Reykjanes Peninsula's historical eruptions: SO$_2$ emissions and future hazard implications

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Abstract

Exposure to volcanic SO\textsubscript{2} can have adverse effects on human health, with severe respiratory disorders documented on the short- and long-term timescale. Here, we use melt inclusion (MI) and groundmass glass data to calculate syn-eruptive SO\textsubscript{2} emission potentials across the Reykjanes peninsula (RP), the most populated area of Iceland that has recently undergone magmatic reactivation with the 2021, 2022, 2023 AD Fagradalsfjall eruptions. We target 16 individual eruptions from the previous volcanic episode at the RP, the 800-1240 AD Fires. Geochemical modelling indicates that pre-eruptive variability in sulfur contents can be explained by fractional crystallization and mixing of geochemically diverse mantle-derived melts. We calculate SO\textsubscript{2} emission potential across the RP to be in the range 0.004-7.4 Mt. These estimates correspond to daily SO\textsubscript{2} emissions in the range 600-53000 tons/day, higher than the mean SO\textsubscript{2} field measurements of 5240 ± 2700 tons/day during the 2021 AD Fagradalsfjall eruption. By using maximum and minimum sulfur values preserved in undegassed MIs, we develop an empirical approach to calculate best- and worst-case SO\textsubscript{2} emission potential scenarios of any past or ongoing RP eruption of known effusion rate. We conclude that the potential sulfur emissions across the RP can be significantly higher than observed during the 2021 AD Fagradalsfjall eruption, mainly because of the more evolved nature and higher sulfur contents of magmas erupted during the 800-1240 AD Fires. Based on dominant wind directions on the RP, eruptions in Brennisteinsfjöll pose the greatest health hazard to the capital area. Our findings enable assessing of SO\textsubscript{2} emission scenarios of future eruptions across the RP and can be used together with gas dispersal models to forecast SO\textsubscript{2} pollution at ground level and its impact on human health.

Introduction

The release of volcanic gases and aerosols during volcanic eruptions can significantly impact the air quality and climate (e.g., Ilyinskaya et al., 2017; Milford et al., 2023), as well as the biodiversity (e.g., Weiser et al., 2022). Among volcanic gases, sulfur species (SO\textsubscript{2}, H\textsubscript{2}S) and associated aerosols (SO\textsubscript{4}, H\textsubscript{2}SO\textsubscript{4}) are the most critical aerial hazard for human health, with short and long-term impacts that have been recorded at variable distances from eruptive vents (e.g., Ilyinskaya et al., 2017; Schmidt et al., 2015). For example, several studies have associated cardiorespiratory heath issues with volcanic sulfur emissions (e.g., Carlsen et al., 2021; Heaviside et al., 2021). Hence, a detailed knowledge of sulfur release potential of active volcanoes located in densely populated areas is critical to understand air quality hazards of future volcanic eruptions. This is the case of the Reykjanes peninsula (RP) in southwest Iceland, an active spreading area segmented into five volcanic systems that hosts ~70% of the Icelandic population. The latest magmatic period in the RP occurred ~800 years ago (Sæmundsson...
et al., 2020) but knowledge about sulfur outputs during those eruptions has been lacking thus far. Each
volcanic system on the RP tend to activate during individual magmatic periods (Sæmundsson et al.,
2020) and the recent 2021, 2022 and 2023 AD Fagradalsfjall eruptions (Barsotti et al., 2023; Einarsson
et al., 2023) suggest the potential initiation of a new magmatic period. Consequently, there is an
increased societal need for a deeper understanding of sulfur emissions across the RP. This is crucial for
a comprehensive assessment of sulfur’s impact during future eruptions across the RP and its potential
consequences for human health.
In this study, we calculate syn-eruptive sulfur release and sulfur emission potential of 16 geologically
and petrochemically well characterized magmatic units erupted in the volcanic systems of Reykjanes,
Svartsengi, Krysuvik and Brennisteinsfjoll during the 800-1240 AD Fires (Caracciolo et al., 2023;
Halldorsson et al., 2022; Peate et al., 2009) and compare those with sulfur emissions from 2021 AD
Fagradalsfjall eruption (Barsotti et al., 2023; Halldorsson et al., 2022). Also, we estimate daily SO2
emissions for the 800-1240 AD Fires and develop an empirical approach to calculate worst- and best-case sulfur emission potentials for any eruption of a given volume emplaced in the RP.

Samples and methods
Samples consist of scoria collected from multiple vents within individual eruptive units erupted during
the 800-1240 AD Fires (Caracciolo et al., 2023). Here, we present new S data for the same groundmass
glass (n=889) and plagioclase-, olivine- and clinopyroxene-hosted melt inclusions (MIs) (n=416) dataset
published in Caracciolo et al. (2023). Sulfur was analysed by electron microprobe analyser (EMPA) at
the University of Iceland using the same analytical settings as in Caracciolo et al. (2020) and MI
compositions have been corrected for post-entrapment processes (PEP) (Caracciolo et al. 2023).
In this work we make use of the ‘petrological method’ (Devine et al., 1984) to calculate eruptive sulfur
emissions by comparing the difference of S concentrations in mineral-hosted MIs with S concentrations
measured in groundmass glass. The idea behind this reconstruction method is that melt inclusions
preserve volatile concentrations at the time of trapping, namely the pre-eruptive volatile content,
whereas quenched groundmass glasses provide an estimate of the post-eruptive volatile content. For
the different magmatic units, the highest S concentration measured in PEP-corrected MIs (S_{MI}) is
selected as the pre-eruptive S concentration, whereas the lowest measurement in groundmass glasses
(S_{glass}) is chosen as post-eruptive S concentration. By combining the volume of erupted magmas with
\Delta S_{pre} [\Delta S_{pre} (ppm)= S_{MI} - S_{glass}], we can assess the syn-eruptive SO2 emission (\Delta S_{syn}, Mt) of individual
eruptions (e.g., Bali et al., 2018; Hartley et al., 2018). Furthermore, we calculate the SO2 emission
potential (\Delta S_{pot}, Mt), which refers to complete degassing of all pre-eruptive sulfur and reflects the
maximum amount of SO2 that a specific eruption could potentially have released. This reconstruction
method has been showed to have matched field-based volatile measurements exceptionally well during the 2014-15 Holuhraun eruption (Bali et al., 2018; Pfeffer et al., 2018) and the 2021 AD Fagradalsfjall eruption (this work, Table 1).

Sulfur concentrations in MI and groundmass glass

Sulfur concentrations in MIs are in the range 200-1900 ppm, with a relatively large variability of S at a given MI Mg#. Particularly, the most primitive MIs (Mg#>65), exclusively preserved in Reykjanes and Krýsuvík, record S contents in the range 580-1070 ppm (Fig. 1). Sulfur concentration in PEP-corrected MI compositions increases with decreasing MI Mg#, as expected for melt compositions controlled by fractional crystallization. Groundmass glasses with Mg# similar to the most evolved MIs have mean S contents varying between 150-450 ppm (Table 1). Groundmass glasses from Brennisteinsfjöll have mean S contents in the range 150-280 ppm, lower than mean S contents measured in glasses from the other volcanic systems (280-450 ppm) (Fig. 1a-d, Table 1). Sulfide globules were not observed in the erupted samples, neither within MIs nor as a stand-alone phase in the groundmass glass. For comparison, MIs from the 2021 AD Fagradalsfjall eruption contain maximum S concentrations of 1200 ppm, whereas the groundmass glasses contain 20-200 ppm S, significantly less than S concentrations preserved in MIs and groundmass glasses from the 800-1240 AD Fires (Fig. 1a-d).

Assessing sulfur variability and degassing during the 800-1240 AD Fires

Considering that historical eruptions on the Reykjanes Peninsula are likely sourced from mantle-derived melts of highly diverse compositions (Caracciolo et al., 2023; Harðardóttir et al., 2022; Halldórsson et al., 2022; Peate et al., 2009), including melts with variable sulfur contents (Ranta et al., 2022), we use our MI record to estimate S contents of the local enriched and depleted end-member melt components. We distinguish between these components from the K2O/TiO2 variability, which has been proved to be a robust tracer of mantle heterogeneities in Iceland and on the RP (Halldórsson et al., 2022; Harðardóttir et al., 2022) (see supplementary information). Starting from estimated end-member melt compositions and considering that sulfur behaves as an incompatible element in basaltic magmas, most of the MI sulfur variability in our dataset can be explained by fractional crystallization (FC) and mixing of at least two end-member melt compositions (Fig. 1a-d).

In order to evaluate S saturation during magma ascent and fractional crystallization through the crust, we calculate sulfur content at sulfide saturation (SCSS) along a FC path, which reflects the amount of $S^2$ present in a melt in equilibrium with a sulfide phase (Smythe et al., 2017) (see supplementary...
Our modelling suggests that melts are sulfide undersaturated during most of magmatic fractionation across the RP (Fig. 1). Only magmas from Svartsengi and Brennisteinsfjöll have a high likelihood to be sulfide saturated prior to eruptions (Fig. 1). Furthermore, sulfide saturation is reached earlier during magmatic differentiation of enriched mantle-derived melts than depleted melts. That is, magmatic differentiation of the depleted and enriched end-member melts leads to sulfide saturation at Mg#<50 and ~55-60, respectively.

Modelling of sulfur degassing with Sulfur_X (Ding et al. 2023) suggest that basaltic melts erupted during the 800-1240 AD Reykjanes Fires are unlikely to degas significant amounts of sulfur at known pre-eruptive magma storage depths (Caracciolo et al. 2023) and that S degassing only takes place during magma ascent in the last 0.2 kbar (<700 m) (Fig. S1).

### Sulfur emissions across the RP

Sulfur release ranges between 1000-1800 ppm across the RP, a typical range for Icelandic rift basalts (Ranta et al. 2022), with the largest $\Delta S$ found in lava flows from Svartsengi (SÖ lavas) and Hvammahraun (h128, Brennisteinsfjöll) (Table 1). $\Delta S$ values can be scaled by the mass of erupted material to estimate $\Delta S_{syn}$ of individual eruptions, using published volumes of individual eruptive units emplaced during the 800-1240 AD Fires (Table 1). In this case, lava flow volumes range between 0.02 km$^3$ to 0.72 km$^3$ (Einarsson et al., 1991; Jónsson, 1978; Sigurgeirsson, 2004). Using a melt density of 2700 kg/m$^3$ estimated with densityX program (Iacovino and Till, 2019), we calculate $\Delta S_{syn}$ between 0.004-7 Mt (Fig. 2a). The voluminous Arnarseturshraun (SÖ-A, 0.55 km$^3$) and Eldvarpahraun lava flows (SÖ-E, 0.28 km$^3$) in Svartsengi and Hvammahraun (h128, 0.72 km$^3$) and Tvíbollahraun (TV, 0.37 km$^2$) lava flows in Brennisteinsfjöll released the highest amount of SO$_2$ into the atmosphere during the 800-1240 AD Fires. The syn-eruptive SO$_2$ released by these latter voluminous lavas is approximately 2 to 7 times larger than syn-eruptive SO$_2$ emissions during the 2021 AD Fagradalsfjall eruption, for which we estimated $\Delta S_{syn} = 0.93$ Mt ($\Delta S_{measured} = 0.97 \pm 0.5$, Barsotti et al. 2023). These are roughly between 20 to 70 % of the syn-eruptive SO$_2$ emissions estimated for the 2014-15 Holuhraun eruption ($\Delta S_{syn} = 10.5$ Mt, Bali et al. 2018).

Similarly, we have calculated $\Delta S_{pot}$, the maximum amount of SO$_2$ that could potentially have been released during the 800-1240 AD Fires, assuming complete degassing of SMI. $\Delta S_{pot}$ ranges between 0.004-7.4 Mt and is only slightly higher than $\Delta S_{syn}$, as most of the sulfur is released into the atmosphere during eruptions on the RP rather than staying dissolved in the lava (Table 1).
Evaluating end-member scenarios of sulfur emissions and hazard potential for future eruptions across the RP.

Based on the MI record of the 2021 AD Fagradalsfjall eruption (Halldórsson et al., 2022) and the 800-1240 AD Fires (this work), we constrain maximum (1900 ppm) and minimum (1200 ppm) SO$_2$ emission potentials and use these to evaluate SO$_2$ emissions of future eruptions in the RP. With these constraints, we developed an empirical approach to assess $\Delta S_{pot}$ for a given eruption of known lava volume, with important applications for forecasting the worst- and best-case scenarios of $\Delta S_{pot}$ of future eruptive events (Fig. 2b). For example, based on our approach, an eruption with an eruptive volume of 0.4 km$^3$, has the potential to release a maximum and minimum of 4.1 Mt and 2.9 Mt SO$_2$, respectively. This method also has an application when it comes evaluating the long-term SO$_2$ impact of ongoing eruptions in the RP. If the mean magma effusion rate is known and fixed, one can roughly estimate the volume of the lava flow and calculate $\Delta S_{pot}$ at any given moment from the onset of the eruption. During an ongoing event, this can be a valuable tool to evaluate best- and worst-case scenarios for S pollution.

Eruptive S emission calculations are strongly dependent on lava flow volumes. Hence, when it comes to comparing the 800-1240 AD Fires with 2021 AD Fagradalsfjall eruption, a more relevant parameter is the mean daily SO$_2$ emissions, which also is an important parameter from a hazard perspective. First, we have estimated eruption durations for the 800-1240 AD Fires starting from published erupted volumes and using mean magma output rates (MOR) calculated by Oskarsson et al. (under review), in the range 1-66 m$^3$/s (Table 1). This yields eruption durations in the range 6 to 177 days. Secondly, we make use of inferred eruption durations to calculate daily SO$_2$ emissions and speculate about potential daily S releases during future eruptive scenarios. Daily SO$_2$ emissions during the 800-1240 AD Fires likely ranged between 600 ton/day to 53000 ton/day (Fig. 2c). In comparison, during the 2021 AD Fagradalsfjall eruption, if we assume a total amount of SO$_2$ equal to 0.97 ± 0.5 Mt (Barsotti et al., 2023), we calculate average daily SO$_2$ emissions of 5240 ± 2700 ton/day. This estimate is in agreement with the majority of measured daily SO$_2$ emissions throughout the 2021 AD Fagradalsfjall eruption, in the range 1000-7600 ton/day (Esse et al., 2023). Hence, future eruptions in the RP have the potential to release significantly more SO$_2$ on a daily basis than the 2021 AD Fagradalsfjall eruption.

SO$_2$ emissions during the 800-1240 AD Fires and 2021 AD Fagradalsfjall are small compared to those during the recent basaltic eruption of Holuhraun (9.2 Mt SO$_2$, Pfeffer et al., 2018). However, volcanic eruptions from the RP are potentially considered to be more hazardous due to their proximity to inhabited areas, to the international airport and to the large number of visitors expected at eruption sites (Fig. 3) (Barsotti et al. 2023). To assess the health hazard for potential future eruptions, we built seasonal wind roses, for the period 2012-2022, reflecting dominant wind speeds and directions in the
RP. We find that most of the time the winds blow in a direction spanning from the NW to the NE, suggesting different SO$_2$ health hazards potential associated with eruptions within different volcanic systems. Eruptions in Brennisteinsfjöll are the most hazardous, especially in spring and autumn seasons, as SO$_2$ is likely to be blown towards the capital area. Eruptions in Reykjanes pose minimal hazard as winds tend to blow away from inhabited areas. During assessment of possible eruptive scenarios in the RP, our estimates can be key input parameters to model the release and dispersion of volcanic SO$_2$ into the atmosphere. Our results can be used to inform SO$_2$ pollution hazard assessments for potential eruptive scenarios and prompt action and mitigation plans during ongoing volcanic crises in the RP.

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Table 1. Eruptive units studied in this work and summary of main results. $S_{MI}$ = pre-eruptive S concentration. $S_{glass}$ = post-eruptive S concentration. $\Delta S$ = sulfur emissions emitted at the vent. $\Delta S_{syn}$ = Syn-eruptive SO$_2$ emissions. $\Delta S_{pot}$ = SO$_2$ emission potential. Eruption durations and daily SO$_2$ emissions are calculated assuming a bulk effusion rate of 9.5 m$^3$/s, as observed during the 2021 Fagradalsfjall eruption (Pedersen et al. 2022). $^a$ Calculations for the 2021 AD Fagradalsfjall eruption are from this work and based on MI and glass data from Halldórsson et al. 2022. Estimated $\Delta S_{syn}$ of 0.93 Mt from this work is in agreement with measured total SO$_2$ emissions of 0.97±0.5 Mt (Barsotti et al. 2023). $^b$ To our knowledge, volumes for Hrútafellshraun (HRF), Svartihryggur (h142), Húsfellsbruni (Hú1 & Hú2) and Kristnitökuhraun (KRT) lava flows are not available in the literature. We estimated the volumes of those lava flows by multiplying their area by an average thickness of 5 m, consistent with average thicknesses of lava flows of known volumes with a similar aerial extent. $^c$ MOR data are from Óskarsson et al. (under review). MOR for the 2021 Fagradalsfjall eruption is from Pedersen et al. (2022).

Fig. 1. (a-d) Variation of S contents in groundmass glasses (filled circles) and PEC-corrected MIs (filled triangles) as a function of Mg# $[\text{Mg# } = 100\cdot \text{Mg}/(\text{Mg}+\text{Fe}^{2+})$, $\text{Fe}^{2+}/\text{Fe}_{\text{tot}} = 0.9]$ in samples from the 800-1240 AD Fires. Data from the 2021 AD Fagradalsfjall eruption are from Halldórsson et al. (2022). Red and blue solid lines indicate fractional crystallization paths calculated for a geochemically enriched and depleted initial melt compositions, respectively (see supplementary text). The black dotted curve indicates SCSS along an empirical fractional crystallization path calculated after Smythe et al., (2017). Data are grouped in different panels according to the volcanic system and coloured according to the lava unit. (e-h) Measured MI S contents vs calculated SCSS, coloured after MgO content. SCSS was
calculated assuming $\text{Fe}^{3+}/\text{Fe}_{\text{tot}} = 0.1$, $P = 2$ kbar, $T=1220^\circ\text{C}$ and sulfide $\text{Fe}/(\text{Fe}+\text{Ni}+\text{Cu}) = 0.65$, after the method of Smythe et al., (2017) (cf. Fig. S3). All SCSS models were implemented in PySulfSat code (Wieser and Gleeson, 2022).

Fig. 2. (a) West to east variation of $\Delta S_{\text{syn}}$ calculated for the different eruptive units. (b) $\Delta S_{\text{pot}}$ as a function of eruption volume for the 800-1240 AD Fires and 2021 Fagradalsfjall eruption. At a given volume, straight lines allow to calculate $\Delta S_{\text{pot}}$ corresponding to maximum (1900 ppm) and minimum (1200 ppm) $S_{\text{MI}}$ values measured across the RP. Inset plot show most common $\Delta S_{\text{pot}}$ across the RP, in the range 0-2 Mt and for lava volumes between 0-0.3 km$^3$. (c) Maximum and minimum daily SO$_2$ emissions calculated for each volcanic system during the 800-1240 AD Fires and for the 2021 AD Fagradalsfjall (see text for calculation details). Blue kernel density estimate curve indicate measured SO$_2$ emission rates during the 2021 AD Fagradalsfjall eruption (Esse et al., 2023). Labels indicate acronyms used for the different eruptive units (c.f. Table 1).

Fig. 3. Simplified geological map of the Reykjanes Peninsula and lava flows emplaced during the 800–1240 AD Fires. The map also illustrates the aerial extent of the 2021 (Pedersen et al., 2022), 2022 (Gunnarson et al., 2023) and 2023 AD (Belart et al. 2023) Fagradalsfjall lavas. The lava flows are colored according to calculated syn-eruptive SO$_2$ emissions, ranging from 0.1 to 7 Mt. Filled, white circles indicate the location of main urban areas in the Reykjanes Peninsula. Geological data are from Iceland GeoSurvey (ÍSOR) (Sæmundsson et al., 2016). Seasonal wind roses reflect data at 900 mPa (~1000 m a.s.l), which is the most common SO$_2$ injection altitude throughout the 2021 Fagradalsfjall eruption (Esse et al. 2023). Spokes indicate the direction the wind is blowing from, and the length of each spoke shows the frequency. Wind data were extracted from ERA5 (Hersbach et al. 2023).

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