peaks of increased seismic activity over the study period, with the largest one occurring in mid-July.

2.3.2 GPS data

The GPS station MOHA started continuous operation in 2010 to monitor crustal deformation in the Krýsuvík region. The station is located just north of the center of uplift observed from 2010 to 2011 (Figure 1).

GPS data were analysed using GAMIT/GLOBK version 10.4 using over 100 continuous global GPS stations to evaluate average daily site positions in the ITRF08 reference frame. In the processing we solve for station coordinates, satellite orbit and earth rotation parameters, atmospheric zenith delay every two hours, and three atmospheric gradients per day. The IGS08 azimuth and elevation dependent absolute phase center offsets were applied to all antennas and ocean loading was corrected for using the FES2004 model. To minimize common mode signal in the time series the de-trended time series from the GPS station NYLA, outside the deforming region, were subtracted from the data. The running weighted average of seven days for the dataset was then calculated. The calculated total subsidence during the year of 2013 was $\approx$ 21 mm. Figure 4 shows clearly the local inflation/deflation periods in the last decade from the GPS time series (2007-2016) of vertical crustal movements at station MOHA, just north of the inflation center (Figure 1), along with daily cumulative seismic moment for the region.

3 Results

3.1 Assessment of the influence of meteorological conditions on the MultiGAS data

Before interpreting the MultiGAS data, we assessed the influence of meteorological conditions on the measurements. The gas ratios (H_2O/CO_2, H_2O/H_2S, H_2O/SO_2, CO_2/H_2S, CO_2/SO_2, H_2S/SO_2) were compared against wind speed (m/s) and precipitation (mm/day) data. Wind data from the three weather stations closest to the location of the MultiGAS station were investigated: Festarfjall, Selvogur and Straumsvík (IMO monitoring data). Based on the topography of the area it was considered that the atmospheric conditions are the most similar between the locations of the Festarfjall weather station and the MultiGAS station (Figure 1). The wind dataset from the Festarfjall station consists of hourly measurements. This resolution allows for an accurate comparison between the wind speed and each MultiGAS acquisition. MultiGAS data were acquired for 30 min starting at 00:00, 06:00, 12:00 and 18:00 each day. The precipitation data (IMO monitoring data) was obtained from the Keflavík airport station (Figure 1), collected twice per day (09:00 and 18:00). The representative precipitation is taken to be the total precipitation per day (where one day is defined from 18:00-18:00 UTC), and used to compare with all MultiGAS acquisitions made that same day.

All gas molar ratios were predominately observed and showed the highest fluctuations during dry periods (< 2 mm/day). However, high values for CO_2/H_2S, CO_2/SO_2 and H_2S/SO_2 ratios are not confined to the dry periods since these gas species are less affected by condensation during rainy days than H_2O. Figure 5 shows the CO_2/H_2S and H_2O/CO_2 gas ratios compared with precipitation (mm/day).

During periods of low wind speed (<5 m/s) the CO_2/H_2S, CO_2/SO_2 and H_2S/SO_2 ratios show the highest fluctuation. Progressive decrease in obtained CO_2/H_2S, CO_2/SO_2 and H_2S/SO_2 ratios and decrease in fluctuation of the ratios is observed with higher wind speeds. No visual correlation is apparent between the H_2O/X ratios (where X stands for CO_2, H_2S and SO_2) with wind speed below 10 m/s. Markedly fewer ratios are obtained when wind speed exceeds 10 m/s. Figure 5 shows the H_2O/CO_2 and CO_2/H_2S gas ratios compared with wind speed (m/s).

Based on these results it was decided to eliminate all gas molar ratios obtained during days of more than 2 mm/day of precipitation. Additionally, CO_2/H_2S, CO_2/SO_2 and H_2S/SO_2 ratios collected in periods of wind speed above 5 m/s were eliminated. This allows for using only ratios calculated from concentrations deemed slightly or not affected by the meteorological conditions, for correlation with geophysical observations.

3.2 Gas composition

The fumarole gas samples from Krýsuvík were dominated by H_2O (96.6-99.6 vol%) with CO_2 as the dominant dry gas component (on average 83.8 vol%), followed by much smaller amounts of H_2S (on average 9.17 vol%), hydrogen (H_2), nitrogen (N_2), methane (CH_4) and argon (Ar) (Table 1). One sample (Seltún 1) had a small component of O_2, a result of atmospheric contamination during sampling.

The detected MultiGAS ratios (where effects of the meteorological conditions have been eliminated) are shown to be highly variable (Table 2). Minor amounts of SO_2 were measured for the first time in Krýsuvík by the MultiGAS sensor. The measured SO_2 concentrations are very low but the bulk (63%) is above the detection limit (0.05 ppmv) of the MultiGAS sensor, allowing for calculations of the X/SO_2 ratios (X = H_2O, CO_2 and H_2S). Concentrations below 0.05 ppmv are considered to be instrumental noise. The SO_2 sensor is not quantitative below 1 ppmv so the measurements (0.05-1 ppmv) should only be viewed as qualitative assessments of SO_2 presence with a great uncertainty.

The calculated ratios of H_2O/CO_2, H_2O/H_2S and CO_2/H_2S from the fumarole samples are compared to the MultiGAS ratios in Table 3. The average value for H_2O/CO_2 and H_2O/H_2S from the fumarole samples (220 and 2812, respectively, excluding Seltún 1 sample), fall within the range of the highest H_2O/CO_2 and H_2O/H_2S ratios measured by the MultiGAS sensor (220 and 10,000, respectively). For CO_2/H_2S, the average and median values of the fumarole samples (13 and 10, respectively) are close to the average and the median values of the MultiGAS data (17 and 12, respectively).

3.3 Soil CO_2 flux and temperature

The highest soil CO_2diffuse degassing was found in areas with intense surface activity, where steam rises through fissures and cracks towards the surface, mostly in the three observation sites (total 0.31 km^2). The CO_2 fluxes ranged between 0 and 29,200 g m^{-2} day^{-1} with an average value of 385 g m^{-2} day^{-1} and median value of 6 g m^{-2} day^{-1}. In Austurengjar (0.09 km^2) 80% of all observation points show a very low CO_2 flux (<10 g $m^{-2}day^{-1}$) 68% in Hveradalir (0.140 km^2) and 51% in Seltún (0.08 km^2) (Figure 6).

The calculation of total flux from the three observation areas shows that the Hveradalir area had by far the greatest total flux of 70.9 T/day. Next was Seltún with 19.6 T/day and at last Austurengjar with 10.9 T/day. The total CO_2 soil flux from the three observation areas was estimated as 101 T/day. The soil temperature ranged between 4.2-99.0 °C with the average value 19°C and median value of 12°C.

4 Discussion

4.1 Application of gas geothermometers

Gas geothermometers (here: Arnórsson et al., 1998 and Arnórsson and Gunnlaugsson, 1985, Table 4) were used to determine the subsurface temperature of the Krýsuvík geothermal system based on the fumarole steam composition (Table 5). The gas geothermometers are based on that the concentrations of CO_2, H_2S and H_2 in geothermal reservoir waters are controlled by temperature dependent equilibria with minerals (Arnórsson and D´Amore, 2000).

The lowest individual variations were observed for both the CO_2 geothermometers, indicating sub-surface temperature of 292.6°C (Table 5). The agreement of other geothermometers was not very good as predicted temperatures varied for individual samples. The average value of the H_2S geothermometers was 267°C and of the H_2 276°C. Also, a substantial discrepancy was observed between the various geothermometers that is known from previous studies (e.g., Arnórsson et al., 1985).

The average sub-surface temperature for all samples was estimated around 278°C which is comparable to previous studies (e.g., Arnórsson and Gunnlaugsson, 1985, Arnórsson, 1987, Yohannes, 2004).

4.2 Correlation of geophysical observations and the MultiGAS data

4.2.1 Correlation with H_2O/CO_2, H_2O/H_2S and X/SO_2 MultiGAS ratios

Distinct periods with peaks in H_2O/CO_2 and H_2O/H_2S gas ratios are observed from the MultiGAS time series. SO_2 was detected almost exclusively at the same time as these detected peaks, allowing for calculations of X/SO_2 ratios (Figure 7). A visual correlation was observed between the H_2O/CO_2 and H_2O/H_2S MultiGAS peaks and crustal movements (micro-seismicity and ground deformation detected with continuous GPS measurements, Figure 7). The largest recorded seismic events for the period of this study were M_W 2.2 (11 July 2013) and M_W 1.5 (26 April 2013). Following these events and throughout the aftershock period (11 July to 7 August and 26 April to 25 may 2013, respectively) the most extensive increase in the H_2O/CO_2 and H_2O/H_2S MultiGAS ratios were observed. Similarly, moderate increases for the same gas ratios were observed during periods of moderately sized peaks of accumulative seismic moment. Peaks of high H_2O/CO_2 and H_2O/H_2S MultiGAS ratios also seemed to follow periods of ground uplift when associated with the recorded seismic events. No gas peaks were observed during period of subsidence associated with low seismic activity (Figure 7).

The H_2O/CO_2 and H_2O/H_2S MultiGAS ratios showed greater variations (1-217 and 9-10,300, respectively) than the ratios in our fumarole samples (H_2O/CO_2 range 184-269 and H_2O/H_2S range 1080-4527). During periods of recorded crustal movements, the H_2O/CO_2 and H_2O/H_2S MultiGAS ratios increase, and are closer, or equal to the gas ratios in the fumarole samples (Table 3).

It is proposed that the high variations in H_2O/CO_2 and H_2O/H_2S MultiGAS ratios are related to the intensity of fumarole activity. The fumarole activity is low during periods of low or no recorded seismic events and land subsidence (Figure 8). During these periods low H_2O/CO_2 and H_2O/H_2S MultiGAS ratios are obtained due to significant H_2O condensation (i.e., removal of H_2O from the gas phase) before the steam reaches the inlet tube. Due to the MultiGAS setup in Krýsuvík (inlet 20 cm above ground) the low H_2O/CO_2 and H_2O/H_2S MultiGAS ratios (<180 and <1000, respectively) are not representative of the emitted fumarole gas composition.

The fumaroles are interpreted to be more active during periods of recorded crustal movements. During these periods the steam rises up faster, because of opening of new pathways and increased boiling by pressure release caused by seismicity (Figure 8). As a result, H_2O is less affected by pre-sampling condensation, which results in higher H_2O/CO_2 and H_2O/H_2S gas ratios measured by the MultiGAS.

4.2.2 Correlation with CO_2/H_2S MultiGAS ratio

No visual correlation is observed between CO_2/H_2S ratios obtained by the MultiGAS and periods of recorded crustal movements (Figure 9). These gas species are significantly less affected by condensation processes than H_2O and their detection was less dependent on the variations in the activity of the fumarole. The CO_2/H_2S ratios obtained from the MultiGAS data generally fall within the range of ratios measured in fumaroles in Krýsuvík (Figure 9) However, the MultiGAS data show more variability than the fumarole data (this study, Arnórsson, 1987 and data from the Iceland GeoSurvey database). This is considered to be the result of the MultiGAS station measuring near-continuously for over 7 months, thereby picking out short-timescale variations that may be missed by point sampling. The observed variations in CO_2/H_2S ratios are therefore believed to be linked to variations in the degassing behaviour of the system.

4.3 Origin of the Krýsuvík gas emissions

The degassing regime in Krýsuvík is shown to be highly variable over short timescales (hours and days) with changes in fumarole activity and fluctuations in gas composition which is linked to variations in seismicity along with ground deformation. The results from the MultiGAS and fumarole samples resemble a typical composition of low temperature (<100°C) fumarole steam (Lee et al., 2005) from a liquid dominated system (Goff and Janik, 2000). The gas composition from this study is in good agreement to previous gas measurements in Krýsuvík (e.g., Arnórsson, 1987) and indicates no increased magmatic gas contribution. Therefore we can’t conclude that the measured SO_2 is connected with new magma intruding into the systems´s roots.

SO_2 concentrations are not expected to be high in geothermal systems like Krýsuvík. This is due to abundant water within the system where hydrolysis reactions change SO_2 into H_2S, sulfuric acid (H_2SO_4) and elemental sulfur (e.g., Ármannsson et al., 1981, 1989, Ármannsson and Hauksson, 1980, Gíslason et al., 1978, Óskarsson, 1978, 1984). However, the SO_2 detected here (0.05-0.5 ppmv) is not considered to be a false signal or interference from other gas species. This is based on two main reasons: (1) SO_2 is observed independently from high concentrations of H_2S and therefore the SO_2 measurements are unlikely to be interference between the H_2S and SO_2 sensors. (2) The MultiGAS station is equipped with highly sensitive sensors that are capable of detecting very low concentrations. It is considered possible that small amounts of SO_2 may be present in emissions from Krýsuvík. Magmatic SO_2 might be able to ascend rapidly to the surface during periods of high seismicity. Another potential source of SO_2 is near surface oxidation of H_2S to SO_2 (Arnórsson, 1987).

Eruptive activity of volcanic systems on RP is characterised by decades-long episodes of fissure eruptions. Based on eruption history, Krýsuvík may enter the next episode within the next decades. During eruptive activity, changes in fumarolic gas composition are likely to occur. Several studies (e.g., Noguchi and Kamiya 1963, Casadevall et al., 1983, Fischer and Arehart, 1996) have shown changes in the fumarole gas composition prior to and during eruptive events. Studies from the rifting episodes in Krafla, NE-Iceland during 1975-1984 (a volcanic system which bears many similarities to Krýsuvík), revealed changes in local fumarolic gas composition (e.g., Óskarsson, 1984, 1978, Gíslason et al., 1984). The gas composition was CO_2-rich during the first weeks of rifting and remained unchanged until 1983. The outgassing CO_2 was released from the deep aquifers beneath the area by the interaction of magmatic gas with the hydrothermal system.

A comprehensive record and monitoring in the Krýsuvík region would be a great asset in understanding of the gas source and the degassing regime of Krýsuvík. When new magma intrudes into the roots of the Krýsuvík system, the fumarolic gas composition can be expected to change similar to what was seen at Krafla 1975-1984 (e.g., Óskarsson, 1984, 1978, Gíslason et al., 1984). The gas composition in Krýsuvík would be expected to become CO_2 richer, resulting in lowering of H_2O/CO_2, and increase in CO_2/H_2S MultiGAS and fumarole ratios. The CO_2 gas fluxes are expected to increase as well.

Several smaller localities with apparent surface activity (mostly related to the Trölladyngja subarea) were not measured as part of this study. We therefore conclude that the total soil CO_2 flux from the Krýsuvík geothermal system during this study is higher than 101 T/day. Furthermore, the total CO_2 soil flux estimated here should be considered as the minimum value given that the amount of CO_2 dissolved in groundwater is unknown.

The total measured soil CO_2 flux from the neighbouring Reykjanes (~0.4 km^2) and Hengill volcanic systems (168 km^2) (using the accumulation chamber method) indicate total soil CO_2 flux of 78.5±13.9 T/day (Óladóttir, 2014) and 1,526 ± 160 T/day of which 453 T/day of volcanic/hydrothermal origin (Hernández et al., 2012), respectively. Studies of the volcanic systems along the RP during the last decades show little evidence of magmatic contribution with the exception of the Hengill volcano (Hreinsdóttir et al., 2001, Einarsson, 2008, Keiding et al., 2008), which suggested minor magma injection into the roots of the volcano from 1995 to 1998 that triggered an intense seismic swarm (Sigmundsson et al., 1997).

It is important to continue the monitoring of gas emissions in Krýsuvík, at least over the next episode of land uplift and elevated seismicity to compare with the dataset from this study. It would be of particular interest to observe if changes in the gas composition will occur during inflation episode, as that will give important information on the source of the inflation.

5 Conclusions

The gas composition from the Krýsuvík geothermal system was studied (April-November 2013) using the MultiGAS method and correlated with geophysical observations. The gas composition is H_2O dominated with CO_2 as the dominant dry gas species, with lesser amounts of H_2S and trace amounts of SO_2.

The gas emissions (in the form of diffuse soil degassing) were measured in three areas of high geothermal surface activity within the Krýsuvík system (Seltún, Hveradalir and Austurengjar). The total emission is estimated as 101 T/day but gives a minimum value for the total emission, as the amount of CO_2 dissolved in groundwater is not known and several smaller localities with apparent surface activity were not measured.

The time series of gas compositon (MultiGAS data) identified short-lived episodes of elevated and highly variable H_2O/CO_2 and H_2O/H_2S ratios (the highest H_2O/CO_2 and H_2O/H_2S ratios followed periods of highest accumulative seismic moment per day (26 April 2013 and 11 July 2013). SO_2 is detected at the same time. Correlation with seismicity and ground deformation shows that the ratio peaks follow increased seismic activity, with the highest H_2O/CO_2 and H_2O/H_2S ratios following elevated seismicity and crustal movements. It is proposed that these peaks represent periods of elevated fumarolic activity that are responding to the seismicity and land uplift due to opening of new pathways in the crust and increased boiling within the system. During these periods the steam reaches the inlet tube faster and is less affected by pre-sampling H_2O condensation. The CO_2 and H_2S gases are significantly less affected by condensation processes and the CO_2/H_2S ratio does not change according to the same pattern. Most of the measured CO_2/H_2S ratios fall within the known range for Krýsuvík fumaroles. However, several markedly higher values are detected, demonstrating more variability in the degassing system than previously known.

It is considered crucial to continue the MultiGAS measurements in the Krýsuvík geothermal due to recent episodes of ground uplift and subsidence as well as future volcanic activity. During the span of the MultiGAS measurements in 2013 the total subsidence in the area was ~21 mm.

Acknowledgements

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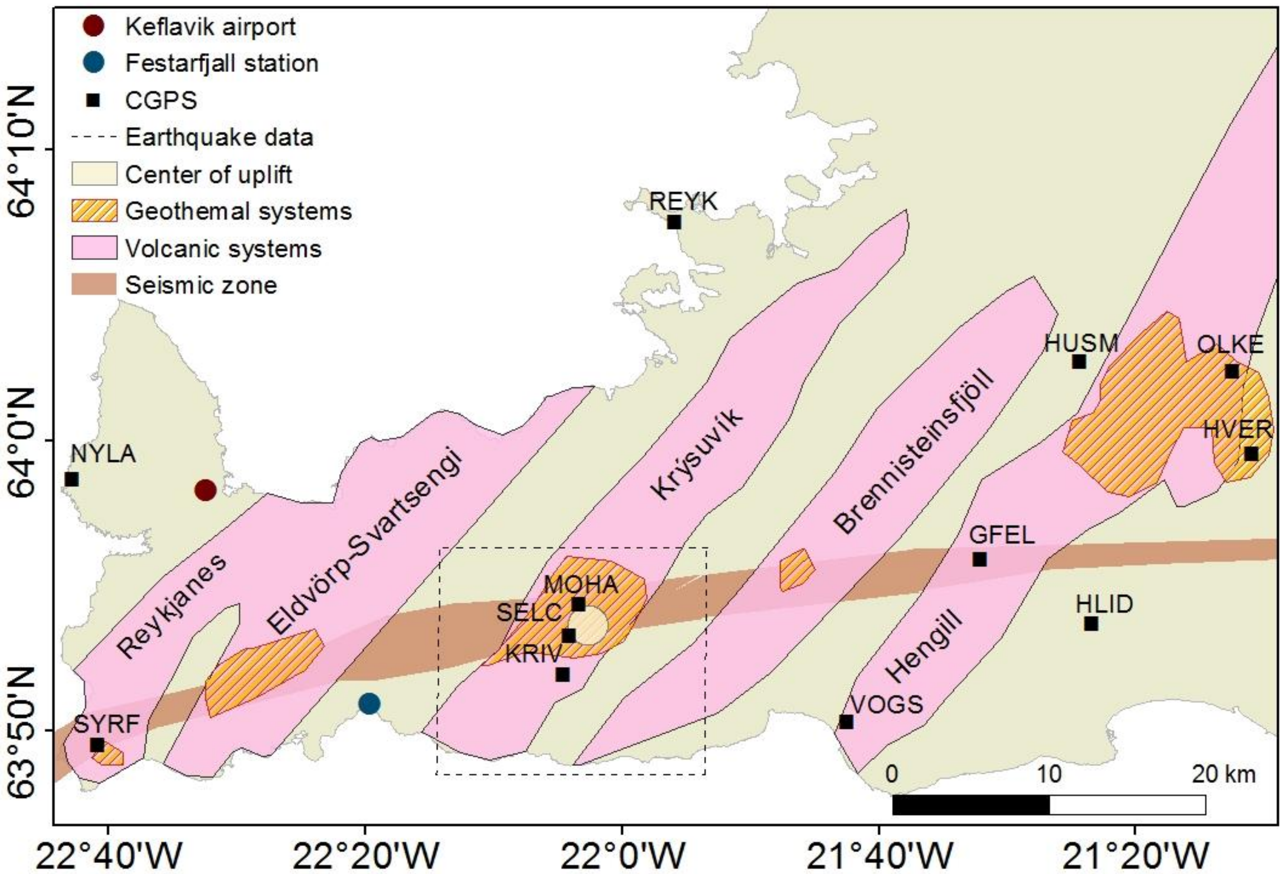


Figure 1 Volcanic systems on the RP (purple) and seismic zone across the Peninsula that marks the axis of the plate boundary (brown) (Einarsson, 2008). High-temperature geothermal areas within the volcanic systems (striped). Modified from (Sæmundsson and Sigurgeirsson, 2013). The five volcanic systems are arranged en echelon along the peninsula, spaced approximately 5 km apart (Clifton et al., 2006). Continuous GPS stations in operation in 2013 on the RP including the region of the

earthquake data (dashed box), and the center of uplift (yellow circle). Blue and dark red dots show the location of Festafjall and Keflavík airport weather stations, respectively.

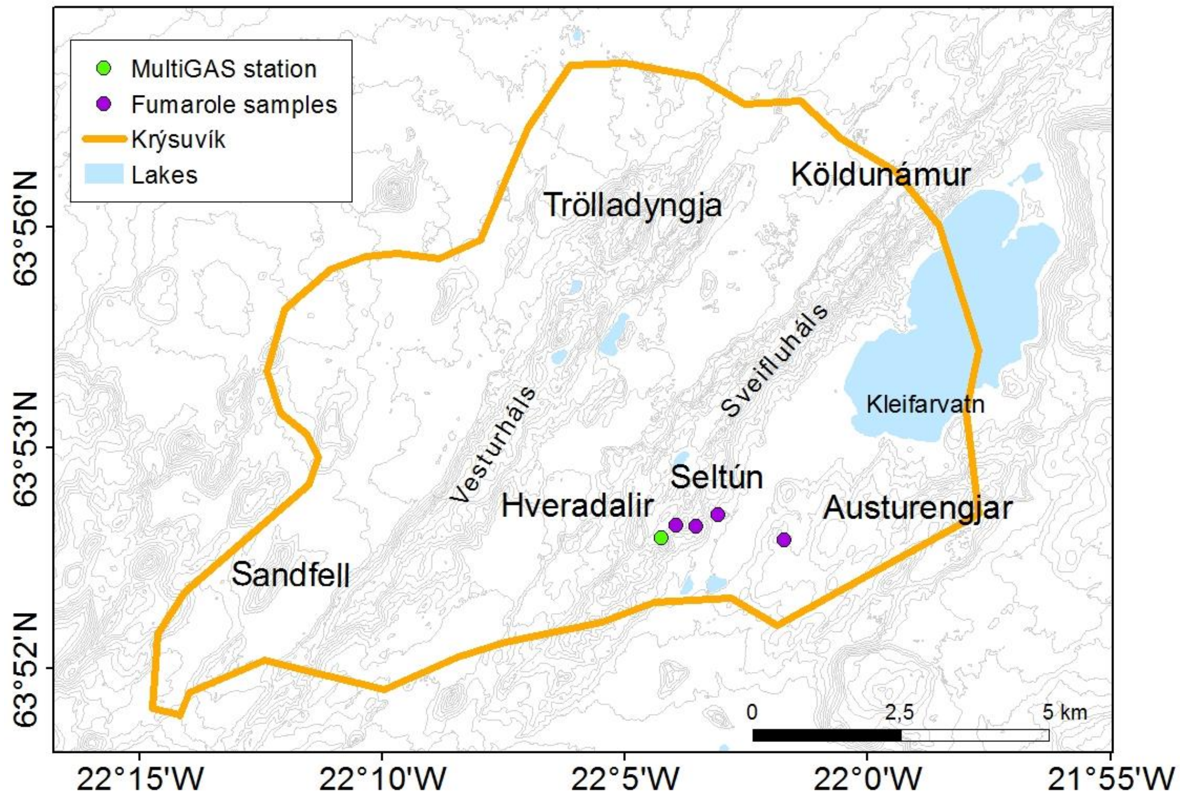


Figure 2 Outlines of the Krýsuvík high-temperature geothermal system identified by resistivity surveys (orange line) (Gudmundson et al., 1975). Krýsuvík sub-areas and the two hyaloclastite ridges, Sveifluháls and Vesturháls, with which the geothermal activity in Krýsuvík is mostly associated.

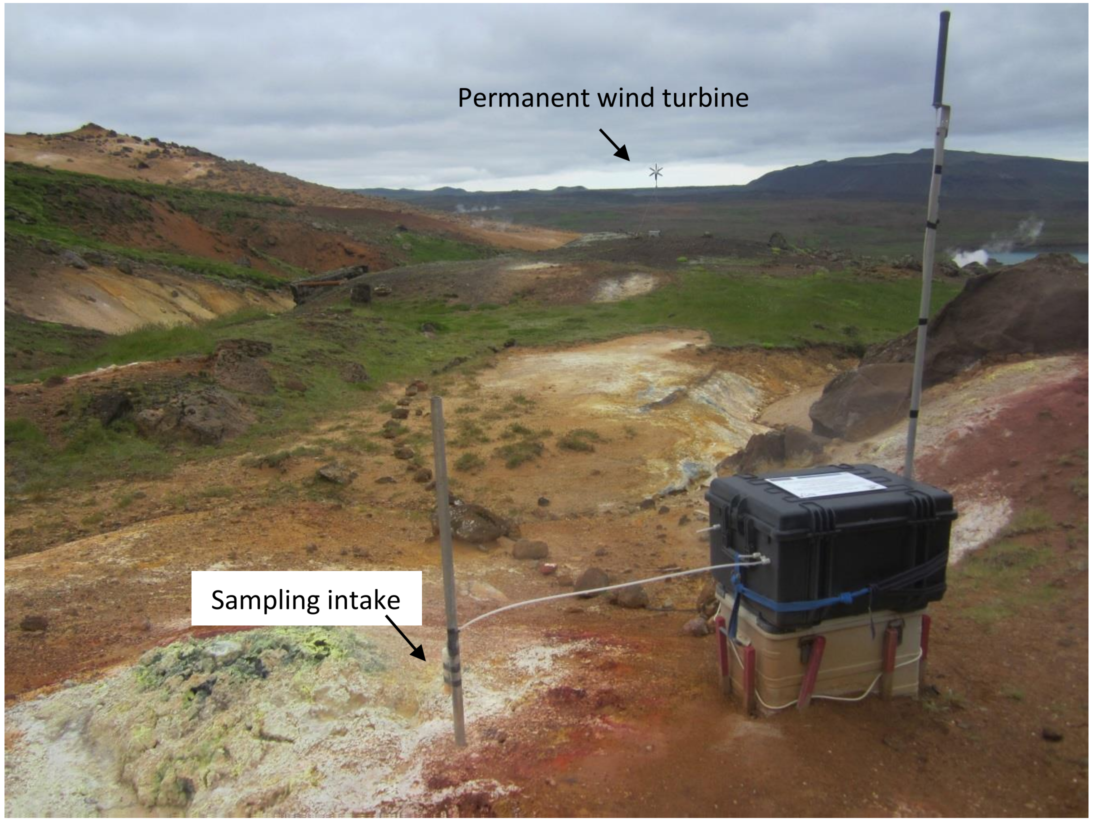


Figure 3 MultiGAS station in Krýsuvík. The station is located next to a fumarole in an area of continuous geothermal activity in Hveradalir, Krýsuvík, and is powered by a wind turbine. The sampling inlet of the MultiGAS station is located ~20 cm above the ground to avoid saturation of the CO_2 sensor.

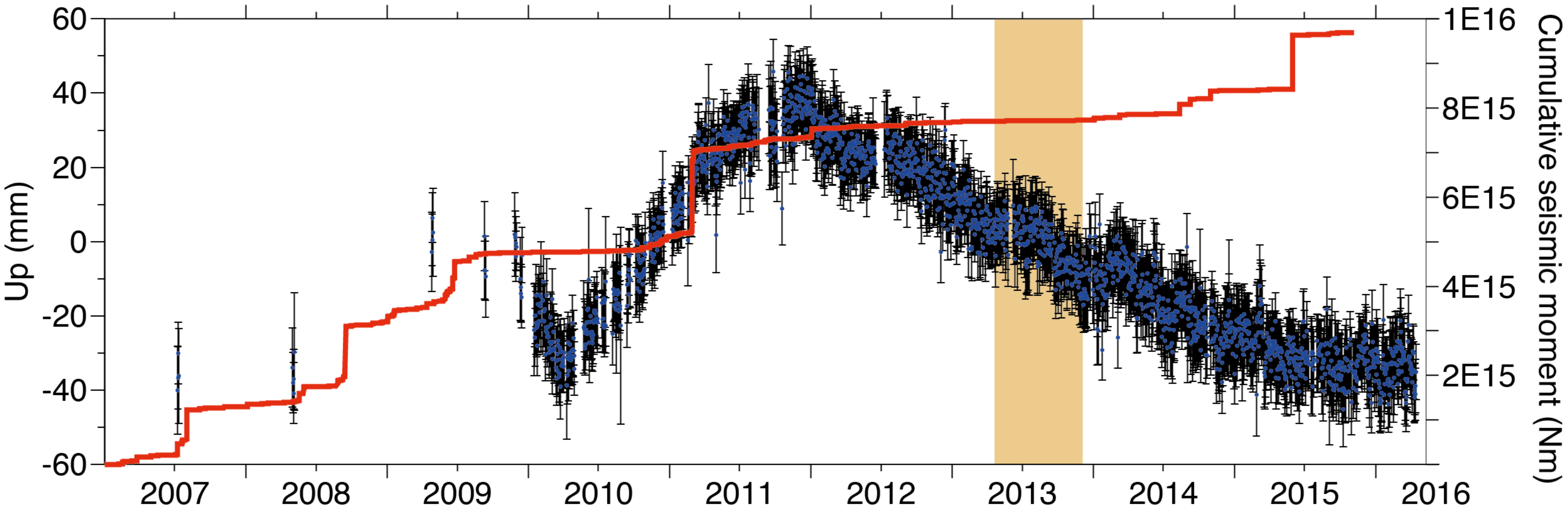
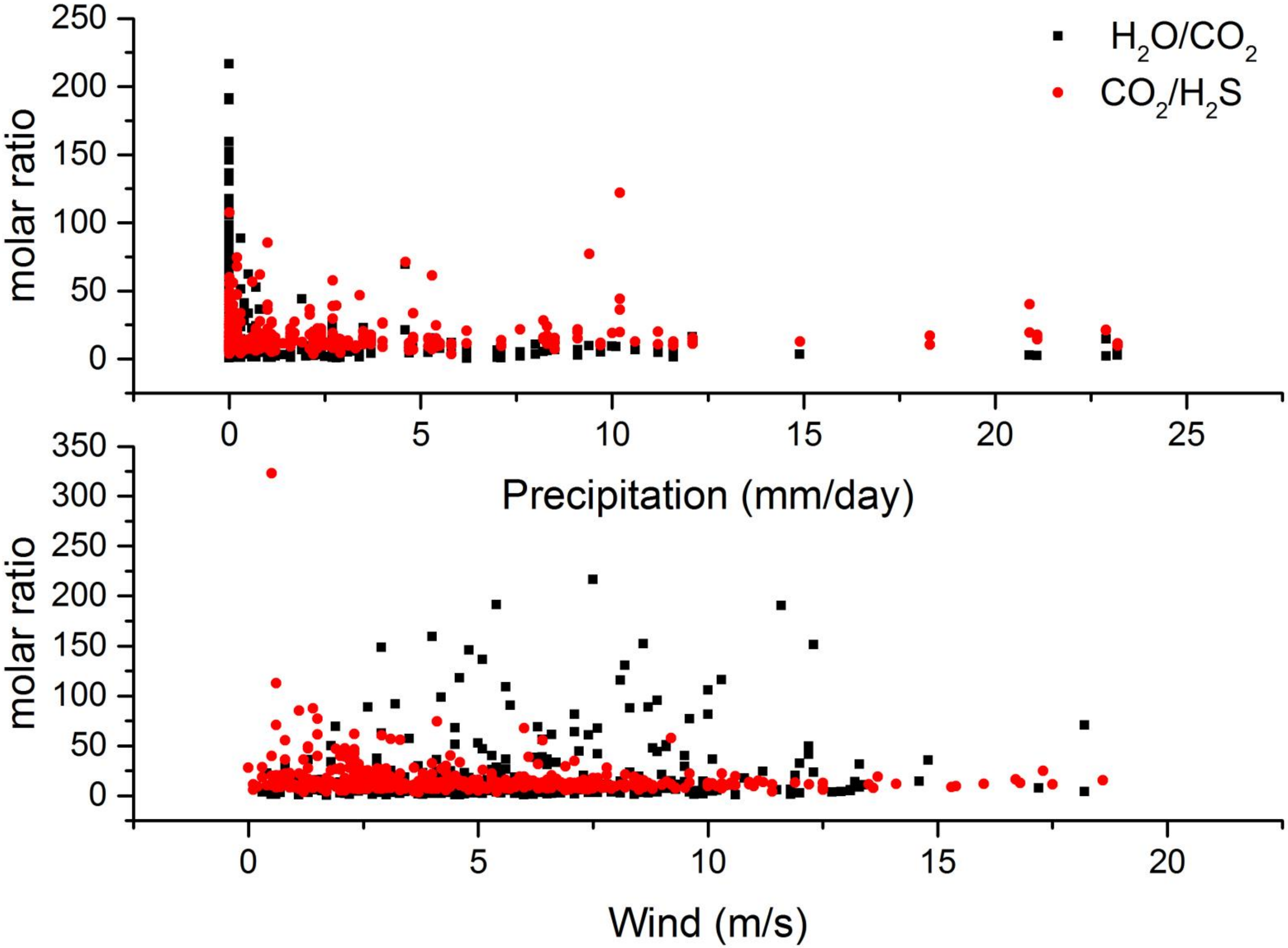


Figure 4 The local inflation/deflation periods from GPS time series (2007-2016) at station MOHA, along with daily cumulative seismic moment. The red band corresponds to the time period of this study.



H2O/CO2
CO2/H2S
molar ratio
Precipitation (mm/day)
molar ratio
Wind (m/s)

Figure 5 (Upper) CO_2/H_2S and H_2O/CO_2 molar ratios as a function of precipitation (mm/day). The CO_2/H_2S ratio are obtained predominantly during periods with <2 mm/day rainfall. However, high ratio values are not confined to the dry periods. All H_2O/CO_2 ratios >19 are obtained during dry periods (< 2mm/day). The most frequently obtained values for H_2O/CO_2 (<19) are visually evenly distubuted between ~ 0-5 mm/day. (Lower) CO_2/H_2S and H_2O/CO_2 molar ratio as a function of wind speed (m/s). The highest ratios of CO_2/H_2S (>75) and largest variations are obtained during relatively low wind speed (approximately <5 m/s). Lower values of CO_2/H_2S (<20) are detected more frequently than the higher values and there is no visible relation to wind speed <10 m/s where marked decrease is seen in the frequency of ratios obtained. Wind speed <10 m/s does not appear to affect detection of H_2O/X molar ratios (X = CO_2, H_2S and SO_2) in >10 m/s markedly fewer ratios are detected.

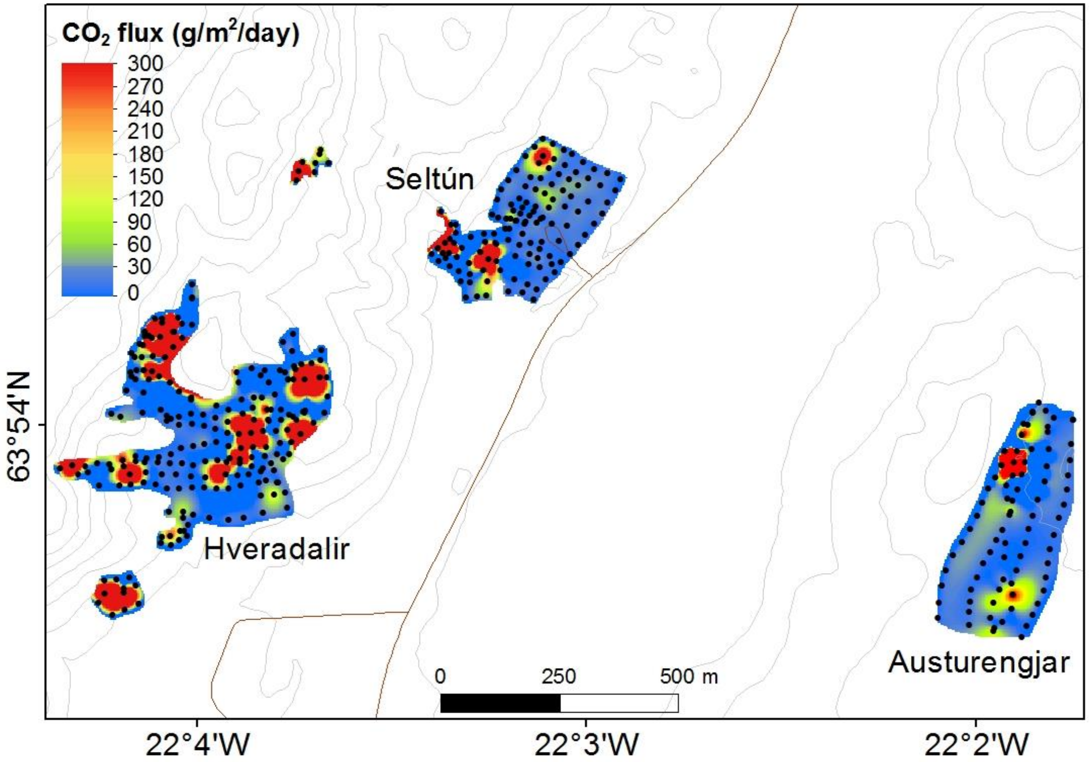


Figure 6 Soil CO_2 flux measurements from the three observation areas, Hveradalir, Seltún, and Austurengjar. The highest flux was observed in Hveradalir, 70.9 T/day. In Seltún the flux was found to be 19.6 T/day and the lowest flux was calculated in Austurengjar, 10.9 T/day. The total flux from the three measured areas was calculated as 101 T/day.

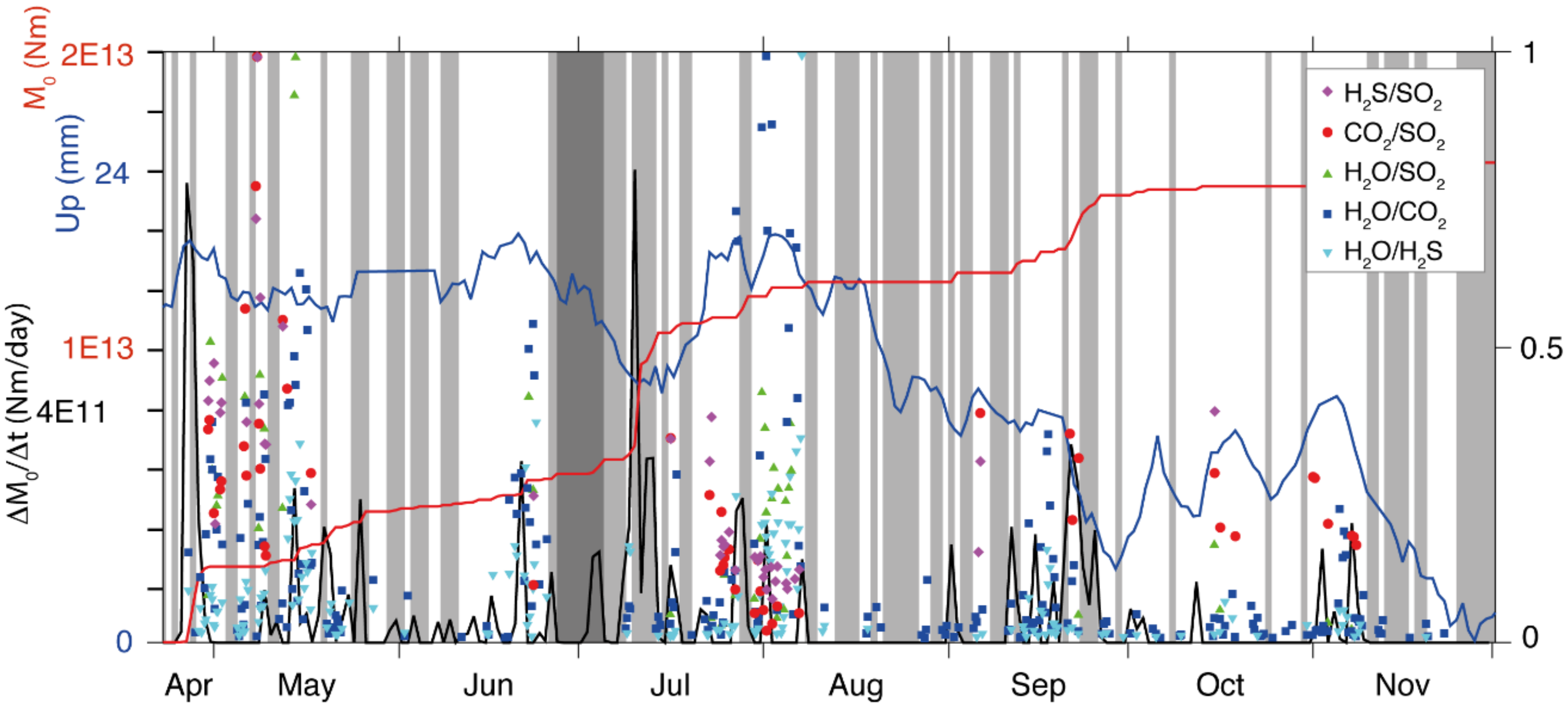


Figure 7 Normalized variations in gas composition as measured by the MultiGAS station (measurements affected by metrological conditions have been eliminated) correlated with geophysical observations. There are distinct intervals with peaks of increased H_2O/CO_2 and H_2O/H_2S ratios. SO_2 is detected during the same intervals allowing calculation of X/SO_2 ratios. Red line: cumulative seismic moment (Nm). Black line: seismic moment per day (Nm/day). Blue curve: Vertical crustal movements measured with GPS (mm). Light grey intervals: rainy days (>2 mm/day). Dark grey interval: station not operating. Peaks of increased H_2O/CO_2 and H_2O/H_2S ratios appear to follow episodes of recorded seismic events and crustal deformation.

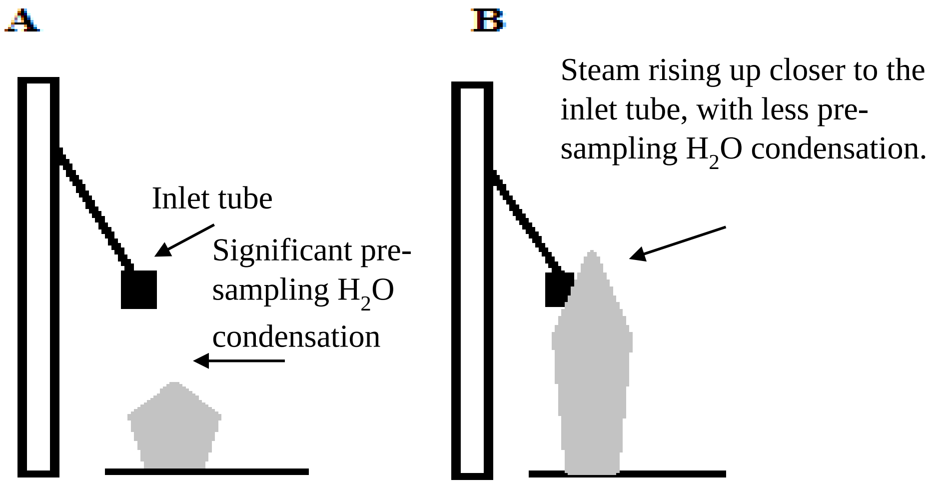


Figure 8 A) Lower fumarole activity resulting in low H_2O/CO_2 and H_2O/H_2S gas ratios measured by MultiGAS. The cause is condensation of H_2O before the gas reaches the inlet tube (20 cm above the ground). These low ratios are not representative of the fumarolic gas composition. B) Higher fumarole activity due to crustal movements. The steam is less affected by pre-sampling condensation resulting in higher H_2O/CO_2 and H_2O/H_2S ratios. These values are closer to the H_2O/CO_2 and H_2O/H_2S ratios as measured in direct samples of the fumaroles.

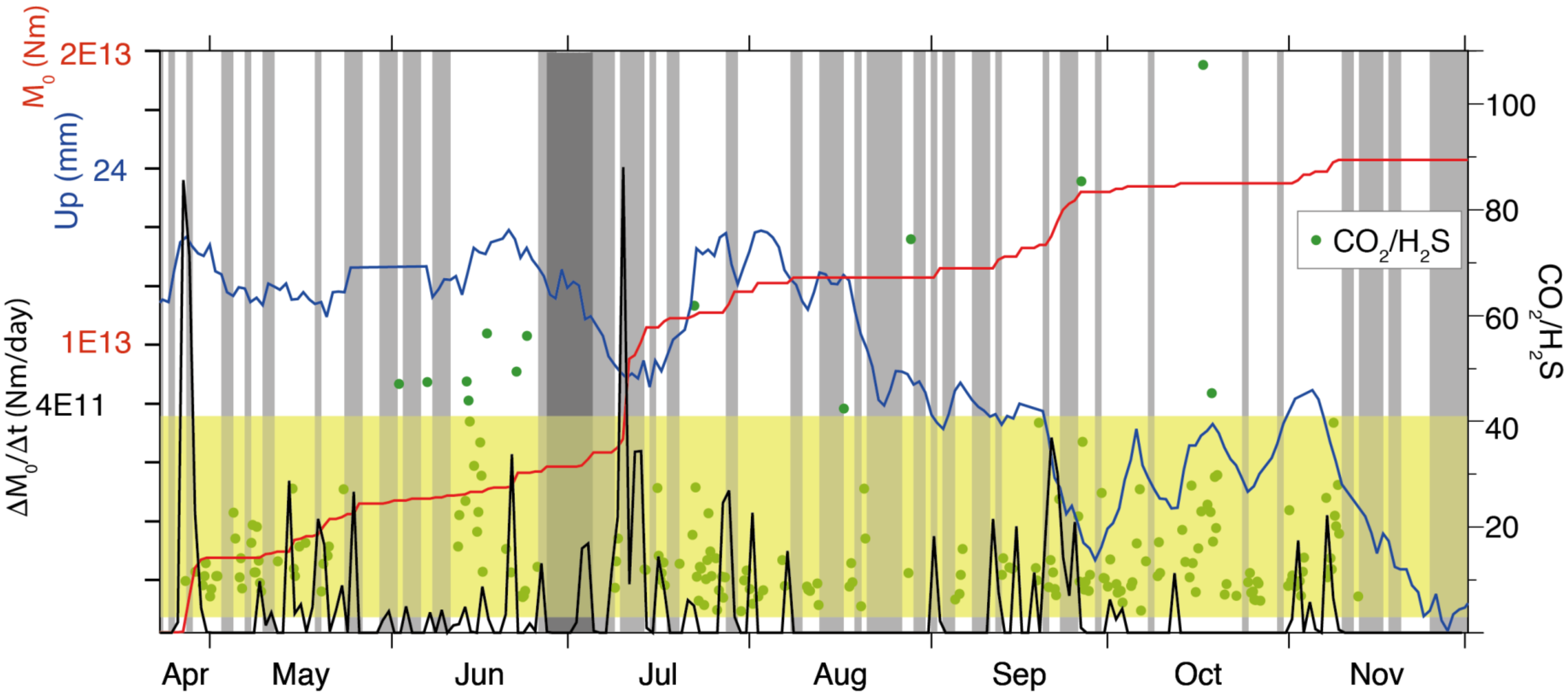


Figure 9 Time series (April-November 2013) of CO_2/H_2S MultiGAS molar ratios (where measurements effected by metrological conditions have been eliminated) compared with crustal movements. The CO_2/H_2S ratio does not show any visible variations related to seismicity or crustal deformation. The yellow band corresponds to CO_2/H_2S ratios (3-41) from fumaroles in the Krýsuvík area (this study, Arnórsson, 1987, data from the Iceland GeoSurvey database).

Table 1 *Vol% concentration in fumarolic steam from the 8 samples. Values within () for CO_2 and H_2S refer to % of total gas volume.*

Sample	**Date**	**H_2O%**	**CO_2%**	**H_2S%**	**H_2%**	**Ar%**	**O_2%**	**N_2%**	**CH_4%**
Seltún 1	18.02.2014	96.9	75.5 (2.3)	13.9 (0.430)	9.32	0.02	0.15	1.03	0.08
Seltún 2	23.04.2014	99.4	79.6 (0.48)	15.3 (0.092)	4.80	-	-	0.32	0.02
Hverahvammur 1	18.02.2014	99.5	88.9 (0.44)	8.69 (0.043)	2.02	-	-	0.40	-
Hverahvammur 2	23.04.2014	99.4	89.3 (0.54)	9.26 (0.056)	0.97	-	-	0.49	-
Austurengjar 1	18.02.2014	99.4	89.3 (0.54)	3.80 (0.023)	6.13	-	-	0.67	0.10
Austurengjar 2	23.04.2014	99.5	81.5 (0.41)	12.9 (0.065)	5.36	-	-	0.19	0.06
Hverahöfði 1	18.02.2014	99.4	73.4 (0.44)	4.00 (0.024)	19.7	0.04	-	2.65	0.22
Hverahöfði 2	23.04.2014	99.6	93.2 (0.37)	5.54 (0.022)	0.76	-	-	0.49	0.02

Table 2 *Variations of the molar gas ratios measured by the MultiGAS station in Hveradalir, Krýsuvík (excluding data affected by meterological factors).*

	Max	**Min**	**Average**	**Median**
H_2O/CO_2	217	1	27	9

H_2O/H_2S	10300	9	640	218
H_2O/SO_2	107000	2380	26900	25100
CO_2/H_2S	107	4	17	12
CO_2/SO_2	3010	58	767	662
H_2S/SO_2	148	11	42	35

Table 3 *The calculated molar ratios of H_2O/CO_2, H_2O/H_2S and CO_2/H_2S for the fumarole samples and from the MultiGAS data. Sample Seltún 1 is excluded from the average calculations due to condensation in the sampling train. The maximum H_2O/H_2S values in the MultiGAS data (10300) is by far the highest value, the second highest value (3850) is, much closer to the average value of the fumarole samples.*

	MultiGAS				Fumaroles								
Ratio	**Max**	**Min**	**Average**	**Median**	**Seltún 2**	**Hvera-hvammur 1**	**Hvera-hvammur 2**	**Austur-engjar 1**	**Austur-engjar 2**	**Hvera-höfði 1**	**Hvera-höfði 2**	**Average**	**Median**
H_2O/CO_2	217	1	27	9.0	207	226	184	184	243	226	269	220	226
H_2O/H_2S	10300 (3850)	9	640	218	1080	2313	1775	4321	1530	4141	4527	2812	2313
CO_2/H_2S	107	4	17	12	5	10	10	24	6	18	17	13	10

Table 4 *Gas geothermometers applied to Krýsuvík fumarole samples.*

Geothermometer	**Temperature function**[1]	**Reference**
CO_2	$4.724Q^3-11.068Q^2+72.012Q+121.8$	(Arnórsson et.al 1998)
H_2S	$4.811Q^2+66.152Q+177.6$	(Arnórsson et.al 1998)
H_2	$6.630Q^3+5.836Q^2+56.168Q+227.1$	(Arnórsson et.al 1998)
CO_2	$-44.1+269.25Q-76.88Q^2+9.52Q^3$	(Arnórsson and Gunnlaugsson 1985)
H_2S	$173.2+65.04Q$	(Arnórsson and Gunnlaugsson 1985)
H_2	$212.2+65.04Q$	(Arnórsson and Gunnlaugsson 1985)

[1]Q refers to the logarithm of the respective gas concentration or ratio in moles per kg of steam.

Table 5 The results from the applied gas geothermometers. The temperature value are in °C.

Sample	**nr.**	**[1]CO_2**	**[1]H_2S**	**[1]H_2**	**[2]CO_2**	**[2]H_2S**	**[2]H_2**	**Average**
Hverahvammur 1	1	292	276	274	288	261	259	275
Hverahvammur 2	2	303	268	256	295	270	243	272
Austurengi 1	3	301	255	323	294	244	295	285
Austurengi 2	4	289	290	309	286	273	286	289
Hverahöfði 1	5	292	257	281	288	245	266	271
Hverahöfði 2	6	292	259	244	288	247	231	260
Seltún 2	7	297	304	313	291	284	289	296