

An overview of carbon dioxide emissions from Icelandic geothermal areas

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ABSTRACT

The origin of CO₂ in fluids from Icelandic high-temperature geothermal systems is predominantly magmatic. Emissions from producing areas have risen with increased production. Abnormal rises have been recorded due to magmatic activity and the onset of boiling due to increase in production. Natural flow is predominantly through soil but to a small extent via steam vents and steam heated pools. The extent of natural steam flow varies considerably between areas, apparently due to the formation of carbonate deposits (mainly calcite) in relatively cool liquid dominated aquifers at shallow depths, where these are present. The CO₂ concentration of fluids from aquifers at higher temperatures apparently decreases with temperature and is for instance very low (< 1000 ppm) in fluid from IDDP-1, Krafla where the source temperature is 450 °C.

1. Introduction

The International Geothermal Association (2002) carried out a survey of CO₂ emissions from geothermal power plants in order to demonstrate the environmental advantage of geothermal energy in mitigating global warming. The results were presented in terms of emitted CO₂ per energy unit (g kWh⁻¹) in relation to production in MW_e (Table 1). The total range for all plants was 4–740 g kWh⁻¹ with a weighted average 122 g kWh⁻¹. In the report it was suggested that the natural emission rate pre-development be subtracted from that released from the geothermal operation, citing Larderello as an example of a field, where a decrease in natural release of CO₂ has been recorded, and suggested to be due to development. Italy has accordingly not presented CO₂ emissions from geothermal production as a part of emissions recorded annually in international protocols.

Geothermal systems are often located in volcanic areas or other areas of high CO₂ flux of magmatic origin, but CO₂ may also be derived from depth where it is mainly produced by metamorphism of marine carbonate rocks. There is often a large flux through soil but CO₂ dissolves in groundwater, where this is present, usually reaching saturation where the flux is sufficiently large. Processes of natural generation are independent of geothermal production. The output is very variable but usually quite substantial. Estimated output from several volcanic and geothermal areas, and a total for the world are shown in Table 2.

A thorough investigation of the proportion of CO₂ emitted through various conduits in Pantelleria Island was conducted by Favara et al. (2001), but estimates of fractions emitted through groundwater on the one hand and soil and fumaroles on the other have been made at

Mammoth Mountain (Sorey et al., 1998; Evans et al., 2002; Gerlach et al., 2001) and Furnas (Cruz et al., 1999). The results for these areas are listed in Table 3, along with results for Reykjanes, Iceland, discussed below.

Thus variations in carbon dioxide concentrations in geothermal fluids may have various causes. The objective of this paper is to investigate such variations at the scale of a country, i.e. Iceland, and at the same time present a detailed overview.

2. Origin of gas in Icelandic high-temperature geothermal fluids

The gas in fourteen of the fifteen areas, in which the carbon-13 isotope ratio has been studied, is apparently magmatic in origin, whereas that in the Öxarfjörður area could originate in organic sediments (Ármannsson, 2016). Stefánsson (2017) surmised that the sources of the magmatic CO₂ and H₂S may be basalt and progressive fluid rock interaction and/or degassing of basaltic melts, either at great depth upon partial melting within the upper mantle and lower crust, or at shallower levels within the crust. Both types of source have been suggested, particularly evidenced in the case of CO₂ (e.g., Stefánsson et al., 2016), whereas H₂S is considered to originate predominantly from basalt upon rock leaching (Stefánsson et al., 2015; Gunnarsson-Robin et al., 2017).

3. Gas emissions from geothermal activity in Iceland

The CO₂ emission from Icelandic geothermal plants has been recorded since about 1970 (Fig. 2). Gas concentrations in steam in Krafla

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Table 1
CO₂ emission and total running capacity of power plants divided into 9 emission categories (International Geothermal Association, 2002).

Emission category (g/kWh)	Running capacity (MW _e)	Average (g/kWh)
> 500	197	603
400–499	81	419
300–399	207	330
250–299	782	283
200–249	346	216
150–199	176	159
100–149	658	121
50–99	1867	71
< 50	2334	24

Table 2
CO₂ output from some volcanic and geothermal areas.

Area	Megaton (10 ⁹ g) yr ⁻¹	Reference
Pantelleria Island, Italy	0.39	Favara et al. (2001)
Vulcano, Italy	0.13	Baubron et al. (1991)
Solfatara, Italy	0.048	Chiodini et al. (1998)
Ustica Island, Italy	0.26	Etiopio et al. (1999)
Popocatepetl, Mexico	14.5–36.5	Delgado et al. (1998)
Yellowstone, USA	10–22 ^a	Werner and Brantley (2003)
Mammoth Mountain, USA	0.055–0.2	Sorey et al. (1998), Evans et al. (2002), Gerlach et al. (2001)
White Island, New Zealand	0.95	Wardell and Kyle (1998)
Mt. Erebus, Antarctica	0.66	Wardell and Kyle (1998)
Taupo Volcanic Zone, New Zealand	0.44	Seward and Kerrick (1996)
Furnas, Azores, Portugal	0.01	Cruz et al. (1999)
Mid-Ocean Volcanic System	30–100	Gerlach (1991), Marty and Tolstikhin (1998)
Total	200–1000	Mörner and Etiopio (2002), Kerrick (2001), Delgado et al. (1998), Marty and Tolstikhin (1998)

^a Diffuse degassing only.

Table 3
Relative CO₂ emission through different conduits from four areas (Favara et al., 2001; Sorey et al., 1998; Evans et al., 2002; Gerlach et al., 2001; Fridriksson et al., 2006).

	Pantelleria Island	Furnas Volcano	Mammoth Mountain	Reykjanes
Soil %	81	49 ^a	63–90 ^a	97
Focussed degassing %	7			
Fumarole %	0.0004			2
Bubbles %	3			
Groundwater %	9	51	10–37	1

^a Total flow directly to atmosphere.

were relatively high during the late seventies and eighties due to magmatic gas. These have stabilized, but the increase seen around 2000 is due to increased production. As is frequently observed the gas concentrations decreased gradually with steady production and seem to have reached stability. The gas concentrations in Svartsengi rose in the early nineties due to the formation of a steam cap and increased production from that cap. A steady value has been reached, which may be expected to decrease if production is not increased. As is expected the gas emissions from Hellisheiði have increased during the power plant's first years of production. A similar rise but not as drastic is observed at Reykjanes.

The emissions from Nesjavellir are low and relatively constant. A comparison between the CO₂ emissions per kWh from the major geothermal plants in Iceland shows that they can be divided into two

groups, i.e. Krafla and Svartsengi on the one hand but Hellisheiði, Reykjanes and Nesjavellir on the other (Table 4). The table also shows that the emissions per kWh in Krafla and Svartsengi have decreased since the year 2000. The effect of cascaded use, i.e. simultaneous production of heat and electricity in the year 2000 in Svartsengi and Nesjavellir is also shown.

Two areas that have been interpreted as ancient high temperature areas that are cooling down may be mentioned here, i. e. Leirá, Borgarfjörður where temperatures up to 170 °C have been logged at 2000 m depth, and Grímsnes (Fig. 1), where temperatures in excess of 200 °C have been logged, and it may still be considered as a high temperature area (Ármannsson, 2016). Carbon dioxide concentrations up to about 500 mg/L have been observed in the water phase from a borehole at Leirá (Ármannsson, 1981) and concentrations up to 2500 ppm in the water phase from a well at Hædarendi, Grímsnes (Sæmundsson et al., 2007). A large amount of free CO₂ is also emitted at Hædarendi, and carbon dioxide produced there is sufficient for all industrial and agricultural use in Iceland (Ármannsson, 2016).

4. Results of gas flux studies in Iceland

Reykjanes: Fridriksson et al. (2006) studied the natural gas flow from the Reykjanes geothermal area prior to the commissioning of the Reykjanes power plant, and their findings are summarized below.

Total discharge of CO₂ to the atmosphere at Reykjanes. Natural atmospheric emissions of CO₂ at Reykjanes take place via three general pathways; soil diffuse degassing, steam vent discharge and gas bubbling through steam heated pools. The combined CO₂ emission via these three pathways at Reykjanes is equal to 13.9 t d⁻¹ or 5060 metric t yr⁻¹. Most of this CO₂, by far (97.4%), is emitted through soil diffuse degassing, while only 1.7 and 0.9% are emitted through steam vents and fractures, and steam heated pools, respectively. It must be noted that the CO₂ flux by soil diffuse degassing was determined directly, whereas the CO₂ emissions from steam vents and steam heated pools were determined by indirect methods. The Reykjanes volcanic system has been dormant during the last 800 years or so, whereas geologic evidence indicates that episodes of volcanic activity occur with about 1000 year intervals (Sigurgeirsson, 2004). The relatively long repose period since the last volcanic episode at Reykjanes suggests that the present rate of CO₂ degassing may be at a minimum and it may have been significantly higher immediately after volcanic episodes with associated dike intrusions.

Several researchers (Favara et al., 2001; Werner et al., 2000; Sorey et al., 1998; Evans et al., 2002; Gerlach et al., 2001), indicate that soil diffuse degassing is generally a major, if not the dominating pathway of CO₂ release from geothermal systems (See Table 3), as appears to be the case at Reykjanes. Ármannsson et al. (2005) estimated that the maximum CO₂ emissions from all Icelandic geothermal systems were 1.3 × Mt yr⁻¹ based on geological observations. Earlier estimates of total CO₂ discharge from Icelandic geothermal systems range between 0.15 × Mt yr⁻¹ (Ármannsson, 1991) to 1 to 2 × 10 Mt yr⁻¹ (Arnórsson, 1991; Arnórsson and Gíslason, 1994; Óskarsson, 1996). The lower value (Ármannsson, 1991) refers to steam vent discharge only, whereas the higher values represent the estimated total release of CO₂ from Icelandic geothermal systems, including atmospheric emissions (via soil diffuse degassing, steam vents, and steam heated pools), as well as CO₂ discharge into groundwater.

Geologic controls of CO₂ emissions at Reykjanes. The spatial distribution of soil diffuse degassing, soil temperature and heat flow indicates a strong tectonic control of both diffuse CO₂ emissions and heat loss. Two well defined linear diffuse degassing and heat loss structures and two or possibly three smaller linear features are observed. The orientation of the diffuse degassing structures (DDSs) is in all cases between N-S and NNE-SSW (between 000° and 020°). The most active parts of the DDSs define a NW-SE trend. The orientation of the DDSs at Reykjanes geothermal area is consistent with the orientation of the right lateral strike-

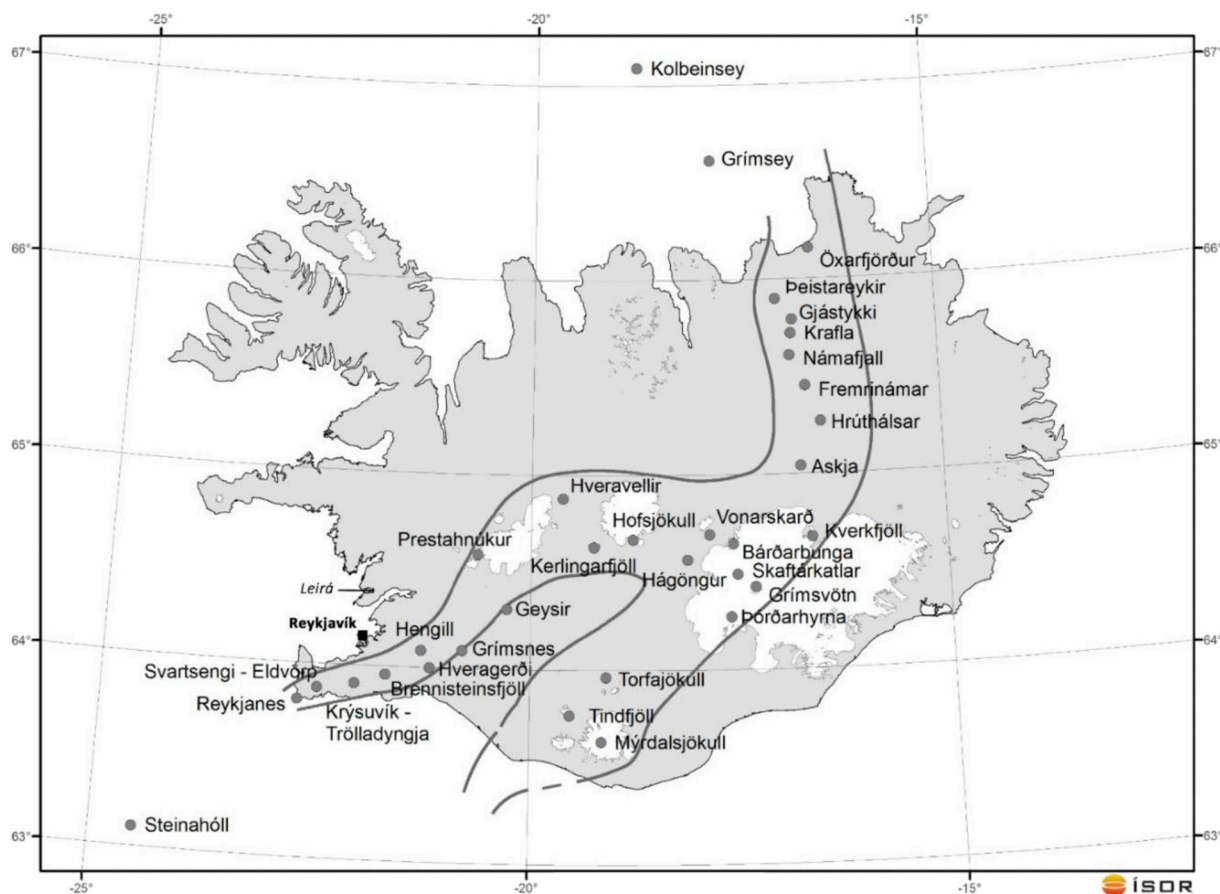


Fig. 1. High temperature areas in Iceland.

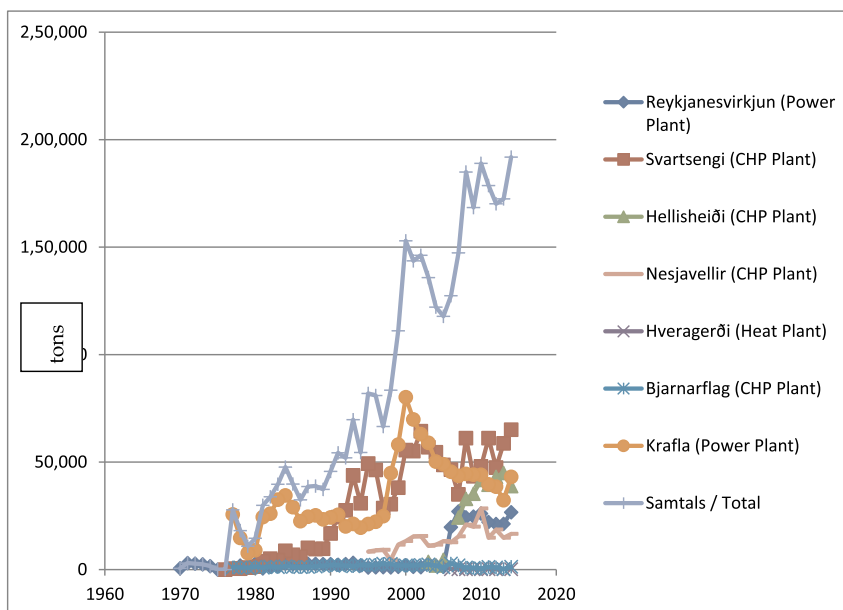


Fig. 2. Carbon dioxide emissions from geothermal activity in Iceland 1970–2014 (From <http://www.os.is/orkustofnun/gagnasofn/talnaefni>).

slip faults reported by Clifton and Schlichte (2003).

Different CO₂-emission/soil-temperature ratio of these two DDSs is probably a result of extensive steam condensation under one, whereas very little condensation seems to occur under the other.

This interpretation is supported by the large discrepancy between the observed heat flow through the surface at Reykjanes, 16.9 MW, and

the thermal energy released by condensing the 4200 t d⁻¹ of steam that must be associated with the CO₂ flux to the atmosphere observed, which is equal to 130 MW. The difference between these values is most probably a result of condensation of a large fraction of the steam (at least 87%) in the subsurface. The thermal energy from steam condensation at depth is likely transported laterally out of the system by

Table 4

CO₂ emissions per kWh from major geothermal power plants in Iceland. (Orkustofnun, 2016 (<http://www.os.is/orkustofnun/gagnasofn/talnaefni>); Ármannsson et al., 2005).

Power plant	Electricity generation only		Heat and electricity production CO ₂ (g kWh ⁻¹) 2000
	CO ₂ (g kWh ⁻¹) 2012	CO ₂ (g kWh ⁻¹) 2000	
Krafla	100	152	
Svartsengi	150	181	74
Reykjanes	18		
Hellisheiði	19		
Nesjavellir	25	26	10

groundwater flow. A portion of the ascending CO₂ must also be dissolved in the groundwater. The observed CO₂ emissions from the Reykjanes geothermal area must be taken to represent a minimum value for the release of CO₂ from the geothermal reservoir. The heat loss inferred from the observed CO₂ release, 130 MW, similarly represents a minimum value for the natural heat loss of the Reykjanes geothermal reservoir.

The extent and modes of surface geothermal manifestations at Reykjanes are probably sensitive to relatively small changes in the hydrological conditions in the groundwater aquifer. Although such changes are not likely to affect the rate of CO₂ release from the deep geothermal reservoir, they can change the relative proportions between discharge of CO₂ into the atmosphere and that into groundwater. Interactions between surface geothermal activity and groundwater will, therefore, tend to amplify temporal variability of surface geothermal activity and thus atmospheric CO₂ discharge from the Reykjanes geothermal system.

Óladóttir and Fridriksson (2015) described a follow-up of the gas flux measurements at Reykjanes, and their conclusions are as follows: The ten years of annual measurements of soil temperature and CO₂ flux in the Reykjanes geothermal area have shown an increased activity both in heat flow and in CO₂ flux. The CO₂ flux has increased from $13.5 \pm 1.7 \text{ t d}^{-1}$ in 2004 to $51.4 \pm 8.9 \text{ t d}^{-1}$ in 2013 according to the results of the soil measurements and there are no clear signs of stabilization in the CO₂ flux in Reykjanes yet. The distribution of CO₂ flux anomalies has changed greatly since 2004 but appears to be very similar in 2011, 2012 and 2013. The temperature anomalies also appear to have changed greatly since 2004 and to be rather stable during the last few years. The heat flow estimate indicates an almost tripled increase in heat flow between 2004 and 2012. The heat flow is derived from the soil temperature and the equation used is very sensitive to high temperature values. It is now known that high temperature values in the soil in Reykjanes vary, therefore reducing the value of the total heat flow estimate as a precise indicator of changes in the surface activity in the Reykjanes geothermal area. The changes in surface activity are expected to approach a steady state, and future measurements are an essential contribution to the understanding of the geothermal system. The CO₂ flux however increased from $51.4 \pm 8.9 \text{ t d}^{-1}$ in 2013 to $78.5 \pm 13.9 \text{ t d}^{-1}$ in 2014 and evidence of stabilization has not been observed yet (Óladóttir and Fridriksson., 2015).

Hengill: Hernández et al. (2012) studied degassing from the Hengill area and their findings are described below.

4.1. Tectonic control of the diffuse degassing structure

The spatial distributions of diffuse CO₂ and H₂S efflux soil temperature and heat flow suggest a strong structural control of both CO₂ and H₂S diffuse emissions and heat loss, indicating well-defined NS lineation diffuse degassing and heat loss structures. Diffuse CO₂ efflux and heat flow anomalies were identified along a NS trend parallel to the NS lines inferred by the seismic activity that occurred between 1994

and 2000 (Árnason and Magnússon, 2001; Björnsson et al., 2003). Jousset et al. (2011) interpreted these earthquakes as resulting from stress changes within the geothermal reservoir, where hot fluid rises in the crust above the heat source. According to Árnason et al. (2010), much of this trend is correlated with a low-rigidity, low-permeability, relatively shallow clay cap, with thermal manifestations occurring at gaps in this cap connecting the thermal manifestations through the base of the clay cap to the immediately underlying reservoir. Comparing the spatial distribution of diffuse CO₂ degassing and heat flow, it was observed that to the north of the DDS, elevated heat flow through soil coincides with DDS. However, the south and the center parts of the DDS do not coincide as clearly with the most prominent heat flow anomaly. A complementary interpretation is that steam condensation beneath DDS is not homogeneous, being weaker in the south, where the main surface thermal anomaly is found. Different CO₂ emission/soil temperature ratios have also been observed at other active volcanic-geothermal areas in Iceland (e.g., Reykjanes, Fridriksson et al. (2006)). The difference observed between heat flow through the surface at Hengill (11.5 MW) (Hernández et al., 2012) and the average thermal energy released by condensation of 40,154 t d⁻¹ of steam to the atmosphere (1237 MW), associated with the volcanic/hydrothermal CO₂ output of 453 t d⁻¹, also supports the observed CO₂ emission/soil temperature ratios. The difference between these values is most probably a result of condensation of a large fraction of the steam in the subsurface, as hypothesized for Reykjanes (Fridriksson et al., 2006). Thermal energy from steam condensation at depth might be transported laterally out of the system by groundwater flow. This hypothesis is supported by TES resistivity and seismic data, which strongly support the existence of a seismically active fault zone located between the Hveragerði and Hengill volcanic systems, acting as a fluid sink, probably due to lateral discharge towards the south (Björnsson et al., 2003).

4.2. Natural geothermal CO₂ emissions compared to emissions from power plants

A comparison of the natural gas emissions from the Hengill central volcano to the emissions from the geothermal power plants Nesjavellir and Hellisheiði, both located in the study area, has been made. In 2010, the Nesjavellir power plant released 30,727 t of CO₂ and 13,340 t of H₂S into the atmosphere (Reykjavík Energy, 2011), whereas the Hellisheiði power plant released 42,688 t of CO₂ and 9384 t of H₂S. The installed capacity at Nesjavellir is 120 MWe and 300 MWt (Reykjavík Energy, 2011), whereas at the Hellisheiði power plant, at the time of this study (phase 1), the installed capacity was 90 MWe, although this was increased to the present production capacity of 303 MWe and 133 MWt by 2011 (<https://www.or.is/en/projects/hellisheiði-geothermal-plant>). The volcanic/hydrothermal CO₂ output of the Hengill volcanic system of 453 t d⁻¹ amounts to an annual CO₂ output of 165,345 t yr⁻¹. The total CO₂ emission from the Reykjavík Energy power plants in the area amounts to 73,415 t yr⁻¹ or slightly less than half the natural emission in 2006. A similar ratio is observed at the Krafla geothermal field in NE Iceland where the natural emission of CO₂ of geothermal origin through diffuse degassing amounts to 84,000 t yr⁻¹ (Ármannsson et al., 2007). This compares to an annual CO₂ emission from the 60 MWe Krafla power plant, whose emissions are about 40,000 t yr⁻¹. The ratio between anthropogenic and natural CO₂ emissions from the Hengill system is more or less the same as that for the Krafla system, i.e., natural emissions amount to slightly more than twice the amount released from the power plants. The ratio of anthropogenic to natural gas emissions from the Reykjanes system is different from that from the Hengill and Krafla areas. Fridriksson et al. (2006) reported observed CO₂ emissions from the geothermal field of about 5100 t yr⁻¹ and they estimated the emissions from the 100-MWe power plant that was under construction at the time as 31,000 t yr⁻¹. After the commissioning of the power plant, the geothermal surface activity increased significantly (Fridriksson et al., 2010), and in 2010, the annual natural emission of

CO₂ via diffuse degassing at Reykjanes amounted to 12,660 t yr⁻¹ (Óladóttir and Snæbjörnsdóttir, 2011) and are still increasing (F. Óskarsson pers. com.), while the CO₂ emissions from the Reykjanes power plant amounted to 26,940 t yr⁻¹ (Óskarsson and Friðriksson, 2011). Thus, while the ratio of power plant emissions to diffuse degassing in Hengill and Krafla are both approximately 1:2, the ratio for Reykjanes is closer to 2:1. The estimated CO₂ output of 453 t d⁻¹ is in the same order of magnitude as estimations reported for other active volcanic areas (Brombach et al., 2001; Chiodini et al., 1996, 2001; 2007; Frondini et al., 2004; Hernández et al., 2001, 2003; Notsu et al., 2005; Pérez et al., 2004; Salazar et al., 2001). However, it should be noted that this is an underestimate of the total CO₂ discharge from the Hengill volcanic system because CO₂ dissolved by groundwater and CO₂ discharged through fumaroles and steam-heated mud pools have not been considered in this study. The absence of extensive surface manifestations in large parts of the productive geothermal reservoirs in the Hengill system, e.g., around the Hellisheidi power plant, suggests that considerable amounts of CO₂ from the reservoir may be dissolved in groundwater before it reaches the surface. On the other hand, the experience from Reykjanes (Friðriksson et al., 2006, 2010) suggests that emissions through steam vents and steam-heated mud pools are probably not as significant as diffuse degassing. In 2004, diffuse degassing constituted 97.5% of the total natural emission, while steam vents and mud pits emitted the remaining 2.5% (Friðriksson et al., 2006). In 2007, after the commissioning of the power plant had invigorated the surface activity at Reykjanes, diffuse degassing still constituted 90% of the total natural CO₂ emission from the field, whereas emission from steam vents and pits amounted to 10% of the total (Friðriksson et al., 2010). Diffuse degassing surveys at regular intervals over a period of several years will be an important geochemical tool to understand the system's behavior, especially concerning the consistency of emission rates and propagation or retreat of fumarolic areas. Such periodic studies are important to evaluate the effect of geothermal production on the surface activity, as has been done in Reykjanes (Friðriksson et al., 2010). It may also be pointed out that an estimation of CO₂ emissions from fumaroles in Icelandic geothermal areas (Ármannsson, 1991) showed them to be about 10% of total CO₂ emissions from these areas estimated by others (Arnórsson, 1991; Arnórsson and Gíslason, 1994; Óskarsson, 1996) and a similar proportion may be expected in most geothermal areas.

Krafla: Ármannsson et al. (2007) have studied the natural gas flux in the Krafla area, and their results are summarized below.

The total CO₂ flux from the areas studied in Leirhnjúkur and Mt. Krafla were 12 and 8 kt/yr, respectively. Subsequent measurements have not revealed a significant change (Kristinsson et al., 2014). The results illustrate a tectonic control over soil gas emissions in the slopes of Mt. Krafla. Two main trends are apparent, a NNE-SSW trend, parallel to the local normal faults, and a WNW-ESE trend. The relationship between soil gas emissions and structural geology is less obvious in Leirhnjúkur, possibly due to the small area of the flux measurement grid.

Using the graphical statistical method of Sinclair (1974) the mean flux of the geothermal population was estimated to be about 115 g/m²/d and it emanates from about 10% of the total area. Two background populations were identified, referred to as background and low background, 6 and 1.6 g/m²/d, respectively. They covered 80% and 10% of the total area, respectively. The total CO₂ flux from the eastern Krafla caldera is about 120 kt yr⁻¹, and about 70% of that is of geothermal origin. This can be considered as an upper limit to the CO₂ flux from Krafla as sampling was skewed towards areas with visible geothermal manifestations. As a result, the relative proportion of the geothermal population might be overestimated, but the mean flux from that population is considered realistic. Significant soil diffuse CO₂ degassing was found in two fumarole fields around the Víti crater lake and one area of a very limited extent in Leirbotnar, east of Hveragil, outside the two areas above.

The CO₂ concentration of cuttings from boreholes in Krafla ranges from 0.0 to 430 kg/m³. The CO₂ concentrations in the bedrock are high near the surface, but decrease steadily towards almost zero at a depth of about 1300 m below surface. The maximum CO₂ concentrations in bedrock are in some wells at the surface but in others at about 200 m depth. As the concentration of fixed CO₂ in the bedrock has reached zero at about 1300 m below surface it is possible to compute the total amount of CO₂ fixed in the bedrock per unit surface area by finite element integration over the CO₂ depth profile for each well. The fixed CO₂ is about 90 t/m² in wells 25 and 32 but the average for the 10 wells is about 70 t/m². If this is representative of the 20 km² eastern Krafla caldera, the total CO₂ fixed in bedrock there is of the order 1400 Mt. Significantly less CO₂ seems to be fixed in bedrock in the southern slopes of Mt. Krafla than in the bedrock west of the Hveragil.

Námafjall: CO₂ flux through soil has been measured on profiles in 2004, 2010 and 2013. In 2013 the mean flux in places of significant geothermal activity was 15.4 g/m²/d and negligible changes had been found since 2004 (Kristinsson et al., 2013a).

Þeistareykir: In 2012 CO₂ flux measurements were carried out in Þeistareykir, and a mean of 18.2 g/m²/d obtained for areas of significant geothermal activity. Earlier measurements had also revealed low flux values (Kristinsson et al., 2013b). Results of modeling studies on the area (Guðmundsson et al., 2008) suggest that fairly cool aquifers are found at relatively shallow levels across a large part of the area, probably causing carbonate deposition and thus weak CO₂ emissions through soil. A strong groundwater current close to the surface is also likely to dissolve the carbon dioxide and prevent its passage to the surface. An extensive survey in 2015 suggested that the CO₂ flow to the surface is extremely patchy but very high values were obtained locally. The total gas flux was calculated about 110 kt yr⁻¹ CO₂ from the whole area (Kristinsson et al., 2015).

Summary: Taking into account uncertainties in estimated areal extent and temporal variations a summary of carbon dioxide flux from the five areas that have been studied in detail is presented in Table 5.

5. Carbon dioxide fixed in Icelandic geothermal systems

To highlight similarities and differences between Hellisheidi, Krafla and Reykjanes, the average CO₂-depth profiles for the three systems are shown in Fig. 3. Since the surfaces of the areas are located at different altitudes, the CO₂-depth profiles are shown in terms of meters below the surface. The graph shows that in Hellisheidi there is almost as much fixed CO₂ as in Krafla while much less CO₂ is captured in the Reykjanes area. This is in agreement with the values for the average CO₂ content (kg/m³). The average CO₂-load is 65.7, 73.1 and 28.2 t/m² for Hellisheidi, Krafla and Reykjanes, respectively.

The total amount of CO₂ that is fixed in the crust of the geothermal systems can be roughly estimated by multiplying the average CO₂-load of the wells in given systems by the areal extent of the geothermal system. Pálmason et al. (1985) estimated the extent of the Reykjanes geothermal area to be 2 km² and Krafla 30 km². The determination of the extent of Hellisheidi is not as straightforward because the Hellisheidi high-temperature field is a subfield of the Hengill system, one of the most extensive geothermal areas in Iceland. A total area of around 110 km² is indicated by temperature distribution, surface and subsurface measurements (Gunnlaugsson and Gíslason, 2010). Since the

Table 5
CO₂ flux from five geothermal in Iceland.

Area	Gas flux g/m ² /d	Reference
Reykjanes	39	Óladóttir and Friðriksson (2015)
Hengill	5	Hernández et al. (2012)
Krafla	115	Ármannsson et al. (2007)
Námafjall	15	Kristinsson et al. (2013a)
Þeistareykir	18	Kristinsson et al. (2013b)

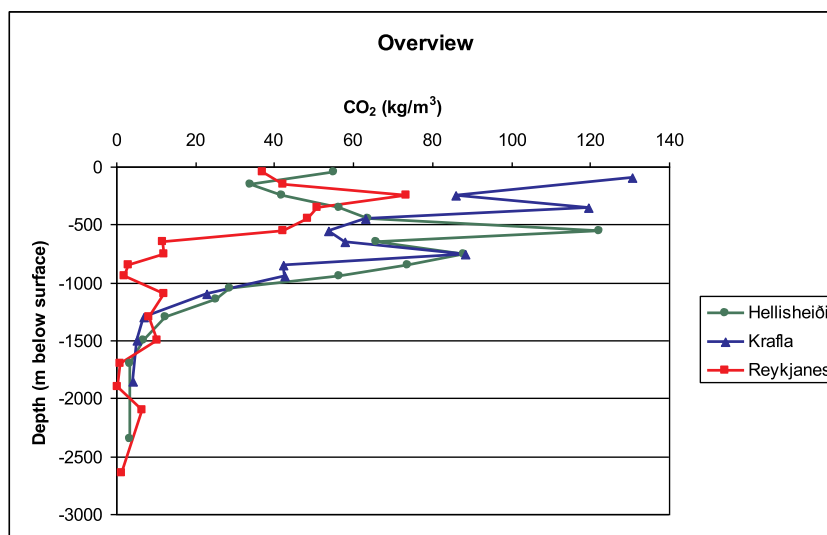


Fig. 3. Average CO₂-depth profile: comparison of the three areas (Wiese et al., 2008).

samples originate from the Hellisheiði subfield the average value can only be applied to this area extending to about 25 km² (estimate based on Björnsson et al., 2006).

The resulting values for the total amount of CO₂ fixed at Hellisheiði, Krafla, and Reykjanes are 1650 Mt, 2200 Mt, and 56 Mt, respectively. In Krafla alone the CO₂ amounts to about 1000 times the annual anthropogenic CO₂ emissions of Iceland (2.2 Mt in 2003; UNFCCC, 2005). That year the total greenhouse gas emissions were 3.9 Mt but had increased to 4.6 Mt in 2013 (Umhverfisstofnun, 2016: <http://www.umhverfisstofnun.is/einstaklingar/loftslagsbreytingar/losun-islands/>). The three high-temperature areas investigated represent less than one tenth of all high-temperature systems in Iceland regarding both surface area (533 km²; Pálmason et al., 1985) and the number of these areas (33; Ármannsson, 2016). Based on the speculative assumption that the CO₂ content of the three investigated systems is representative, the total carbon dioxide fixed in active high-temperature systems in Iceland amounts to 30–40 Gt of CO₂. If geothermal systems related to extinct central volcanoes are included in this estimate the total amount of CO₂ fixed in the Icelandic crust may be 10 to 15 times higher than this number (assuming that about 30–40 geothermal systems have been active in the volcanic zone throughout the geologic history of Iceland).

In order to evaluate the importance of calcite fixation in geothermal systems as a geochemical sink of CO₂ it is necessary to estimate the time it has taken the calcite to accumulate. Unfortunately, the ages of the geothermal systems considered in this study are poorly constrained; age estimates for the Hellisheiði geothermal system range between 70,000 and 400,000 years (Franzson et al., 2005). For Krafla, K. Saemundsson gives a range of 110,000 to 290,000 years (K. Saemundsson, pers. comm. March 2016, Saemundsson, 1991, Saemundsson et al., 2000, Björnsson et al., 2007) and Reykjanes is estimated to be between 10,000 and 100,000 years old (H. Franzson, pers. comm. March 2016, Franzson (2007)).

These age estimates, estimated areal extents of the systems, and the average CO₂-load of the crust in the three geothermal systems (see Table 6) were used to evaluate the calcite fixation rate in these systems. Accordingly, for Hellisheiði the estimated CO₂ fixation rate in calcite is 4100 to 23,500 t/yr and for Krafla and Reykjanes the estimated CO₂ fixation rates are 7500 to 20,000 and 560 to 5600 t/yr, respectively. These values can be compared to natural atmospheric CO₂ emissions observed from these systems. In 2004 the atmospheric emissions from Reykjanes were 5000 t/yr⁻¹ (Fridriksson et al., 2006), and preliminary data analysis indicates that geothermal soil diffuse degassing from Krafla is of the order 100,000 to 150,000 t/yr (Ármannsson et al.,

Table 6

CO₂ fixation rate and CO₂ emissions.

	Area (km ²)	Fixed CO ₂ (kg/m ²)	Age (yr)	Fixation Rate (kg/m ² yr ⁻¹)	CO ₂ Emissions (kg/m ² yr ⁻¹)
Hellisheiði	25 ⁽¹⁾	65700	70.000–400.000 ⁽³⁾	0.2–0.9	
Krafla	30 ⁽⁴⁾	73100	110.000–290.000 ⁽⁵⁾	0.3–0.7	4.25 ⁽²⁾
Reykjanes	2 ⁽⁴⁾	28200	10.000–100.000 ⁽⁶⁾	0.3–2.8	2.5 ⁽⁷⁾
Iceland ^a	533 ⁽⁸⁾	55667 ⁽⁹⁾	100.000–1.000.000 ⁽¹⁰⁾	0.1–0.6	0.2 – 3.8 ⁽¹¹⁾

(1) Björnsson et al. (2006).

(2) Preliminary data analysis of CO₂ flux measurements 2004 to 2006 (Ármannsson et al., 2007).

(3) Franzson et al. (2005).

(4) Pálmason et al. (1985).

(5) Saemundsson, K. personal communication March 2016, Saemundsson (1991), Saemundsson et al., 2000, Björnsson et al., 2007).

(6) Franzson, H., personal communication March 2016., Franzson 2007.

(7) Fridriksson et al. (2006).

(8) Wiese et al. (2008).

(9) Average of Hellisheiði, Krafla and Reykjanes.

(10) Arnórsson (1995).

(11) Calculation based on data from Ármannsson et al. (2005).

^a Total numbers for Iceland are speculated.

2007). Comparison of the CO₂ fixation rate determined in this study and the observed atmospheric emissions from Reykjanes and Krafla shows that the magnitude of the CO₂ fixation is somewhere between 7.5% of the atmospheric emissions to being equal to them. These results illustrate that calcite fixation plays a considerable role in the CO₂-budget of geothermal systems, even if the lower estimates for the CO₂ fixation were true. In Reykjanes Fridriksson et al. (2016) found that some gas samples seem to be depleted in CO₂ relative to He, and plot significantly below the atmospheric and geothermal CO₂ mixing line, with a CO₂/He ratio of ~5000 compared to 25000 for well samples. Interpreting this as an indication of CO₂ depletion it corresponds to 80% CO₂ loss from the geothermal gas.

6. CO₂ in recently drilled hot deep wells

Until recently the CO₂ concentrations in fluids from most wells have followed the temperature as is to be expected (Arnórsson and Gunnlaugsson, 1985). Exceptions were very high concentrations during

Table 7
CO₂ in steam at 1 bar in some wells at Krafla and Þeistareykir.

Well	Year	Type	Inflow depth (m)	Temperature (°C)	CO ₂ (ppm)
KJ-15	1980	Affected by magmatic gas	1500	330	67836
KG-24	2006	Upper part „cool“ well	700	210	978
KJ-34	2006	Conventional deep well	1500	320	15961
IDDP-1	2011	Recent „hot“ well	2000	450	760
THG-04	2007	Recent „hot“ well	1900	330	719

volcanic activity in Krafla and occasionally upon drawdown, e.g. in Svartsengi. Recently much lower CO₂ concentrations have been observed in fluids from deep wells at temperatures in excess of 320–330 °C in Krafla, Þeistareykir and Námafjall. Examples are shown in Table 7.

Thus it seems that if deeper and hotter wells will be more common in the future that the problem of gas emissions may be reduced.

7. Summary and conclusions

It is surmised that in Reykjanes and Svartsengi there has probably been a considerable increase in CO₂ emissions after the start of production, and that probably about 80% of the emissions would be counted as an addition. In Hengill, Námafjall and Krafla it would however seem that the increase is very small, and only a negligible amount would count as added emissions. This shows that it is extremely important to establish firmly the background emissions from geothermal areas by determining the amount of gas emitted from soil, steam vents, and if possible water pools before production starts, and monitor these parameters as well during production so that both possible increases in such emissions and emissions due to production can be evaluated and reported.

The potentially productive high-temperature areas in Iceland are magmatic in origin, except possibly Óxarfjörður. The CO₂ concentrations of their fluids depend on equilibrium between carbonates in the rock and the fluid, except in special cases such as the Krafla fires 1975–1984 during which excess CO₂ invaded the geothermal system. The CO₂ concentration may also rise upon increased boiling in a geothermal system usually as a result of increased production, e.g. Svartsengi in the 1990s and more recently Reykjanes. In such cases there is usually a sharp concentration increase at the beginning which gradually slows down and eventually decreases to former levels. CO₂ concentrations may be very high in peripheral fluids and in fluids from old high-temperature systems that are cooling down such as Leirá, Borgarfjörður, and Grímsnes, but such areas are not likely to become utilized for power production, although Grímsnes is used for CO₂ production.

Recently results of deep drilling into relatively high temperature production zones indicate that at temperatures in excess of 320–340 °C the CO₂ concentration of the fluids is relatively low and decreases with temperature, e.g. a very low CO₂ concentration is observed in fluids from IDDP-1, Krafla at a temperature of 450 °C.

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