Bulletin of Volcanology (2024) 86:60
https://doi.org/10.1007/s00445-024-01751-7

SHORT SCIENTIFIC COMMUNICATION

CO_2 emissions during the 2023 Litli Hrútur eruption in Reykjanes, Iceland: $\delta^{13}C$ tracks magma degassing

Tobias P. Fischer[1] · **Céline L. Mandon**[2] · **Scott Nowicki**[1] · **John Ericksen**[3] · **Felipe Rojas Vilches**[1] · **Melissa A. Pfeffer**[4] · **Alessandro Aiuppa**[5] · **Marcello Bitetto**[5,6] · **Angelo Vitale**[5] · **G. Matthew Fricke**[3] · **Melanie E. Moses**[3,7] · **Andri Stefánsson**[2]

Received: 28 February 2024 / Accepted: 19 May 2024

Abstract

We report CO_2 emission rates and plume $\delta^{13}C$ during the July 2023 eruption at Litli Hrútur in the Fagradalsfjall region of the Reykjanes Peninsula. The CO_2 emission rates were measured by UAV utilizing a new method of data extrapolation that enables obtaining rapid flux results of dynamic eruption plumes. The $\delta^{13}C$ values are consistent with degassing-induced isotopic fractionation of the magma during and after the eruption. Our results show that rapid, real-time CO_2 flux measurements coupled with isotopic values of samples collected at the same time provide key insights into the dynamics of volcanic eruptions and have the potential of forecasting the termination of activity.

Keywords Reykjanes eruptions · CO_2 emissions · Carbon isotopes · Magma degassing · UAV (uncrewed aerial vehicle)

Introduction

Rapid measurements of volcanic CO_2 emissions prior to and during an eruption are an important forecasting tool (Aiuppa et al. 2010) because of CO_2's low solubility in silicic magmas (Holloway 1976; Holloway and Blank 1994) resulting in the early release of CO_2 from ascending melts. CO_2 is a greenhouse gas (Arrhenius 1896), and constraining its emissions from volcanoes during quiescence and eruptions is central to reconstructing the preindustrial geological carbon cycle (Berner 2004) and assessing its role in climate modulations over geologic time (Sleep and Zahnle 2001). Due to seemingly insurmountable challenges of directly measuring CO_2 in plumes emitted from volcanoes during passive degassing and eruptions utilizing currently available satellite-based remote sensing approaches, such measurements remain exceedingly rare. Johnson et al. (2020) utilized the Orbiting Carbon Observatory-2 (OCO-2) and successfully measured the CO_2 emissions from the 2018 Kīlauea eruption. Their measurements were made during 1 day of observations where conditions were ideal to enable collection of high-quality data. The OCO-2 16-day repeat cycle currently makes this method impractical for frequent, high-rate CO_2 flux measurements from erupting volcanoes, and the only other successful volcanic CO_2 emission study using satellites was made at Yasur, Vanuatu (Schwandner et al. 2017). Therefore, these satellite-based methods are not practical for routine, high-rate measurements during eruptions. This leaves ground-based remote sensing (Burton et al. 2013) or methods utilizing uncrewed aerial vehicles (UAV) equipped with sensors (Liu et al. 2020) for volcanic plume CO_2 flux determinations. As a result of these issues, almost all CO_2 flux data from volcanic plumes reported to date are from non-erupting volcanoes and based on measurements of SO_2 flux using satellite- or ground-based methods combined with

Editorial responsibility: J. Gardner

✉ Tobias P. Fischer
fischer@unm.edu

1 Department of Earth and Planetary Sciences, University of New Mexico, Albuquerque, NM, USA
2 Nordic Volcanological Center, Institute of Earth Sciences, University of Iceland, Reykjavík, Iceland
3 Department of Computer Science, University of New Mexico, Albuquerque, NM, USA
4 Icelandic Meteorological Office, Reykjavík, Iceland
5 Dipartimento di Scienze della Terra e del Mare (DiSTeM), Università Di Palermo, Palermo, Italy
6 Istituto Nazionale di Geofisica e Vulcanologia (INGV), Sezione di Palermo, Palermo, Italy
7 External Faculty, Santa Fe Institute, Santa Fe, NM, USA

CO_2/SO_2 that has been measured in the plume at some point in time and generally not during the same time as the SO_2 flux measurements (Fischer and Aiuppa 2020; Fischer et al. 2019; Werner et al. 2019). CO_2 flux determinations during eruptions depend on eruptive CO_2/SO_2 ratios, but due to obvious challenges, these are extremely scarce, and out of 89 eruptions in the period from 2005–2017, only 26 have this ratio measured just before the eruption (Fischer et al. 2019). FTIR (Fourier transform infrared)-based methods alleviate this issue by enabling measurements during eruptions (Burton et al. 2007; Oppenheimer et al. 2014, 2018) but require favorable geometries and a strongly emitting IR source such as a lava lake or fountaining limiting applicability to a broad range of volcanoes. Combined approaches (FTIR + drone-based Multi-GAS) are especially useful, but unfortunately still rare (Burton et al. 2023; Halldórsson et al. 2022).

The Reykjanes peninsula, in southwest Iceland, is a subaerial trans-tensional oblique rift which comprises five volcanic complexes (Fig. 1a). Recent volcanic activity started in March 2021 near Fagradalsfjall mountain in Geldingadalir valley (Fig. 1b), after a year of tectonic unrest within Svartsengi and Fagradalsjfall complexes. The magma sources, evolution, and transport have been studied throughout the 6-month eruption, with detailed geophysical, petrologic, and geochemical studies (e.g., Halldórsson et al. 2022; Pedersen et al. 2022; Scott et al. 2023). Another eruption started on 3 August 2022 in Meradalir valley, which lasted 3 weeks. The third eruption within the Fagradalsfjall complex started on July 10, 2023, with a fissure opening near Litli Hrútur mountain, about 30 km from Reykjavík, Iceland's capital city (Kornei 2023) and continued for almost a month until August 5th.

We report here the first CO_2 flux from this eruption that was directly measured, rather than calculated from SO_2 flux and CO_2/SO_2 ratios. We collected data starting on July 16, 2023, less than a week after eruption initiation at Litli Hrútur, and used a new method for measuring plume CO_2 fluxes directly by utilizing UAV-based sensing systems coupled with novel extrapolation approaches. We compare this method of CO_2 flux determination with the conventional method of combining remotely sensed SO_2 flux measurements with in-plume CO_2/SO_2 ratios to obtain volcanic CO_2 fluxes. While this method has initially been tested at the 2021 eruption of Tajogaite on La Palma Island, Spain (Ericksen et al., 2024), the 2023 eruption at Litli Hrútur provided an ideal laboratory to further refine

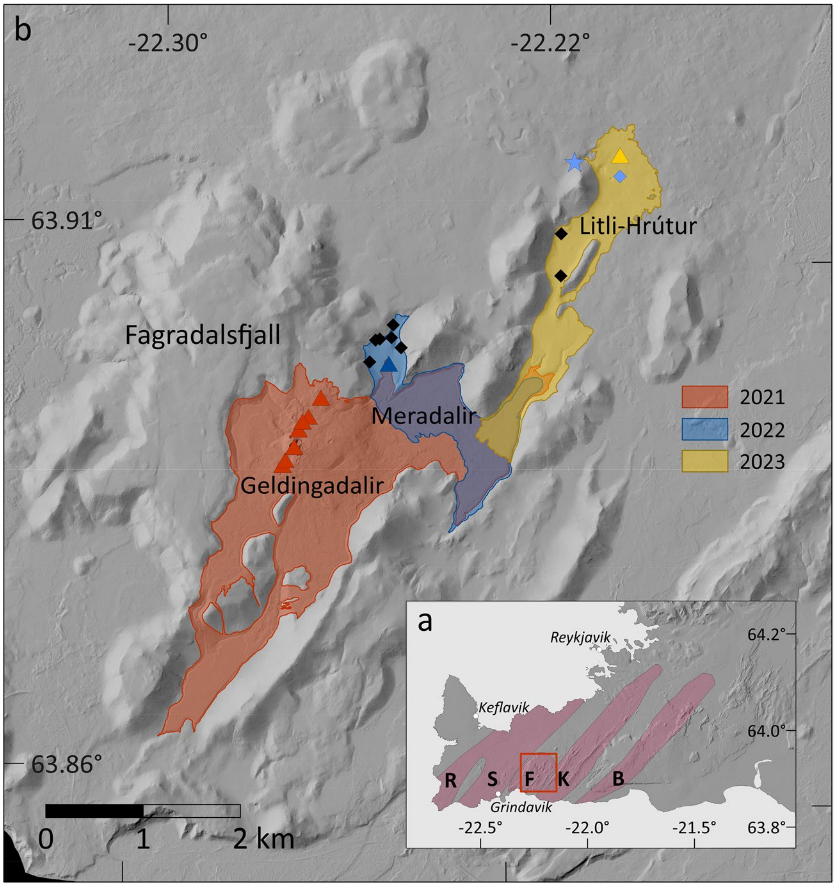

Fig. 1 Fagradalsfjall complex in the Reykjanes peninsula. (b) Map of the Reykjanes peninsula and its volcanic complexes: R, Reykjanes; S, Svartsengi; F, Fagradalsfjall; K, Krýsuvík; B, Brennisteinsfjöll (Sæmundsson et al. 2010). The red square indicates the area displayed in (a). (a) Map showing the vents (triangles) and outlines of lava fields from the 2021, 2022, and 2023 eruptions of the Fagradalsfjall complex (Geldingadalir, Meradalir, Litli Hrútur). The black diamonds are the ground-based sampling locations for $\delta^{13}C$ measurements. The drone launch site during the 2023 eruption is represented by the blue star, with the blue diamond showing the approximate location of drone-based samples for $\delta^{13}C$ measurements. Lava field outlines are modified from this source https://atlas.lmi.is/mapview/?application=umbrotasja

the method and compare the results with conventional measurements made during the same period of time. We also report $\delta^{13}C$ values of CO_2 gas samples collected by UAV in the eruption plume and analyzed by isotope ratio infrared spectroscopy at the University of Iceland utilizing the University of New Mexico's Delta Ray instrument following the approach of Fischer and Lopez (2016). We compare the data with gas samples collected from vents and lavas at the 2022 eruption site at Fagradalsfjall.

Methods

We present here a novel approach that is similar to the "ladder" technique that utilizes CO_2 sensors mounted on fixed-wing aircraft or helicopter (Gerlach et al. 1997; Werner et al. 2013). However, we use a PP Systems SBA-5, a much lower-cost and smaller sensor mounted on a UAV (Ericksen et al., 2024; Ericksen et al. 2022). In our approach, we fly one UAV equipped with a Multi-GAS, the same unit as used in Halldórsson and al. (2022), Liu et al. (2020), and Burton et al. (2023), in a vertical profile up through the visually estimated center of the plume to locate the highest concentration of CO_2 in the vertical. We then fly a UAV with an SBA-5 sensor in a horizontal transect through the plume, perpendicular to the wind direction and at the altitude of the highest concentration. Because the UAV measurements are made much closer to the source than aircraft-borne measurements, the plume has had less time to mix with the surrounding air and is more turbulent and more heterogeneous (Sparks et al. 1997; Wang and Law, 2002). In our approach, the transect flights are automated with the CO_2 data and UAV location transmitted in real time to the pilot. In the ideal case, multiple transects are flown through the plume at various altitudes to make a CO_2 concentration map as is done during the "ladder" approach, but this is often challenging for highly variable or rapidly evolving, distant, or wide plumes.

For this experiment, we flew multiple consecutive horizontal transects through the gas plume during the eruption. We flew these at the same distance from the vent and roughly (± 10 m) at the same altitude. We found the plume to be highly dynamic over short periods of time, indicating that the "ladder method" (Gerlach et al. 1997; Werner et al. 2013) in which multiple transects are used to characterize the cross section of the plume is not well suited for the Litli Hrútur plume during this phase of the eruption. To still obtain CO_2 emission rate estimates, we use individual one-dimensional horizontal transects to describe the plume over a short period of time by assuming a normally distributed plume concentration and fit a Gaussian curve to the CO_2 concentration profile (Fig. 2). We use the following procedure to obtain the emission rate: (1) Set an ambient CO_2 concentration threshold and discard concentrations below this threshold from the sample; (2) calculate the mean and standard deviation of the plume; (3) fit the Gaussian model amplitude to the plume concentration by minimizing the χ^2 error between the data and the model; (4) extrapolate the Gaussian curve to two dimensions by assuming symmetrical horizontal and vertical dimensions of the plume. This Gaussian distribution is integrated to calculate a cross-sectional concentration. We then multiply the integrated Gaussian transect distribution by the measured wind speed to yield the CO_2 flux (Gerlach, et al 1997; Werner et al. 2013; Ericksen et al., 2024). This method, termed the VolCAN method, is shown in detail with all data presented in the publicly accessible Jupyter Notebook linked here: https://github.com/BCLab-UNM/iceland-2023-expedition/tree/ATM2023.

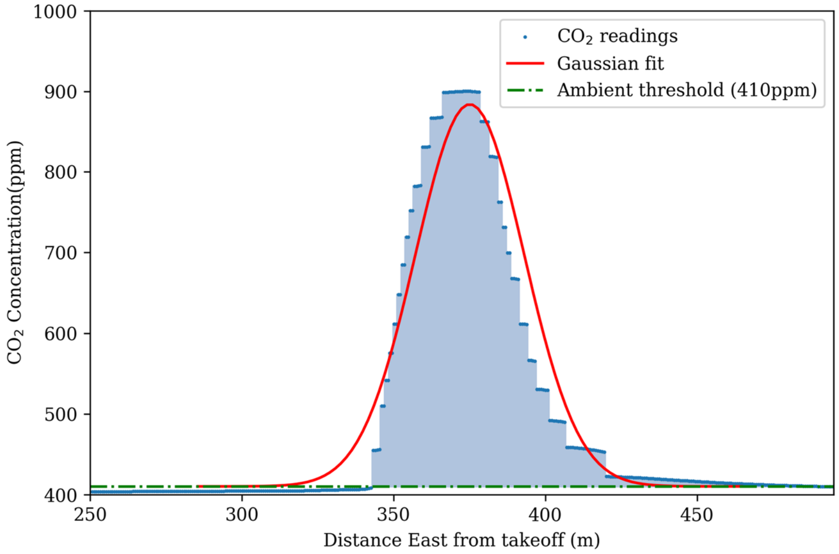

Fig. 2 Gaussian model fit to the sample CO_2 data collected at distances from takeoff location along the plume transect. The red curve represents the Gaussian fit to the data points (blue dots), where the mean, standard deviation, and amplitude are calculated to minimize the χ^2 error. The one-dimensional model is mirrored to two dimensions, integrated, and multiplied by wind speed to calculate the plume flux

Our approach should provide the most accurate results if the plume is transected through its highest vertical concentration point and the plume approaches a Gaussian shape of concentration (Fig. 2). Considering the observed highly dynamic nature of the plume, this approach should result in a self-consistent estimate of flux. In order to test the accuracy of our rapid measurements, we compare the results to CO_2 fluxes obtained by making vehicle-based traverses under the plume using a miniDOAS SO_2 sensor (Galle et al. 2002) and in plume UAV-Multi-GAS collected CO_2/SO_2 ratios (Liu et al. 2020).

Figure 3 shows the experimental observations made on July 14 and July 16, 2023, at the vent of Litli Hrútur. On July 14, at a distance of 7.5 km downwind from the vent, we performed two driving traverses using UNM's miniDOAS system mounted on a vehicle. The spectra were collected using the freely available MobileDOAS software. The first transect was 4.5, and the second 1.7 km long. Both transects captured the entire width of the plume. On July 16, we launched the Dragonfly UAV (a UNM-designed and built quadcopter described in Ericksen et al. (2022)) from a location about 450 m from the active vent to perform several transects through the plume. Prior to the launch of the Dragonfly, we measured the CO_2 concentration of the vertical plume profile using a DJI Matrice UAV (a large quadcopter) equipped with the Multi-GAS sensor system from the University of Palermo. We performed the vertical transect at a distance of 375 m from the vent, and the highest CO_2 concentration of 1472 ppm was detected at a height of 60 m above the launch site at 16:45 h local time. We measured CO_2/SO_2 ratios using the University of Palermo's UAV-mounted Multi-GAS system on July 16 at 18:30 local time. We also collected one Tedlar bag sample of the plume on July 16 using our DJI Phantom 4 (a small quadcopter) at a distance of 125 m from the vent that was analyzed by isotope ratio infrared spectrometer and contained 650 ppm CO_2, or about 230 ppm above ambient.

While the source of the degassing CO_2 during the eruption is clearly magmatic, we utilize carbon isotopes to constrain the extent of magma degassing. To achieve this, we use the UAV to collect gas samples of the dilute volcanic plume during the eruption. This approach was pioneered by Chiodini et al. (2011) who collected dilute plume samples from the ground at Solfatara and Vulcano and Fischer and Lopez (2016) who used a helicopter at Aleutian volcanoes. Our UAV-based sampling system has previously been used at Manam volcano in Papua New Guinea for passively degassing plumes (Galle et al. 2021; Liu et al. 2020) and at Tajogaite volcano, La Palma, Spain, during an eruption (Ericksen et al., 2024). At Litli Hrútur, we mounted the sampling system on a DJI Phantom 4. The sample was collected by pumping about 500 ml of gas into a Tedlar bag. The pump was triggered with a timer that was set based on previous flights which allowed us to locate the densest part of the plume. Generally, two samples were collected during each flight. After collection, the

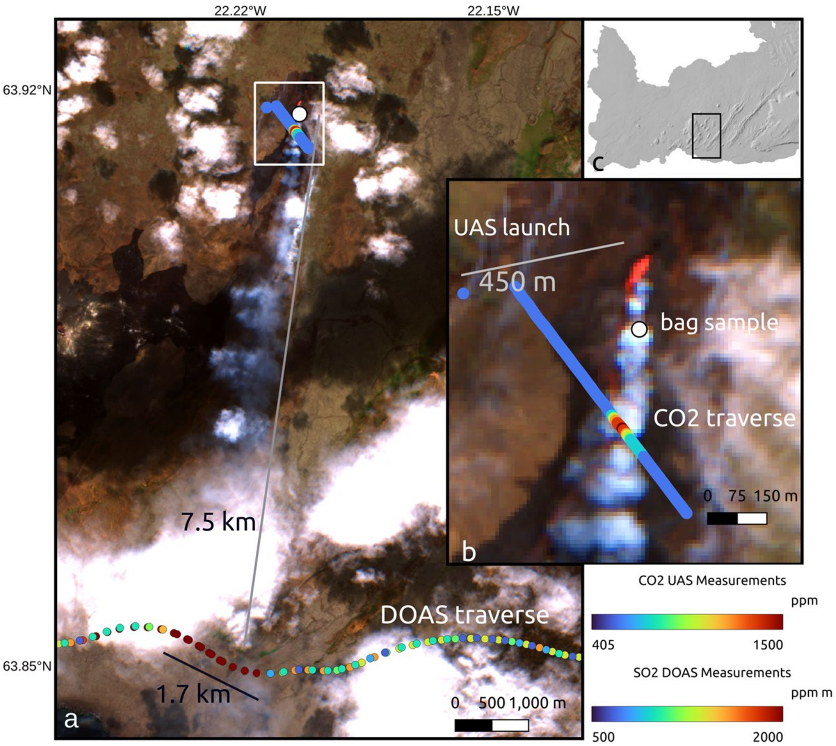

Fig. 3 Map of observations and eruptive conditions overlain on Sentinel 2 visible image. (a) the July 14, 2023, 19:15 local time DOAS traverse was collected by a vehicle-mounted telescope along the paved road 7.5 km from the eruption. (b) CO_2 traverse using the UNM Dragonfly drone, launched about 450 m from the vent and the location of the one bag sample collected on July 16. (c) Overall location of the experiment. Sentinel image was acquired on July 13 during which the wind direction was nearly identical to conditions on July 16, 2023

samples were analyzed by isotope ratio infrared spectroscopy on an instrument that we had temporarily installed at the University of Iceland. We obtained the δ^{13}C values and the CO_2 concentrations of the samples using analytical techniques identical to those reported previously (Fischer and Lopez 2016; Galle et al. 2021; Liu et al. 2020). The error on the δ^{13}C analyses is < 0.1‰ and for CO_2 concentrations < 10 ppm. During 2022, we also collected gas samples from vents and fissures on the lava field produced by the 2022 eruption of Fagradalsfjall (Fig. 1). In December 2023, after the 2023 Litli Hrútur eruption ended, we collected gas samples from vents and fissures on that eruption site. These samples were collected by pumping gas into a Tedlar bag using a syringe and analyzed in the same way as the samples collected by UAV.

Results

The results are shown in Table 1 for gas measurements. The SO_2 flux measurements ranged from 73 to 80 kg/s (wind speed of 17 m/s). The Multi-GAS molar CO_2/SO_2 measurement using the Matrice UAV + Palermo Multi-GAS on July 16 was 2.97 ± 0.52. The Multi-GAS CO_2/SO_2 measurements using the Phantom UAV and UNM Multi-GAS system were 2.35 ± 1.61, 5.40 ± 2.72, and 5.7 ± 2.83 molar on July 19 and 27 and August 2, respectively. The CO_2 flux computed using our SO_2 flux measurements of 73–80 kg/s (wind speed of 17 m/s) and the CO_2/SO_2 molar ratio of 3 results in a CO_2 flux of 150–160 kg/s. The CO_2 flux computed directly using the VolCAN method (Dragonfly + SBA-5) with a wind speed of 10 m/s from NNE results in a CO_2 flux of 20 and 50 kg/s for two passes during the same out and back flight, occurring between 17:20 and 17:23 h local time.

The results of the δ^{13}C analyses of samples collected from the 2023 Litli Hrútur eruption and following this eruption are shown in Table 2. The results of samples collected from degassing fissures and lava flows at the 2022 eruption site are shown in Supplementary Table S1.

Discussion

The two methods for obtaining CO_2 emissions compared here are fundamentally different in their observations, but it is important to state that neither method collects a complete instantaneous cross section of CO_2. Both methods are highly dependent upon the wind speed used to determine the CO_2 flux, but the SO_2 ratio method is further dependent upon the derived SO_2/CO_2 ratio determined from an averaged set of separate observations. We know that CO_2 and SO_2 may not be emitted from the same vents or traveling down-wind in consistent relative concentrations based on observations at Kīlauea (Gerlach et al. 2002) for instance. Our SO_2 flux measurements were made on July 14 while our CO_2/SO_2 ratios and CO_2 transects were made on July 16, i.e., 2 days apart. Our measurements made with UAV-mounted Multi-GAS are variable, and the applied ratio is an average of

Table 1 Results of gas composition and flux measurements on July 14 to August 2, 2023

Method	Date	Time local	Max CO_2 (ppm) vertical	Max CO_2 (ppm) horizontal	Molar CO_2/SO_2	Plume width (m) DOAS transect	Wind speed (m/s)	SO_2 flux (kg/s)	CO_2 flux (kg/s)*
miniDOAS	14-Jul	19:30				4515	17	80	
miniDOAS	14-Jul	19:15				1753	17	73	
Matrice + Palermo MG	16-Jul	16:45	1400						
Matrice + Palermo MG	16-Jul	18:30	1220		2.97 ± 0.52				
Phanom + Bag	16-Jul	16:00	650						
Phantom + UNM MG	17-Jul	13:55–16:20	1196		2.35 ± 1.60				
Phantom + UNM MG	27-Jul	15:50, 16:50	1777		5.40 ± 2.72				
Phantom + UNM MG	2-Aug	14:00–16:00	1721		5.70 ± 2.83				
CO_2 flux techniques									
miniDOAS and MG	14 and 16 Jul				2.97	1752–4515	17	73–80	**150–160**
VolCAN (Dragonfly + SBA-5)	16-Jul	16:30	1400	1472			10		**20–50**

*Note: CO_2 flux computed by miniDOAS and Multi-GAS utilizes the CO_2/SO_2 ratio of 2.97 measured by Matrice + Palermo MG

Data in bold emphasis are the CO_2 fluxes determined with the combined (miniDOAS and Multi-GAS) and the VolCAN method

Table 2 Carbon isotope data collected for this study. Errors on δ^{13}C values are < 0.1‰ and on CO$_2$ concentrations < 10 ppm. Also indicated are methods of sampling and the person who sampled. All July 2023 samples, with the exception of the air samples collected at the University of Iceland (Air RKJ), were collected at the eruption site. The December 2023 samples were collected from the eruption vents after the eruption ended (see Figs. 1 and 2 for locations)

Date	Sample information	δ^{13}C of CO$_2$	CO$_2$ (ppm)	Location information/sample collector name
2023 eruption				
7/10/23	Eruption ground	−8.82	478	Ground/Melissa
7/10/23	Eruption ground	−8.37	470	Ground/Melissa
7/16/23	80-m elevation	−13.53	1291	Plume/John
7/16/23	140-m elevation	−7.14	650	Plume/Scott
7/19/23	35-m elevation	−9.40	520	Plume/Tobias
7/19/23	Air RKJ	−7.32	432	U of I/Tobias
7/23/23	Air RKJ	−7.45	428	U of I/Tobias
7/23/23	Air Vogar	−7.43	434	Vogar/Tobias
7/22/23	L.Hrutur #2	−10.13	578	Plume/Tobias
7/22/23	L.Hrutur #3	−8.19	466	Plume/Tobias
7/24/23	L.Hrutur #1	−9.91	1145	Plume/Tobias
7/24/23	L.Hrutur #2	−7.15	563	Plume/Tobias
7/24/23	L.Hrutur #3	−6.18	1045	Plume/Tobias
7/24/23	L.Hrutur #4	−5.90	1054	Plume/Tobias
7/24/23	L.Hrutur #5	−6.81	579	Plume/Tobias
7/24/23	L.Hrutur fire	−14.10	1304	Moss fire/Tobias
7/24/23	Air RKJ	−8.13	465	U of I/Tobias
7/26/23	Air RKJ	−7.67	438	U of I/Tobias
7/29/23	Air RKJ	−7.88	443	U of I/Tobias
7/25/23	L.Hrutur #1	−7.59	547	Plume/Felipe
7/25/23	L.Hrutur #2	−7.30	540	Plume/Felipe
7/25/23	L.Hrutur #3	−6.45	790	Plume/Felipe
7/27/23	L.Hrutur #1	−7.19	758	Plume/Felipe
7/27/23	L.Hrutur #2	−7.83	630	Plume/Felipe
Post-eruption				
12/2/23	L.Hrutur #1	−13.91	2473	Vent 1/Celine
12/2/23	L.Hrutur #2	−12.70	1644	Vent 1/Celine
12/2/23	L.Hrutur #3	−11.41	954	Vent 1/Celine
12/2/23	L.Hrutur #4	−8.92	559	Vent 2/Celine
12/2/23	L.Hrutur #5	−9.10	549.3	vent 2/Celine

that variance. Rarely is there any spatial knowledge of the plume dynamics when collecting Multi-GAS data, and we do not know how CO$_2$ and SO$_2$ align spatially in the plume at the fine scale. The direct CO$_2$ VolCAN method does not measure a column-integrated profile of the CO$_2$ plume, but the flight path is a two-dimensional sample through a plume rather than a full cross section of the CO$_2$. Given the highly variable nature of the emitted plumes (see the puffy undulating plume shape in the visible Sentinel Imagery in Fig. 2 and Pering et al. (2019)), there is likely to be a high degree of variability between successive observations even when taken only minutes apart. Future work should address these issues by making a high number of vertical and horizontal transects through the plume with the CO$_2$ VolCAN method, collecting mini DOAS SO$_2$ emission data and Multi-GAS data at the same plume location, and combining these with vent FTIR data. This would require an "ideal" eruption that allows such measurements to be made at the same time—a highly challenging objective. The decrease in flux between the SO$_2$ ratio method measured on July 14 (150–160 kg/s) and direct CO$_2$ VolCAN method measured on July 16 (20 and 50 kg/s) is, however, consistent with the visual observations of decreasing overall emissions and CO$_2$ flux values until the eruption ended on August 5, 2023.

Figure 4 shows δ^{13}C and 1/CO$_2$ concentration values of all samples collected during the 2022–2023 period at Fagradalsfjall and Litli Hrútur. Using the Keeling-plot approach (Keeling 1958), our δ^{13}C and CO$_2$ abundance data show mixing between ambient air and pure CO$_2$. We fit extrapolation lines through our various datasets: (1) samples collected from the 2022 vents, following the 2022 eruption at Meradalir until June 2023; (2) samples of the vents of Litli Hrútur, collected after the 2023 eruption; (3) samples contaminated by smoke from moss fires; and (4) samples

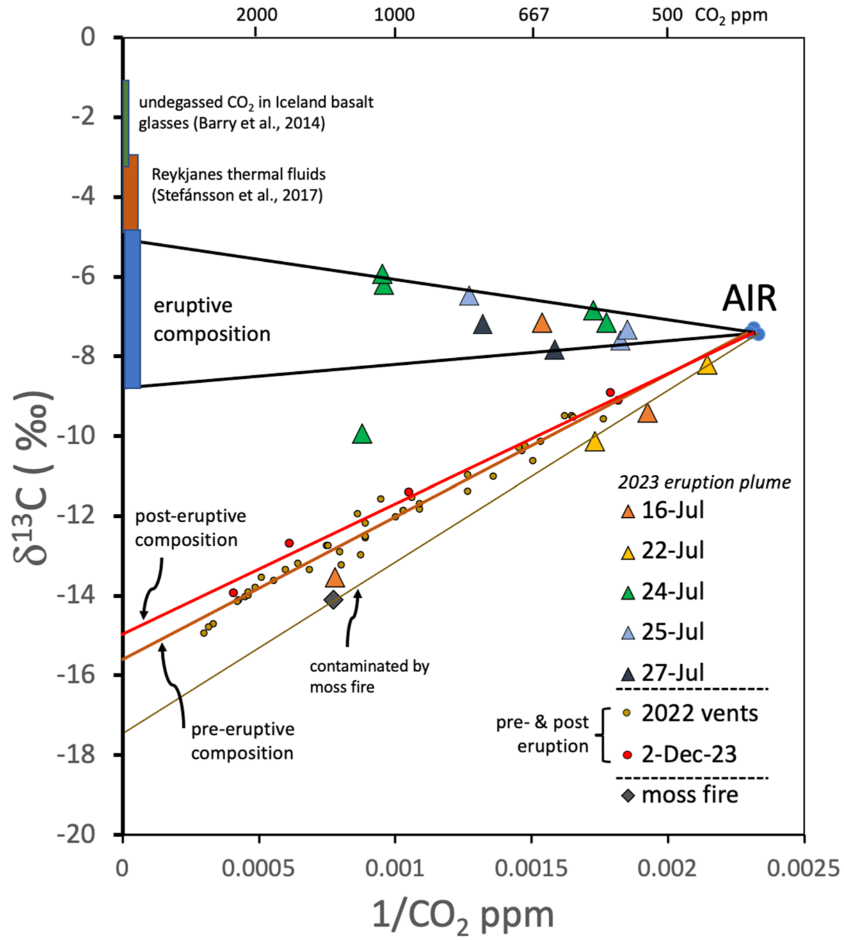

Fig. 4 Carbon isotope data collected using bag sampling and isotope ratio infrared spectroscopy analyses. The 2022 vent samples and the 2-Dec-23 samples were collected from the ground. Analytical errors are smaller than symbol sizes. Also shown are modeled $\delta^{13}C$ values of undegassed magma from Barry et al. (2014) and values measured in fluids from the Reykjanes Peninsula (Stefánsson et al. 2017)

collected by UAV during the 2023 Litli Hrútur eruption. In our extrapolated values, we observe distinct ~ 10‰ differences between samples collected during the eruption and those preceding and following the eruption. This significant difference lies outside of any analytical or sampling error.

Our data are extrapolated to pure CO_2 and show light $\delta^{13}C$ values (~ − 16‰) following the 2022 eruption, while during the 2023 eruption, values between − 5 and − 9‰ are observed approaching the range of the Icelandic mantle in the region as sampled by hydrothermal fluids (− 3.6 ± 0.6‰, Stefánsson et al. 2017). Samples collected at the eruption site in December 2023, about 3 months after the eruption ended, again show light $\delta^{13}C$ values. The samples collected from the 2022 Fagradalsfjall/Meradalir eruption site during the time from September 2022 to June 2023 extrapolate to light $\delta^{13}C$ values of approximately − 16‰. These values are consistent with the lightest $\delta^{13}C$ values of low CO_2 content (~ 10–40 ppm) vesicles and glasses from the Western Rift Zone that are highly degassed (Barry et al. 2014). Barry et al. (2014) also consider the possibility that some glasses with light $\delta^{13}C$ values were affected by wall rock assimilation of crustal organic materials during magma emplacement. While we cannot rule out this possibility, we consider it unlikely because we collected the pre- and post-eruption samples directly at the eruption vents where any crustal organic material has likely been combusted and removed during magma emplacement. In addition, pervasive organic crustal assimilation would result in lighter $\delta^{13}C$ values during the eruption than what we observe.

Several of our samples collected during the 2023 Litli Hrútur eruption were contaminated by smoke from the moss fires that were ignited by the erupting lava. To characterize this contamination, we collected a sample of the moss fire smoke plume which extraoplates to − 17.5‰. We collected our first samples from the 2023 eruption on July 12, 2023, at ground level hundreds of meters from the site. Due to the large distance from the source, the plume gases were significantly diluted by air and showed light values that extrapolate to − 17.5‰. Plume samples collected by UAV on July 16 and 22, 2023, also align with the sample collected directly from the moss fire and extrapolate to − 17.5‰. These light values are clearly distinct from the other 2022 and 2023 samples. Most likely, these lightest samples are contaminated by organic carbon derived from either the lava flowing over vegetation at the start of the eruption in the case of the ground-based sample collected on July 12 and by smoke mixing with plume gases in the case of the one light July 16, the July 22 samples, and the one light July 24 sample. We do not consider these samples representative of a magmatic source.

Notably, samples collected by UAV during July 16 (one sample) and during July 22–27, 2023 (nine samples), extrapolate to δ^{13}C values (− 5 to − 9‰) that are slightly lower than observed in hydrothermal gases at Reykjanes (− 4.96 to − 2.99‰) (Stefánsson et al. 2017) and estimated for undegassed basaltic melts beneath Iceland (− 2.5 ± 1.0‰, Barry et al. (2014)). Although we were not able to collect uncontaminated samples right at the start of the eruption on July 10, 2023, our data show that by July 16, the erupting lava had a δ^{13}C signature (− 5 to − 9‰) that represented fresh and relatively undegassed melts feeding the eruption. Samples collected at the eruption site in December 2023, 3 months after the eruption stopped, show light values extrapolated to − 15‰. Magma degassing results in fractionation of carbon isotopes to progressively lighter values in the gas and remaining melt (Holloway and Blank 1994) and the light values of December 2023 indicate that the magma had extensively degassed 3 months after the eruption had stopped.

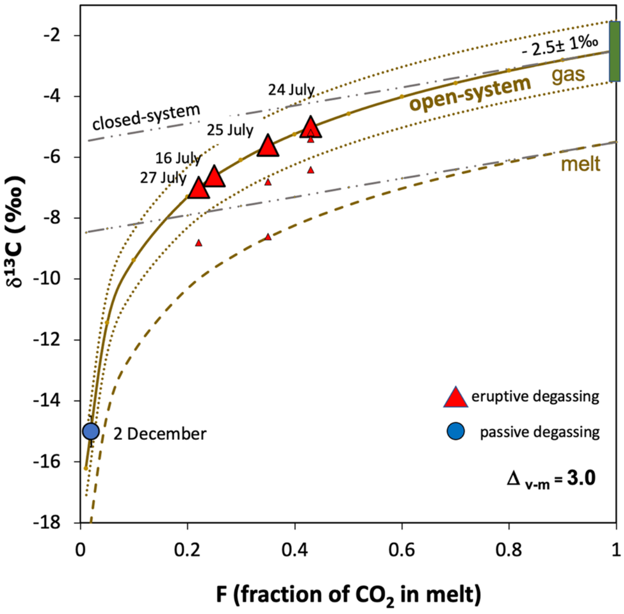

Fig. 5 Degassing fractionation modeled using the approach of Holloway and Blank (1994) for open-system and closed-system equilibrium degassing of CO_2. Gas–melt fractionation factor is from Javoy et al. (1978) and Mattey (1991). We use estimates for the undegassed mantle beneath Iceland (− 2.5 ± 1 from Barry et al. (2014)) as the starting composition. We use the samples with maximum CO_2 contents (shown in Fig. 4) to plot the extrapolated δ^{13}C values collected during and shortly after the eruption in 2023. Smaller red triangles show data from samples with lower CO_2 contents

This magma degassing process can be further assessed and visualized by plotting the extrapolated δ^{13}C values against the degassing fraction (*F* in Fig. 5). The fractionation of CO_2 degassed can be computed using an open-system, classic Rayleigh-type degassing model, assuming experimentally determined isotope fractionation factors for basaltic melts (Holloway and Blank 1994; Rayleigh 1896). Experimentally determined equilibrium isotope fractionation factors for carbon in coexisting vapor and natural basaltic melts range from 2.0 (± 0.2) to 4.3 (± 0.4) (Javoy et al. 1978; Mattey 1991). Consistent with the basaltic composition of erupted lavas, we choose an intermediate vapor-melt distribution coefficient of 3‰ for basaltic melts to illustrate the tracking of the melt degassing process by our δ^{13}C values collected during and following the eruption. We use the values published by Barry et al. (2014) to represent the undegassed melt composition underlying the Reykjanes peninsula (− 2.5 ± 1‰) which is also broadly similar to DMM-values of − 5 ± 1‰ sampled by MORB (Marty and Zimmermann 1999). In Fig. 5, we plot the extrapolated δ^{13}C values of the samples with the maximum CO_2 concentrations, i.e., those least affected by air dilution. Using the model, we can estimate the extent of degassing over this time period. While our data show variability, the results indicate magma degassing from July 24 to July 27, 2023. Based on our data, we infer that the magma had lost about 60% of its gas by July 24 ($F = 0.4$) and was about 80% degassed ($F = 0.2$) by July 27, represented by our last sample collected during the eruption. The eruption ended on August 6, only 10 days after. By December 2023, the magma had lost about 98% ($F = 0.02$) of its initial CO_2. Degassing processes can be more complex and closed-system degassing (Brown et al. 1985) or hybrid models discussed in Aubaud (2022) have been documented. During closed-system degassing, the maximum isotopic shift in the gas is limited to the magnitude of the fractionation factor, i.e., 3‰, and does not explain our data (Fig. 5). For application to volcanic gases, knowledge of *F* is generally not available and δ^{13}C values of gas samples are plotted against assumed (not measured) wt% of dissolved CO_2 (Gerlach and Taylor 1990). In our case, assuming an open-system degassing process enables a relative estimate of the fraction of CO_2 that has left the magma. Our observations utilizing δ^{13}C of CO_2 emitted prior to, during, and after the eruption are consistent with the idea of volatile-charged magma injections into deep crustal levels that drive the eruptive episodes on the Reykjanes Peninsula as proposed by Halldórsson et al. (2022). Once this injected magma is degassed, as shown with progressively lighter δ^{13}C values, the eruption stops until new volatile-rich magma is again injected, as indicated by heavy and mantle-like δ^{13}C emitted during the next eruption. Initial CO_2 contents of Icelandic primitive melts have been estimated at up to 4800 ppm CO_2 and would become saturated with a fluid phase at up to 25 km depth (White et al. 2019). Sixty to eighty percent degassing as estimated by our carbon isotope data would result in melt that contains about 1000–2000 ppm CO_2, consistent with the CO_2 contents of Iceland melt inclusions (up to 1200 ppm (Maclennan 2017)) and fluid saturation at 6–10 km depth. By the time the eruption ends, this melt has lost 98% of its initial CO_2 and is equivalent to 100 ppm remaining in the magma. These low CO_2 contents with light $\delta^{13}C$ values are consistent with those of subglacial glasses sampled by Barry et al. (2014). The light sample of 16 July, collected only 6 days after eruption initiation, may represent CO_2 that has more extensively degassed from a deeper source at initial fluid saturation, has accumulated, and was released during the initial stages of the eruption.

Conclusions

The Litli Hrútur eruption emitted 20–160 kg CO_2/s during its initial stages in mid-July 2023. Our UAV-based approach to directly obtain the CO_2 flux during eruptions is a valid alternative to more complex or involved methods that rely on several types of measurements or extensive traverses through plumes. The source of eruptive CO_2 was from fresh, relatively undegassed magma and distinct from that of the extensively degassed magma that was left at depth after the end of the 2022 eruption at Meradalir to the south. Our observations are consistent with the idea of volatile-charged magma injections into deep crustal levels that drive the eruptive episodes on the Reykjanes Peninsula (Halldórsson et al. 2022). Once this injected magma is degassed, the eruption stops. Samples that are collected prior to, during, and following an eruption like the ones that have occurred in 2022 and 2023 on the Reykjanes peninsula and are rapidly analyzed for $\delta^{13}C$ values give insights into eruption dynamics, magma supply, and timing of eruption cessation.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s00445-024-01751-7.

Acknowledgements TF likes to thank Nicolas Fischer for help during field work and for UAV spotting. We thank the Icelandic civil defense for safety during work on the eruption. We thank Taryn Lopez and Saemundur Halldórsson for constructive reviews that improved the manuscript.

Funding This work was supported by the NSF-funded VolCAN project (NRI 2024520) to MM and NSF RAPIDs (EAR-2336217, EAR-IF-1664246) to TF.

References

Aiuppa A, Burton M, Caltabiano R, Guidice G, Guerrieri S, Liuzzo M, Muré F, Salerno G (2010) Unusually large magmatic CO_2 gas emissions prior to a basaltic paroxysm. Geophys Res Lett 37:L17303. https://doi.org/10.1029/2010GL043837

Arrhenius S, (1896) On the influence of carbonic acid in the air upon the temperature of the ground. Philosophical Magazine and J Sci, 5 41: 237–276

Aubaud C (2022) Carbon stable isotope constraints on CO_2 degassing models of ridge, hotspot and arc magmas. Chem Geol 605:120962

Barry PH, Hilton DR, Füri E, Halldórsson SA (2014) Carbon isotope and abundance systematics of Icelandic geothermal gases, fluids and subglacial basalts with implications for mantle plume-related CO2 fluxes. Geochim Cosmochim Acta 134:74–99

Berner RA (2004) The Phanerozoic carbon cycle. Oxford University Press, New York, p 150

Brown PE, Bowman KW, Kelly WC (1985) Petrologic and stable isotopic constraints on the source and evolution of skarn-forming fluids at Pine Creek. California Econ Geol 80:72–92

Burton M, Allard P, Muré F, La Spina A (2007) Magmatic gas composition reveals the source depth of slug-driven Strombolian explosive activity. Sci 317:227–230

Burton MR, Sawyer GM, Granieri D (2013) Deep carbon emissions from volcanoes. Rev Mineral Geochem: Carbon Earth 75:323–355

Burton M, Aiuppa A, Allard P, Asensio-Ramos M, Cofrades AP, La Spina A, Nicholson EJ, Zanon V, Barrancos J, Bitetto M, Hartley M, Romero JE, Waters E, Stewart A, Hernández PA, Lages JP, Padrón E, Wood K, Esse B, Hayer C, Cyrzan K, Rose-Koga EF, Schiavi F, D'Auria L, Pérez NM (2023) Exceptional eruptive CO_2 emissions from intra-plate alkaline magmatism in the Canary volcanic archipelago. Commun Earth Environ 4(1):467

Chiodini G, Caliro S, Aiuppa A, Avino R, Granieri D, Moretti R, Parello F (2011) First 13C/12C isotopic characterisation of volcanic plume CO_2. Bull Volcanol 73(5):531–542

Ericksen J, Fricke GM, Nowicki S, Fischer TP, Hayes JC, Rosenberger K, Wolf SR, Fierro R, Moses ME (2022) Aerial survey robotics in extreme environments: mapping volcanic CO_2 with flocking UAVs. Front Control Eng 3:836720. https://doi.org/10.3389/fcteg.2022.836720

Ericksen J, Fischer TP, Fricke GM, Nowicki S, Perez NM, Hernandez Perez P, Padron Gonzalez E, Fierro R, Moses ME (2024) Drone CO_2 measurements during the Tajogaite volcanic eruption. Atmos Meas Tech Discus. https://doi.org/10.5194/amt-2023-229

Fischer TP, Aiuppa A (2020) AGU centennial grand challenge: volcanoes and deep carbon global CO_2 emissions from subaerial volcanism: recent progress and future challenges. G-cubed 21(3 e2019GC008690)

Fischer TP, Arellano S, Carn S, Aiuppa A, Allard P, Lopez T, Shinohara H, Kelly PJ, Werner C, Cardelini C, Chiodini G (2019) The emissions of CO_2 and other volatiles from the world's subaerial volcanoes. Sci Rep 9:18716. https://doi.org/10.1038/s41598-41019-54682-41591

Fischer TP, and Lopez TM, (2016). First airborne samples of a volcanic plume for d13C of CO_2 determinations. Geophys Res Lett, 43

Galle B, Arellano S, Bobrowski N, Conde V, Fischer TP, Gerdes G, Gutmann A, Hoffmann T, Itikarai I, Krejci T, Liu EJ, Mulina K, Nowicki S, Richardson T, Rüdiger J, Wood K, Xu J (2021). A multi-purpose, multi-rotor drone system for long range and high-altitude volcanic gas plume measurements. At Meas Tech Discuss. , P. 1–33

Galle B, Oppenheimer C, Geyer A, McGonigle AJS, Edmonds M, Horrocks L (2002) A miniaturised ultraviolet spectrometer for remote sensing of SO_2 fluxes; a new tool for volcano surveillance. j. Volcanol Geotherm Res 119:241–254

T Gerlach G Delgado H McGee K Doukas M Venegas JJL Cárdenas 1997 Application of the LI-COR CO2 analyzer to volcanic plumes: a case study, volcán Popocatépetl, Mexico, June 7 and 10, 1995 J. Geophys. Res. 102 B4 8005 8019

Gerlach TM, Taylor BE (1990) Carbon isotope constraints on degassing of carbon dioxide from Kilauea volcano. Geochimica Cosmochimica Acta 54:2051–2058

Gerlach TM, McGee KA, Elias T, Sutton AJ Doukas MP, (2002) Carbon dioxide emission rate of Kīlauea volcano: implications for primary magma and the summit reservoir. J Geophys Res: Solid Earth, 107(B9): ECV 3–1-ECV 3–15

Halldórsson SA, Marshall EW, Caracciolo A, Matthews S, Bali E, Rasmussen MB, Ranta E, Robin JG, Guðfinnsson GH, Sigmarsson O, Maclennan J, Jackson MG, Whitehouse MJ, Jeon H, van der Meer QHA, Mibei GK, Kalliokoski MH, Repczynska MM, Rúnarsdóttir RH, Sigurðsson G, Pfeffer MA, Scott SW, Kjartansdóttir R, Kleine BI, Oppenheimer C, Aiuppa A, Ilyinskaya E, Bitetto M, Giudice G, Stefánsson A (2022) Rapid shifting of a deep magmatic source at Fagradalsfjall volcano. Iceland Nature 609(7927):529–534

Holloway JR (1976) Fluids in the evolution of granitic magmas: consequences of finite CO2 solubility. Geol Soc America Bulletin 87:1513–1518

Holloway JR, and Blank J (1994) Experimental results applied to C-O-H in natural melts. In: M.R.C.J.R. Holloway (Editor), Volatiles in magmas. Reviews in Mineralogy. Mineral Soc Ameria, Fredericksburg, VA, pp. 187–230

Javoy M, Pineau F, Iiyama I (1978) Experimental determination of the isotopic fractionation between gaseous CO_2 and carbon dissolved in tholeitic magma. Contrib Mineral Petrol 67:35–39

Keeling CD (1958) The concentration and isotopic abundances of atmospheric carbon dioxide in rural areas. Geochim Cosmochim Acta 13:322–334

Kornei K (2023) Third time's the charm for Iceland's Fagradalsfjall. EOS Trans Am Geophys Union 104:2023. https://doi.org/10.1029/2023EO230267.Publishedon19July

Liu E, Aiuppa A, Alan A, Arellano S, Bitetto M, Bobrowski N, Carn S, Clarke R, Corrales E, de Moor JM, Diaz JA, Edmonds M, Fischer TP, Freer J, Fricke GM, Galle B, Gerdes G, Giudice G, Gutmann A, Hayer C, Itikarai I, Jones J, Mason E, McCormick Kilbride BT, Mulina K, Nowicki S, Rahilly K, Richardson T, Rüdiger J, Schipper CL, Watson IM Wood K (2020) Aerial strategies advance volcanic gas measurements at inaccessible, strongly degassing volcanoes: Manam, Papua New Guinea. Sci Adv, 6, (no. 44): eabb9103

Marty B, Zimmermann L (1999) Volatiles (He, C, N, Ar) in mid-ocean ridge basalts: assessment of shallow-level fractionation and characterization of source composition. Geochim Cosmochim Acta 63:3619–3633

Mattey DP (1991) Carbon dioxide solubility and carbon isotope fractionation in basaltic melt. Geochim Cosmochim Acta 55:3467–3473

Oppenheimer C, Fischer TP, Scaillet B (2014) Volcanic degassing: processes and impact. Treatise on Geochemistry (second edition). 4, the crust p.111–179 https://doi.org/10.1016/B1978-1010-1008-095975-095977.000304-095971

Oppenheimer C, Scaillet B, Woods A, Sutton AJ, Elias T Moussallam Y (2018) Influence of eruptive style on volcanic gas emission chemistry and temperature. Nat Geosci https://doi.org/10.1038/s41561-018-0194-5

Pedersen GBM, Belart JMC, Óskarsson BV, Gudmundsson MT, Gies NH, T al, e, (2022) Volume, effusion rate, and lava transport during the 2021 Fagradalsfjall eruption: results from near real-time photogrammetric monitoring. Geophys Res Lett, 49, e2021GL097125

Rayleigh JWS (1896) Theoretical considerations respecting the separation of gases by diffusion or similar processes. Phil Mag 42:493

Sæmundsson K, Jóhannesson, H Hjartarson Á, Kristinsson SG, Sigurgeirsson MA, (2010) Geological map of southwest Iceland, 1: 100000. Iceland geosurvey

Schwandner FM, Gunson MR, Miller CE, Carn SA, Eldering A, Krings T, Verhulst KR, chimel DS, Nguyen HM, Crisp D, O'Dell CW, Osterman GB, Iraci LT Podolske JR (2017) Spaceborne detection of localized carbon dioxide sources. science, 358: eaam5782.

Scott S, Pfeffer MA, Oppenheimer C, Bali E, Lamb OD, Barnie T, Woods AW, Kjartansdóttir R, Stefánsson A, (2023) Near-surface magma flow instability drives cyclic lava fountaining at Fagradalsfjall, Iceland. Nature Communications, 14

Sleep NH, Zahnle K (2001) Carbon dioxide cycling and implications for climate on ancient earth. J Geophys Res 106:1373–1399

Sparks RSJ, Bursik M, Carey S, Gilbert J, Glaze L, Sigurdsson H, Woods A (1997) Volcanic plumes. Wiley

Stefánsson A, Hilton DR, Sveinbjörnsdóttir ÁE, Torssander P, Heinemeier J, Barnes JD, Ono S, Halldórsson SA, Fiebig J, Arnórsson S (2017) Isotope systematics of Icelandic thermal fluids. J Volc Geotherm Res 337:146–164

Wang H, Law AWK (2002) Second-order integral model for a round turbulent buoyant jet. J Fluid Mech 459:397–428

Werner C, Fischer TP, Aiuppa A, Edmonds M, Cardellini C, Carn S, Chiodini G, Cottrell E, Burton M, Shinohara H, Allard P (2019) Carbon dioxide emissions from subaerial volcanic regions. IN Deep carbon: past to present. Edited by Beth N. Orcutt, Isabelle Daniel, Rajdeep Dasgupta. Camb Univ Press

Werner C, Kelly P, Doukas M, Lopez T, Pfeffer M, McGimsey G, Neal C (2013) Degassing associated with the 2009 eruption of Redoubt Volcano, Alaska. J Volcanol Geoth Res 259:270–284

Johnson MS, Schwandner FM, Potter CS, Nguyen HM, Bell E, Nelson RR, Philip S, O'Dell CW (2020) Carbon dioxide emissions during the 2018 Kilauea volcano eruption estimated using OCO-2 satellite retrievals. Geophys Res Lett, 47(24): e2020GL090507

Maclennan J (2017) Bubble formation and decrepitation control the CO_2 content of olivine-hosted melt inclusions. Geochem Geophys Geosyst 18(2):597–616

Pering TD, Ilanko T, Liu EJ (2019) Periodicity in volcanic gas plumes: a review and analysis. Geosciences 9(9):394

White RS, Edmonds M, Maclennan J, Greenfield T, Agustsdottir T (2019) Melt movement through the Icelandic crust. Philos Trans Roy SoC: Math Phys Eng Sci 377(2139):20180010