

RESEARCH LETTER

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Key Points:

- Subglacial volcanoes are underrepresented in terms of gas monitoring, but we show that they can be major emitters of CO₂.
- Katla volcano is found to be one of largest volcanic sources of CO₂ on the planet, contributing up to 4% of all nonerupting volcanoes.
- High-precision airborne measurements combined with atmospheric modeling are a powerful method to monitor poorly accessible volcanoes.

Supporting Information:

- Supporting Information S1

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Globally Significant CO₂ Emissions From Katla, a Subglacial Volcano in Iceland

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Abstract Volcanoes are a key natural source of CO₂, but global estimates of volcanic CO₂ flux are predominantly based on measurements from a fraction of world's actively degassing volcanoes. We combine high-precision airborne measurements from 2016 and 2017 with atmospheric dispersion modeling to quantify CO₂ emissions from Katla, a major subglacial volcanic caldera in Iceland that last erupted 100 years ago but has been undergoing significant unrest in recent decades. Katla's sustained CO₂ flux, 12–24 kt/d, is up to an order of magnitude greater than previous estimates of total CO₂ release from Iceland's natural sources. Katla is one of the largest volcanic sources of CO₂ on the planet, contributing up to 4% of global emissions from nonerupting volcanoes. Further measurements on subglacial volcanoes worldwide are urgently required to establish if Katla is exceptional, or if there is a significant previously unrecognized contribution to global CO₂ emissions from natural sources.

Plain Language Summary We discovered that Katla volcano in Iceland is a globally important source of atmospheric carbon dioxide (CO₂) in spite of being previously assumed to be a minor gas emitter. Volcanoes are a key natural source of atmospheric CO₂, but estimates of the total global amount of CO₂ that volcanoes emit are based on only a small number of active volcanoes. Very few volcanoes that are covered by glacial ice have been measured for gas emissions, probably because they tend to be difficult to access and often do not have obvious degassing vents. Through high-precision airborne measurements and atmospheric dispersion modeling, we show that Katla, a highly hazardous subglacial volcano that last erupted 100 years ago, is one of the largest volcanic sources of CO₂ on Earth, releasing up to 4% of total global volcanic emissions. This is significant in a context of a growing awareness that natural CO₂ sources have to be more accurately quantified in climate assessments, and we recommend urgent investigations of other subglacial volcanoes worldwide.

1. Introduction

Volcanoes are one of the most important natural sources of carbon dioxide (CO₂), but empirical measurements are available for only ~20% of major volcanic gas emission sources (reviewed in Burton et al., 2013). Extrapolations of these measurements give an estimated a global subaerial geological emission rate of ~1,500-kt/d CO₂ (Burton et al., 2013), which is ~2% of the anthropogenic emission rate of ~96,000 kt/d (Friedlingstein et al., 2010). Updated measurements of degassing from arc volcanoes, for example, Aiuppa et al. (2017), demonstrate that there are still large uncertainties. The quantification of CO₂ emissions from previously unmeasured volcanic sources is therefore critical. While subglacial volcanoes are numerous, they are grossly underrepresented in terms of volcanic gas measurements (3 out of the 33 volcanoes reviewed in Burton et al., 2013), potentially because they often lack a visible gas plume and/or are more difficult to access. In Iceland, gas measurements of CO₂ fluxes from the 32 active volcanic systems are sparse, and only 2 out of its 16 subglacial volcanoes (Grímsvötn and Eyjafjallajökull) have been measured (Table 1). The reported fluxes CO₂ from nonerupting volcanoes are relatively low, with a maximum of 0.5 kt/d from Grímsvötn (Ágústsdóttir & Brantley, 1994). Due to the low number of available measurements, the estimates of total volcanic CO₂ flux in Iceland, 2.7–5.8 kt/d (Arnórsson & Gislason, 1994; Hernández et al., 2012;

Table 1

CO₂ Flux (kt/d With Standard Error) From Katla Volcano Compared With Other Volcanoes in Iceland (kt/d, Minimum and Maximum Values) for Which Data Have Been Published

| Volcano | Date (flight number for Katla) | CO ₂ flux (kt/d) | Approach | Methods | | | |
|--|--------------------------------|-----------------------------|--|-------------------------|---------------------------|---|-------------------------|
| | | | | Number of flight tracks | CO ₂ max (ppm) | Katla only | |
| | | | | | | Altitude of CO ₂ plume (m above sea level) | Flux calculation method |
| Katla, western flank | 18 Oct 2016 (B987) | 19.6 ± 3.2 | Airborne direct observations | 12 | 432 | 100–600 | IDW |
| | | 15 | Simulation | | | | SMF |
| | 20 Oct 2016 (B989) | 14.6 ± 3.2 | Airborne direct observations | 13 | 413 | 840–1,200 | IDW |
| | | 11.9 ± 5.4 | Simulation | | | | Gaussian |
| Katla, central caldera | 04 Oct 2017 (C060) | 5–10 | Airborne direct observations | 3 | 432 | 890–970 | SMF |
| | | 12.8 ± 1.3 | Simulation | | | | IDW |
| | | 5–10 | Airborne direct observations | 7 | 415 | 380–650 | SMF |
| | | 0.53 | Simulation | | | | IDW |
| Grímsvötn (Ágústsdóttir & Brantley, 1994) | 1954–1991 | 0.007–0.070 | Subglacial melt water from the caldera | | | | |
| Eyjafjallajökull (Gíslason, 2000) | 2000 | 0.012–0.019 | Subglacial melt water from the caldera | | | | |
| Hekla (Gíslason et al., 1992) | 1988–1991 | 0.19 | Gas dissolved in groundwater | | | | |
| Hekla (Ilyinskaya et al., 2015) | 2012–2013 | 0.044 | Diffuse soil emissions | | | | |
| Reykjanes (Fridriksson et al., 2006, Fridriksson et al., 2010) | 2004–2009 | 0.012–0.019 | Diffuse soil emissions | | | | |
| Hengill (Hernández et al., 2012) | 2006 | 0.45 | Diffuse soil emissions | | | | |
| Krafla (Ármannsson et al., 2007) | 2004–2006 | 0.23 | Diffuse soil emissions | | | | |

Note. For Katla airborne measurements, the table shows the number of flight tracks that passed through the plume, the max CO₂ concentration measured on each flight, and the altitude at which the CO₂ plume was found. Methods used for Katla CO₂ flux calculations: IDW, inverse distance weighting; Gaussian, fitting of a Gaussian plume dispersion model; SMF, specified mass flux.

Pálmasón et al., 1985), are poorly constrained and are likely too low (Ármannsson et al., 2005). The CO₂ flux from Grímsvötn and Eyjafjallajökull were estimated by analyzing gas content dissolved in melt water accumulating under the ice that likely underestimates the flux as CO₂ degasses very rapidly when the water is depressurized. Our study is the first to report the CO₂ flux from a subglacial volcano in Iceland by measuring the gas directly in the atmosphere.

Measurements of gas emissions from subglacial volcanic systems are important for understanding the underlying magma systems and, subsequently, for forecasting their eruptions, which are typically highly hazardous due to the generation of ash and jökulhlaups (flash floods of glacial melt water). Recent studies across different tectonic and geographical settings have demonstrated that increases in CO₂ output can precede eruptions by months to years, for example, at Redoubt in the Aleutians (Werner et al., 2012), Kilauea in Hawaii (Poland et al., 2012), and Villarica in Chile (Aiuppa et al., 2017) but it is not yet known if this applies to any of the Icelandic volcanoes.

1.1. Katla Volcanic System

The subglacial Katla volcanic system is one of the largest and most active ones in Iceland and has erupted 1–3 times per century since the settlement of Iceland 1,100 years ago (Larsen, 2000), and up to 6 times per century in prehistoric times (Óladóttir et al., 2008). The current repose period is the longest one on record, with the last confirmed eruption in 1918 C.E. Katla system consists of a central volcano (max altitude 1,500 m above sea level [asl]) and 80-km long fissure system. The central volcano is partially covered by the vast 590-km² Mýrdalsjökull glacier, which is on average ~200 m thick, reaching 700-m thickness in

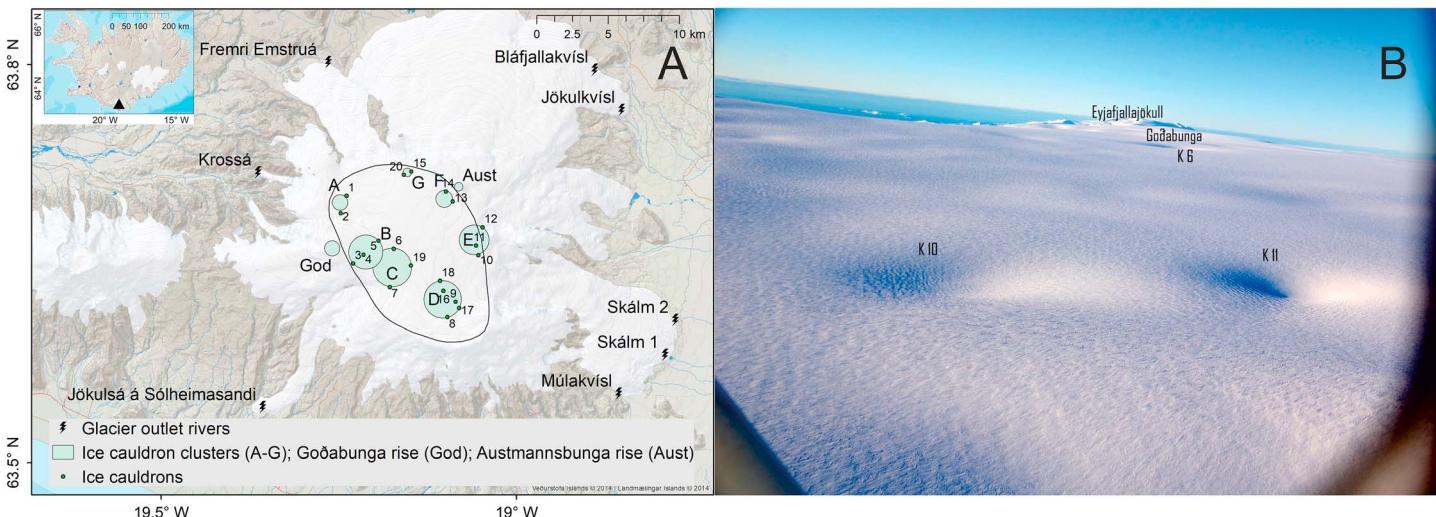


Figure 1. (a) Map and (b) photograph of Katla. The map shows the outlines of the subglacial caldera and locations of glacier river outlets ($n = 8$), ice cauldrons ($n = 20$), Goðabunga rise (God), and Austmannsbunga rise (Aust). For model simulations of the gas source, the 20 ice cauldrons were combined into seven clusters (A–G). The photograph, taken in November 2017, shows ice cauldrons 10 and 11 (K10 and K11, respectively) and Goðabunga rise. The cauldrons are several hundreds of meters in diameter. The summit of the neighboring Eyjafjallajökull volcano is seen behind the Katla caldera.

places. The central volcano contains a large, ice-filled caldera (110 km^2 , Figure 1). The eruptions within the glaciated part are typically accompanied by tephra generation (bulk volume $0.02\text{--}2 \text{ km}^3$) and jökulhlaups due to the magma-ice interaction (Larsen, 2000). The fissure swarm has produced large effusive basaltic eruptions with lava volumes $\geq 18 \text{ km}^3$ (Thordarson et al., 2003). The size and proximity to populations of Katla mean that the next eruption will likely have major local and possibly regional impacts, whether it occurs within the glaciated or nonglaciated part of the system. Disturbance to international aviation by ash is likely, even if the eruption is small in size (Biass et al., 2014).

Katla has had recurring geophysical unrest (seismicity and ground deformation), but the presence of glacial ice makes the subsurface signals difficult to interpret. Previous studies have disagreed on whether unrest in different parts of the system is caused by movements of magma (e.g., Soosalu et al., 2006; Sturkell et al., 2008), or movements of glacial ice and its seasonal changes (e.g., Jónsdóttir et al., 2009; Spaans et al., 2015). Katla has an annual average of ~ 300 earthquakes (Icelandic Met Office monitoring data) and periodic escalations of up to a few thousand earthquakes. The majority of the earthquakes are at 0- to 5-km depth and < 2.5 in magnitude, with rarer occurrences of deeper (up to 20-km depth) and larger events (magnitude ≥ 4). There are two main areas of geophysical unrest—within the caldera, and near the Goðabunga rise on the western part of the central volcano (e.g., Jónsdóttir et al., 2009). The largest unrest periods since the last confirmed eruption have occurred in 1955, 1999, and 2011 C.E. These periods had increased seismicity for months to years, increased geothermal activity, and significant jökulhlaups that caused damage to infrastructure (Sturkell et al., 2008). It has not been conclusively shown whether these episodes were associated with small subglacial eruptions.

Katla has no obvious degassing vents or areas, or visible gas plumes. Presence of subglacial activity is manifested by 20 ice cauldrons, which are 10- to 50-m deep depressions in the glacier surface (Figure 1) caused by geothermal melting of the glacier base. Geothermal melt water escapes through the glacier drainage systems and is periodically flushed out from the outlet rivers (Figure 1). The number, size, and shape of Katla's ice cauldrons and the activity of the outlet rivers change over time as the subglacial system is highly dynamic (Guðmundsson et al., 2007), likely influenced both by the state of the volcanic system, and short- and long-term variations in weather and climate. The smell of hydrogen sulfide (H_2S) is commonly reported near the outlet rivers, in particular during major and minor jökulhlaups (Bergsson, 2016). Conversely, there are no known reports of visible gas plumes or gas smell in the vicinity of the ice cauldrons. A DOAS UV spectrometer installed on the flanks of Katla since July 2017 has never detected sulfur dioxide (SO_2) (Icelandic Met Office monitoring data).

The only eruption of Katla where gas release has been estimated using the petrological method is the Eldgjá flood basalt eruption 934–40 C.E. (Thordarson et al., 2003). Its current gas emission rate has not been

quantified. Here we measured Katla's gas emissions from an aircraft in October 2016 and October 2017. This work builds on previous airborne measurements of CO₂-rich plumes in other countries using in situ sensors (Delgado et al., 1998; Doukas & McGee, 2007; Gerlach et al., 1999, 1997; Werner et al., 2006, 2008, 2012, 2013) and serves as a proof-of-concept for monitoring gas emissions from other Icelandic volcanic systems.

2. Methods

2.1. Airborne Observations

The airborne observations were made using the atmospheric research aircraft (a highly modified BAE-146 aircraft) of the Facility for Airborne Atmospheric Measurements (<http://www.faam.ac.uk>). Details about the instrumentation are in Text S1 in the supporting information. Flight paths were selected based on the prevalent wind direction in order to obtain downwind measurements of active volcanoes. Low-altitude cloud distribution and topography influenced the flight path planning for safety reasons. No flights traversing the subglacial caldera were possible in 2016 or 2017 due to cloud cover over the glacier. The full tracks of the flights reported in this paper are shown in Figure S1 in the supporting information.

2.2. Gas Source Modeling

In order to identify the source of the excess CO₂, we applied two approaches. The first was to use back-trajectories based on simple, low-resolution forecast wind fields; we used the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) Lagrangian dispersion model driven by GFS forecast winds (full details about the model in Text S1). The second involved simulating the effects of a variety of plausible sources within a very high resolution numerical weather prediction model (Weather Research and Forecasting model [WRF]; full details about the model Text S1) and comparing the distribution of dispersed gases within the model with the observations. HYSPLIT was run from numerous measurement points along the aircraft track for 12 hr back in time in order to determine which trajectories coincided with likely sources. The relatively long run time was chosen so that there were no initial constraints on the gas source within Iceland (e.g., other volcanic systems and anthropogenic activities). Results of HYSPLIT are included in supporting information (Figure S2). For the sources in the WRF simulations we initially used the 32 volcanic systems in Iceland (Figure S1) and ran the WRF model with CO₂ as a passive tracer. This confirmed unequivocally that the source was in the region of Katla, leading us to make further measurement flights in 2017, and more detailed simulations of the Katla region in order to identify the source of the gas. For these simulations, we specified as potential sources 8 glacier outlet rivers from Katla, 20 ice cauldrons within the caldera that were combined into 7 cauldron clusters (A–G), and Goðabunga rise (a location of long-term seismic activity on the volcano's west flank), giving a total of 16 sources (Figure 1). All sources were treated as a point release of a dense gas with a specified emission rate (full details in Text S1). For most of the simulated cases, HYSPLIT and WRF indicated the same source locations; notable differences are described in section 3.

2.3. Gas Emission Rate Calculations

As the exact location and number of the degassing sources within the large glacier (590 km²) overlying Katla were unknown, the calculation of the CO₂ emission rate ("flux") presented a challenge not previously reported in studies using airborne measurements. We calculated the CO₂ flux using two independent methods, direct calculations and model simulations. The model simulations provided an independent means of mass flux estimation and hence a corroboration of the principal findings of the paper.

The first method was a direct calculation of the measured mass flux by integration of interpolations of the measured wind and CO₂ concentration fields (we used two different interpolation techniques). The interpolation techniques were inverse distance weighting (IDW in Table 1) for all of the flights and fitting of a Gaussian plume dispersion model (Gaussian in Table 1). The Gaussian method provided an independent flux estimate in addition to IDW. Several restrictions on its use (the requirement for a Gaussian plume, the need for wind speeds above 5 m/s, and the wind direction and flight track alignment to be perpendicular) meant that the Gaussian method could only be used for flight B989 (Table 1). It is included here for completeness. See Text S1 for further details about both interpolation techniques.

Motivated by the large emission rates given by IDW and Gaussian calculations (11–20 kt/d of CO₂, Table 1), we designed the second method of estimating emission rates using a state-of-the-art numerical model, WRF (the

simulations described in section 2.2). WRF is able to resolve the complex, unsteady flows associated with such a topographically complex region. Coupled with dense gas dynamics, WRF is essential here for effective source identification. Additionally, the use of WRF enabled various emission scenarios to be tested, adjusting the source strength to maximize correlation between observed and modeled CO₂ concentrations along the aircraft tracks (specified mass flux in Table 1). See Text S1 for further details.

Agreement between the different methods in this challenging degassing scenario provided additional confidence that assumptions concerning the dispersion mechanisms were sound and that the measurements were representative of the gas plumes.

3. Results and Discussion

3.1. Katla as a Source of Elevated CO₂

Gas plumes of elevated CO₂ were detected on three airborne measurement campaigns at Katla, on 18 October 2016 (flight number B987), 20 October 2016 (B989), and 4 October 2017 (C060). Background concentrations of CO₂ were around 400 ppm. CO₂ concentrations exceeding background levels up to 32 ppm were detected in the immediate vicinity of Katla (Figures 2a, 2c, and 2e) and up to 8–15 ppm in excess of background ~80 km to the east of it (B987 only, Figure 2e). Significant anthropogenic sources of CO₂ are highly unlikely upwind of the areas where the elevated concentrations were observed. H₂S smell was noticed in the aircraft cabin on several flights, both to the south and north of the glacier, but instrumental measurements were not obtained (see Text S1 for further details). SO₂ was below the 3-sigma detection limit of the fluorescence photometer of 1.5 ppb for 1-s measurements. The CO₂/CH₄ ratio in the gas plumes was ~200 (r^2 between 0.81 and 0.99, Figure 2); this gas composition suggests an interaction with a geothermal system in the roots of the subglacial caldera (Chiodini, 2009). This CO₂/CH₄ ratio is very similar to the ratios measured in fumarole direct samples from other Icelandic volcanoes that are primarily ice-capped (Kverkfjöll: 200, Grímsvötn, 300–350; Icelandic Met Office monitoring data). We first describe and discuss the results of 20 October 2016 (flight B989) and 4 October 2017 (C060), followed by 18 October 2016 (B987), which had a more complex gas dispersal pattern than the first two.

On 20 October 2016 (B989) the airborne measurements detected a well-defined CO₂ plume with maximum concentration of 413 ppm (~13 ppm above background) to the north of the Katla glacier at an altitude between 840 and 1,200 m asl (Figure 2a). The edge of the glacier is at ~500 m asl, and it then rises fairly steeply towards the middle of the caldera, the floor of which is at ~1,400–1,500 m asl. Model simulations emitting 5–10 kt/d of CO₂ identified that the likely source of excess CO₂ was on the western flank of Katla volcano where two outlet rivers (Fremri-Emstrúá and Krossá, ~500 m asl) and Goðabunga rise (1,500 m asl) are located. Ice cauldron cluster A (~1,500 m asl) on the western edge of the caldera is also a possible source (Figure 2b). We consider that the outlet rivers are possible sources of the CO₂ even when the flight tracks passed above the rivers. The CO₂ concentrations of up to ~30 ppm above background represent a small fraction of the background air and are unlikely to restrict the vertical motion associated with very complex underlying topography (Figure 3).

On 4 October 2017 (C060) elevated CO₂ concentrations were measured in two locations, as two separate and well-defined gas plumes. The first gas plume was to the northwest of Katla (up to 432 ppm, 890–970 m asl, Figure 2c) and could be reproduced by the model when 5–10 kt/d of CO₂ was released from Fremri-Emstrúá River on the western flank of Katla (Figure 2d), which is in good agreement with B989. The second plume was to the southeast of Katla (up to 415 ppm, 380–650 m asl, Figure 2c) and was reproduced with 5- to 10-kt/d emission rate when the gas was emitted from ice cauldron clusters E, F, or G within the caldera (Figure 2d). Cauldron cluster E contains two of the currently most active ice cauldrons (Guðmundsson et al., 2007), nr 10 and 11. This source area also includes a nunatak (elevated bedrock exposed within a glacier) named Austmannsbunga, ~1,400 m asl, a location of frequent current seismic unrest and surface deformation (Icelandic Met Office monitoring data).

On 18 October 2016 (B987) elevated CO₂ concentrations (up to 432 ppm) were measured immediately to the southeast of Katla at altitude 100–600 m asl (Figure 2e), at a similar location and altitude to that detected by C060. Elevated CO₂ (up to 409 ppm) was also measured in the coastal regions ~80 km to the east of Katla at altitudes between 200 and 1,600 m asl, with the highest values (408–409 ppm) between 210 and 540 m asl

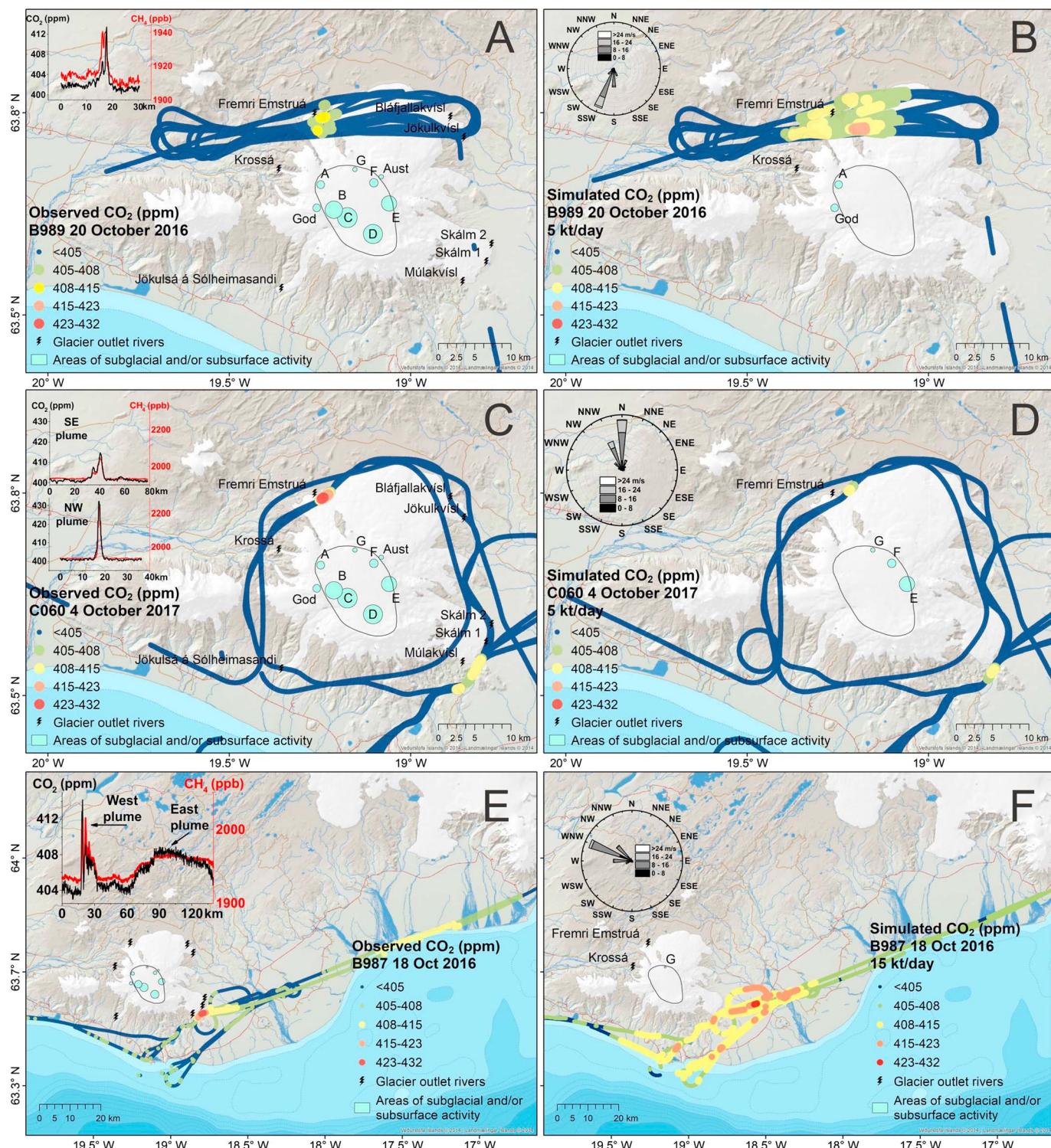


Figure 2. Observed and simulated CO₂ plumes from Katla volcano on 18 and 20 October 2016 and 4 October 2017. The panels show the wind direction and speed measured during each flight and along-track points of CO₂ and CH₄ concentrations. (a) Observed CO₂ peak on 20 Oct 2016 (flight B989). (b) The best-fit simulated sources for the CO₂ peak observed on 20 Oct 2016: rivers Fremri Emstruá and Krossá, ice cauldron cluster A, and Goðabunda rise. The figure shows simulation of 5 kt/d of CO₂, but good agreement was also reached with 10 kt/d. (c) Two observed CO₂ peaks on 4 Oct 2017 (flight C060), to the northwest (NW) and southeast (SE) of the caldera. (d) The best-fit simulated sources for the CO₂ peaks observed on 4 Oct 2017: river Fremri Emstruá and ice cauldron clusters E, F and G. The figure shows simulation of 5 kt/d of CO₂, but good agreement was also reached with 10 kt/d. (e) Two observed CO₂ peaks on 18 Oct 2016 (flight B987), to the west and east of the caldera. (f) The best-fit simulated sources for the CO₂ peaks observed on 18 Oct 2016—rivers Fremri Emstruá and Krossá and ice cauldron cluster G. Both of the observed CO₂ peaks can be traced to Katla when CO₂ is simulated as a dense gas with emission rate of 15 kt/d.

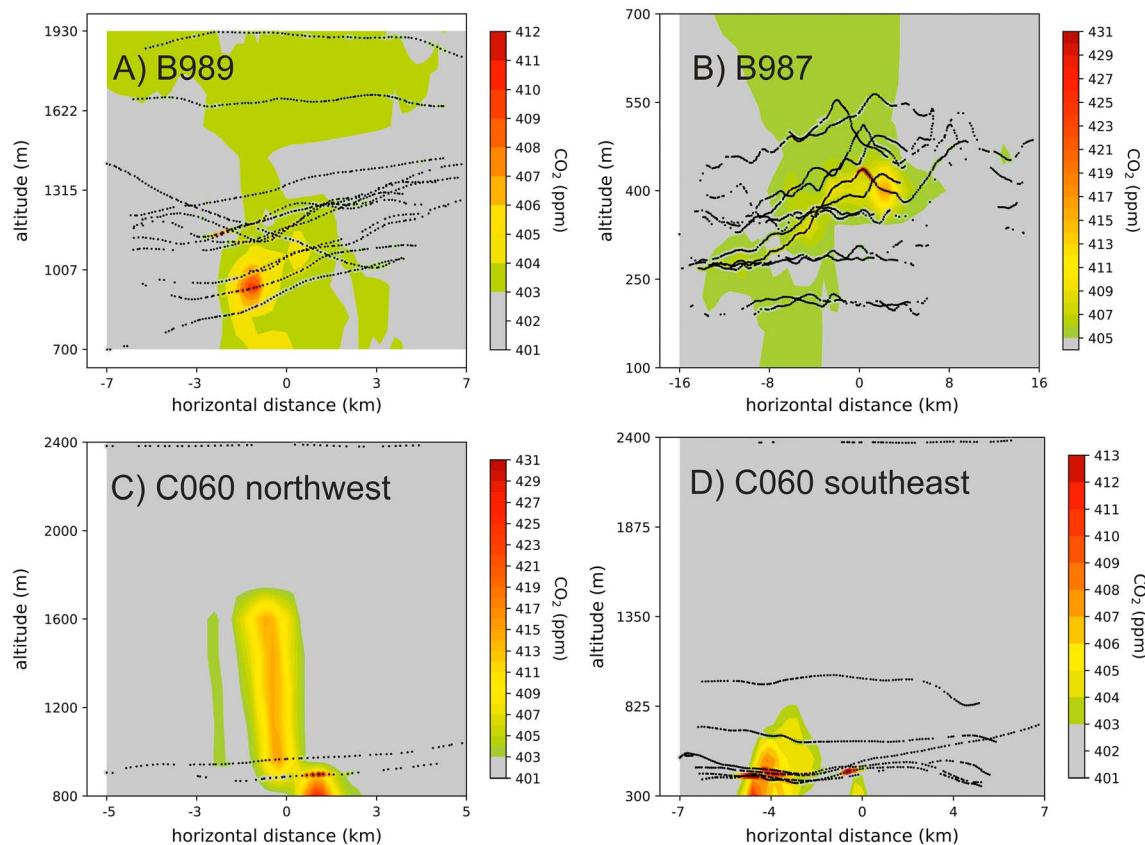


Figure 3. Vertical cross section through the interpolated CO₂ gas plumes from flights (a) B989 (20 October 2016); (b) B987 (18 October 2016), and C060 (4 October 2017), which intersected two gas plumes in 2 locations; (c) to the northwest of Katla; and from (d) to the southeast of Katla. The black dots show the measurement locations on the flight tracks. The x axis represents the horizontal distance from the center of the plume (the location with the highest measured gas concentration). The flight paths close to ground level (<1,500 m above sea level) were influenced by the highly irregular mountainous topography.

(Figure 2e). Dense gas simulations, emitting 15 kt/d of CO₂, were able to trace the gas observed to the east of Katla when Fremri-Emstrúá and Krossá Rivers were considered as sources (Figure 2f), with the gas flowing through and accumulating in valleys. Ice cauldron cluster G was also a possible source (Figure 2f). However the simulated concentrations were much lower (of the order of 0.1 ppm above background) than observed. This may be caused by gas accumulating over a longer time period than the duration of the simulation, or the existence of a possible extra source not accounted for here. Flight B987 is a notable example of the complexity (fine-scale variability and unsteadiness) of the air flow around a volcanic edifice. Simple models (e.g., HYSPLIT) cannot be used for source identification in such circumstances. Further observations coupled with model simulations would be required to understand scenarios such as B987 better.

The relatively well-defined structure of the gas plumes (Figure 3) suggests that the degassing source(s) within the Katla volcanic system are focused rather than diffuse, and we have made used model simulations to show that these sources could be located on the western flank of Katla, and within the central caldera (Figure 2). However, more detailed observations, ground- and/or airborne, will be needed to determine the location(s) of the gas source with more accuracy. It is likely that the gas source location(s) are dynamic in this subglacial volcanic environment and change over time.

3.2. CO₂ Emission Rate From Katla

Emission rate calculations (section 2.3) showed that a source associated with the western side of Katla (see Figure 2 for likely source locations) was emitting 11.9–19.6 kt/d of CO₂ in October 2016 and 12.8 kt/d in October 2017 (Table 1). In 2017, we were able to identify a second source of CO₂ (Table 1) most likely within the caldera, emitting 11.4 kt/d (best-fit sources ice cauldron clusters E, F and G in Figure 2d), although the low

number of flight passes through this plume gives a lower confidence in the flux calculation than for the other flights (Figure 3 and Table 1). It is possible that there was no significant degassing from within the caldera in 2016, as the gas fluxes and exit paths are likely to be unstable in this highly dynamic volcanic-glacial system; the emission rate from 2016 (12–20 kt/d) is compatible with the total emission rate (flank + caldera) from 2017 (13–24 kt/d). Emission rate of 12–24 kt/d is significant on a global level. Table 1 compares the Katla fluxes to other volcanic sources in Iceland. In Iceland, the previous estimates of total natural CO₂ flux amounted to 2.7–5.8 kt/d (Ármannsson et al., 2005) and included emissions from only four volcanic systems (Grímsvötn, Eyjafjallajökull, Hekla, and Krafla). The emissions from Katla (12–24 kt/d) are therefore at least double the previous estimates of total natural CO₂ from Iceland. Compared to the top global volcanic CO₂ emitters, Katla is one of the top three, potentially exceeded only by Nyiragongo (1–95 kt/d, Arellano et al., 2017, Le Guern, 1987) and Popocatépetl (9–40 kt/d, Gerlach et al., 1997, Delgado et al., 1998).

It should not be assumed that this airborne study has captured all the CO₂ sources from Katla under all conditions. For example, CO₂ emission has also been detected near several other outlet rivers by ground-based gas sensors (Icelandic Met Office monitoring data). These measurements are ill-suited to determining the total flux of CO₂ being released, nor are they suitable for determining the maximum concentrations of CO₂ released, as this would need to be measured at the mouth of the outlet river, an unstable, dynamic environment where permanent installations are unsustainable. We share these results here to show that there are additional, noncontinuous, ground-level emissions of CO₂ from Katla volcano that may not be captured in our aircraft-based assessment. Ground-based CO₂ concentration measurements during a jökulhlaup were made at the outlet river Jökulsá á Sólheimasandi in July 2014, with values of up to 12,000-ppm CO₂. These measurements did not start until after the peak of the flood. Measurements were also made during small jökulhlaups at the outlet river Múlakvísl in August 2016 (concentration in excess of 1,400 ppm) and November 2017 (concentration in excess of 4,000 ppm). These ground-based observations demonstrate that at least during flooding events, additional CO₂ sources exist at Katla.

Studies from Kilauea (Poland et al., 2012) and Redoubt volcanoes (Werner et al., 2012) showed that increases in CO₂ flux of up to 10–20 kt/d may precede eruptive activity by weeks to months. It is not yet known if Katla's large degassing rate is part of its steady state, or if it has been increasing recently. The depth of the degassing is also unknown; basaltic melts can become saturated in CO₂ at tens of kilometer depths. We estimate the lower and upper limits of the mass of magma degassing per day required to sustain the fluxes we observed using magmatic CO₂ contents measured in recent basaltic eruptions in Iceland (no data are available for Katla): 0.14 wt % from Holuhraun 2014–2015 (lower limit, Bali et al., 2018) and 1.1 wt % CO₂ from Fimmvörðuháls 2010 (upper limit, Burton et al., 2015). Using the lower limit of the observed CO₂ flux of 12 kt/day the required minimum magma mass flux (assuming 1.1 wt % CO₂) is 0.18 km³ per year. If the lower limit of 0.1 wt % is assumed, the volume of magma increases by an order of magnitude (1.8 km³ per year). Further ground deformation studies at Katla are recommended to shed light on the volumes suggested by the gas measurements.

4. Conclusions

The discovery of a very large CO₂ emission from Katla volcano is novel, as Katla was thought to be a minor emitter of gases between the periodic jökulhlaups and eruptions (last eruption in 1918 C.E.). We have shown unequivocally that Katla volcanic system as a whole is a source of CO₂, but the exact location(s) of the degassing sources is still unknown (and are potentially dynamic). Using model simulations, we have made an attempt to show that the degassing sources are likely to be located on the western flank and within the central Katla caldera. However, further direct observations are needed to locate these sources with greater accuracy.

The globally significant CO₂ emission from Katla may indicate that this volcanic system is supplied by magmatic gas from depth and that the ongoing geophysical unrest in Katla is due to magma movements. It is not yet known if this is Katla's steady-state or if the gas flux is changing. The collection of a CO₂ flux time series and measurements of other gas species, including, for example, hydrogen sulfide and methane, will therefore be critical for furthering our understanding. Regular gas measurements, airborne and/or ground-based, should be established as part of routine monitoring at this highly active and hazardous volcano.

It is not known how representative Katla may be of other subglacial volcanoes, in Iceland or globally. A global total volcanic CO₂ flux from passively degassing subaerial volcanoes was estimated at 540 kt/d (Burton et al., 2013) based on extrapolation of CO₂ flux measurements from 33 volcanoes to an estimated 150 volcanoes (Global Volcanism Program, 2013). Only 3 of the measured 33 volcanoes were subglacial (Redoubt, Spurr, and Grímsvötn). In comparison, our measurements of CO₂ flux from Katla represent around 2–4% of this total. If degassing from subglacial volcanoes occurs widely on the same scale as Katla, then the total contribution from subglacial volcanoes would change the global CO₂ degassing estimate very significantly. However, the size and relatively recent activity of Katla may make it an exceptional emitter; this will remain an open question until similar measurements can be made on more subglacial volcanoes. We conclude that further airborne measurements using sensitive gas sensors are urgently required, targeted on subglacial volcanoes to establish if Katla volcanism is the exception, or the rule.

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References

Ágústsdóttir, A. M., & Brantley, S. L. (1994). Volatile fluxes integrated over four decades at Grímsvötn volcano, Iceland. *Journal of Geophysical Research*, 99(B5), 9505–9522. <https://doi.org/10.1029/93JB03597>

Aiuppa, A., Bittetto, M., Francofonte, V., Velasquez, G., Parra, C. B., Giudice, G., et al. (2017). A CO₂-gas precursor to the March 2015 Villarrica volcano eruption. *Geochemistry, Geophysics, Geosystems*, 18, 2120–2132. <https://doi.org/10.1002/2017GC006892>

Arellano, S., Yalire, M., Galle, B., Bobrowski, N., Dingwell, A., Johansson, M., et al. (2017). Long-term monitoring of SO₂ quiescent degassing from Nyiragongo's lava lake. *Journal of African Earth Sciences*, 134, 866–873. <https://doi.org/10.1016/j.jafrearsci.2016.07.002>

Ármannsson, H., Fridriksson, T., & Kristjánsson, B. R. (2005). CO₂ emissions from geothermal power plants and natural geothermal activity in Iceland. *Geothermics*, 34(3), 286–296. <https://doi.org/10.1016/j.geothermics.2004.11.005>

Ármannsson, H., Fridriksson, T., Wiese, F., Hernandez, P., & Perez, N. (2007). CO₂ budget of the Krafla geothermal system, NE-Iceland, in: *Proceedings of the 12th International Symposium on Water-Rock Interaction 2007* (pp. 189–192). London: Taylor & Francis Group.

Arnórsson, S., & Gíslason, S. R. (1994). CO₂ from magmatic sources in Iceland. *Mineralogical Magazine*, 58A(1), 27–28. <https://doi.org/10.1180/minmag.1994.58A.1.17>

Bali, E., Hartley, M. E., Halldórsson, S. A., Gudfinnsson, G. H., & Jakobsson, S. (2018). Melt inclusion constraints on volatile systematics and degassing history of the 2014–2015 Holuhraun eruption, Iceland. *Contributions to Mineralogy and Petrology*, 173, 9. <https://doi.org/10.1007/s00410-017-1434-1>

Bergsson, B. (2016). Volcanogenic floods at Sólheimajökull. Hazard identification, monitoring and mitigation of future events, (Master's thesis, 97 pp.). Faculty of Life and Environmental sciences, University of Iceland, Reykjavík.

Biass, S., Scaini, C., Bonadonna, C., Folch, A., Smith, K., & Höskuldsson, A. (2014). A multi-scale risk assessment for tephra fallout and airborne concentration from multiple Icelandic volcanoes—Part 1: Hazard assessment. *Natural Hazards and Earth System Sciences*, 14(8), 2265–2287. <https://doi.org/10.5194/nhess-14-2265-2014>

Burton, M., Ilyinskaya, E., La Spina, A., Salerno, G., Bergsson, B., Donovan, A., et al. (2015). Contrasting gas compositions and fluxes produced by the Holuhraun 2014/2015 eruption and the Fimmvörðuháls 2010 eruption, Iceland. *Geophysical Research Abstracts*, 17, EGU2015-15899. Retrieved from <http://meetingorganizer.copernicus.org/EGU2015/EGU2015-15899.pdf>

Burton, M. R., Sawyer, G. M., & Granieri, D. (2013). Deep carbon emissions from volcanoes. *Reviews in Mineralogy and Geochemistry*, 75(1), 323–354. <https://doi.org/10.2138/rmg.2013.75.11>

Chiodini, G. (2009). CO₂/CH₄ ratio in fumaroles a powerful tool to detect magma degassing episodes at quiescent volcanoes. *Geophysical Research Letters*, 36, L02302. <https://doi.org/10.1029/2008GL036347>

Delgado, H., Piedad-Sánchez, N., Galvan, L., Julio, T., Alvarez, M., & Cardenas, L. (1998). CO₂ flux measurements at Popocatepetl volcano: II magnitude of emissions and significance. *EOS TransSupplement*, 79, F926.

Doukas, M. P., & McGee, K. A. (2007). A compilation of gas emission-rate data from volcanoes of Cook Inlet (Spurr, Crater Peak, Redoubt, Iliamna, and Augustine) and Alaska Peninsula (Douglas, Fourpeaked, Griggs, Mageik, Martin, Peulik, Ukinrek Maars, and Veniaminof), Alaska, from 1995–2006 (USGS Numbered Series No. 2007–1400), Open-File Report. Geological Survey (U.S.).

Fridriksson, T., Kristjánsson, B. R., Ármannsson, H., Margrétardóttir, E., Ólafsdóttir, S., & Chiodini, G. (2006). CO₂ emissions and heat flow through soil, fumaroles, and steam heated mud pools at the Reykjanes geothermal area, SW Iceland. *Applied Geochemistry*, 21(9), 1551–1569. <https://doi.org/10.1016/j.apgeochem.2006.04.006>

Fridriksson, T., Oladóttir, A. A., Jonsson, P., & Eyjolfsdóttir, E. I. (2010). The response of the Reykjanes geothermal system to 100 MW_e power production: Fluid chemistry and surface activity. In *Proceedings World Geothermal Congress*. Bali, Indonesia.

Friedlingstein, P., Houghton, R. A., Marland, G., Hackler, J., Boden, T. A., Conway, T. J., et al. (2010). Update on CO₂ emissions. *Nature Geoscience*, 3(12), 811–812. <https://doi.org/10.1038/ngeo1022>

Gerlach, T. M., Doukas, M. P., McGee, K. A., & Kessler, R. (1999). Airborne detection of diffuse carbon dioxide emissions at Mammoth Mountain, California. *Geophysical Research Letters*, 26(24), 3661–3664. <https://doi.org/10.1029/1999GL008388>

Gerlach, T. M., McGee, K. A., Elias, T., Sutton, A. J., & Doukas, M. P. (1997). Application of the LI-COR CO₂ analyzer to volcanic plumes: A case study, volcán Popocatépetl, Mexico, June 7 and 10, 1995. *Journal of Geophysical Research*, 102(B4), 8005–8019. <https://doi.org/10.1029/96JB03887>

Gíslason, S. (2000). Carbon dioxide from Eyjafjallajökull and chemical composition of spring water and river water in the Eyjafjallajökull–Mýrdalsjökull region (no. RH-06-2000). Science Institute, University of Iceland.

Gíslason, S. R., Andrásdóttir, A., Sveinbjörnsdóttir, Á., Óskarsson, N., Thordarson, T., Torssander, P., et al. (1992). Local effects of volcanoes on the hydrosphere: Example from Hekla, southern Iceland. *Water-rock interact. Rotterdam Balkema*, 1, 477–481.

Global Volcanism Program (2013). Volcanoes of the World, v. 4.7.3. Venkze, E (ed.). Smithsonian Institution. Downloaded 7 Sep 2018. <https://doi.org/10.5479/si.GVP.VOTW4-2013>

Guðmundsson, M., Högnadóttir, b., Kristinsson, A., & Guðbjörnsson, S. (2007). Geothermal activity in the subglacial Katla caldera, Iceland, 1999–2005, studied with radar altimetry. *Annals of Glaciology*, 45, 66–72. <https://doi.org/10.3189/172756407782282444>

Hernández, P., Pérez, N., Fridriksson, T., Egbert, J., Ilyinskaya, E., Thárhallsson, A., et al. (2012). Diffuse volcanic degassing and thermal energy release from Hengill volcanic system, Iceland. *Bulletin of Volcanology*, 74(10), 2435–2448. <https://doi.org/10.1007/s00445-012-0673-2>

Ilyinskaya, E., Aiuppa, A., Bergsson, B., Di Napoli, R., Fridriksson, T., Óladóttir, A. A., et al. (2015). Degassing regime of Hekla volcano 2012–2013. *Geochimica et Cosmochimica Acta*, 159, 80–99. <https://doi.org/10.1016/j.gca.2015.01.013>

Jónsdóttir, K., Roberts, R., Pohjola, V., Lund, B., Shomali, Z. H., Tryggvason, A., et al. (2009). Glacial long period seismic events at Katla volcano, Iceland. *Geophysical Research Letters*, 36, L11402. <https://doi.org/10.1029/2009GL038234>

Larsen, G. (2000). Holocene eruptions within the Katla volcanic system, south Iceland: Characteristics and environmental impact. *Jökull*, 49, 1.

Le Guern, F. (1987). Mechanism of energy transfer in the lava lake of Nyiragongo (Zaire), 1959–1977. *Journal of Volcanology and Geothermal Research*, 31(1–2), 17–31. [https://doi.org/10.1016/0377-0273\(87\)90003-5](https://doi.org/10.1016/0377-0273(87)90003-5)

Óladóttir, B. A., Sigmarsdóttir, O., Larsen, G., & Thordarson, T. (2008). Katla volcano, Iceland: Magma composition, dynamics and eruption frequency as recorded by Holocene tephra layers. *Bulletin of Volcanology*, 70(4), 475–493. <https://doi.org/10.1007/s00445-007-0150-5>

Pálmasón, G., Johnsen, G. V., Torfason, H., Sæmundsson, K., Ragnars, K., Haraldsson, G. I., et al. (1985). Assessment of geothermal energy in Iceland (no. OS-85076/JHD-10). Orkustofnun.

Poland, M. P., Miklius, A., Sutton, A. J., & Thornber, C. R. (2012). A mantle-driven surge in magma supply to Kilauea volcano during 2003–2007. *Nature Geoscience*, 5(4), 295–300. <https://doi.org/10.1038/ngeo1426>

Soosalu, H., Jónsdóttir, K., & Einarsson, P. (2006). Seismicity crisis at the Katla volcano, Iceland—Signs of a cryptodome? *Journal of Volcanology and Geothermal Research*, 153(3–4), 177–186. <https://doi.org/10.1016/j.jvolgeores.2005.10.013>

Spaans, K., Hreinsdóttir, S., Hooper, A., & Ófeigsson, B. G. (2015). Crustal movements due to Iceland's shrinking ice caps mimic magma inflow signal at Katla volcano. *Scientific Reports*, 5(1). <https://doi.org/10.1038/srep10285>

Sturkell, E., Einarsson, P., Roberts, M. J., Geirsson, H., Gudmundsson, M. T., Sigmundsson, F., et al. (2008). Seismic and geodetic insights into magma accumulation at Katla subglacial volcano, Iceland: 1999 to 2005. *Journal of Geophysical Research*, 113, B03212. <https://doi.org/10.1029/2006JB004851>

Thordarson, T., Self, S., Miller, D. J., Larsen, G., & Vilmundardóttir, E. G. (2003). Sulphur release from flood lava eruptions in the Veidivötn, Grímsvötn and Katla volcanic systems, Iceland. *Geological Society of London, Special Publication*, 213(1), 103–121. <https://doi.org/10.1144/GSL.SP.2003.213.01.07>

Werner, C., Christenson, B. W., Hagerty, M., & Britten, K. (2006). Variability of volcanic gas emissions during a crater lake heating cycle at Ruapehu volcano, New Zealand. *Journal of Volcanology and Geothermal Research*, 154(3–4), 291–302. <https://doi.org/10.1016/j.jvolgeores.2006.03.017>

Werner, C., Evans, W., Kelly, P., McGimsey, R., Pfeffer, M., Doukas, M., et al. (2012). Deep magmatic degassing versus scrubbing: Elevated CO₂ emissions and C/S in the lead-up to the 2009 eruption of Redoubt volcano, Alaska. *Geochemistry, Geophysics, Geosystems*, 13, Q03015. <https://doi.org/10.1029/2011GC003794>

Werner, C., Hurst, T., Scott, B., Sherburn, S., Christenson, B. W., Britten, K., et al. (2008). Variability of passive gas emissions, seismicity, and deformation during crater lake growth at White Island volcano, New Zealand, 2002–2006. *Journal of Geophysical Research*, 113, B01204. <https://doi.org/10.1029/2007JB005094>

Werner, C., Kelly, P. J., Doukas, M., Lopez, T., Pfeffer, M., McGimsey, R., et al. (2013). Degassing of CO₂, SO₂, and H₂S associated with the 2009 eruption of Redoubt volcano, Alaska. *Journal of Volcanology and Geothermal Research*, 259, 270–284. <https://doi.org/10.1016/j.jvolgeores.2012.04.012>