Medieval and recent SO$_2$ budgets in the Reykjanes Peninsula: implication for future hazard

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Supplementary Information

The Supplementary Information includes:

- Calculation of SO$_2$ Emissions
- Modelling Sulfur Degassing
- The Occurrence of Degassed and Partially Degassed Melt Inclusions
- Selection of Enriched and Depleted End Member Melt Compositions
- Modelling SCSS
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Calculation of SO$_2$ Emissions

The total mass of sulfur ($M_S$, [kg]) which was released from the magma and dispersed into the atmosphere can be calculated as:

$$M_S = V_{DRE} \times \rho \times \Delta C_S ,$$  \hspace{1cm} (S-1)

where $V_{DRE}$ is the dense rock equivalent volume (vesicle-free) of the erupted lava unit in m$^3$, $\rho$ is the density of the melt in kg/m$^3$ and $\Delta C_S$ the mass of sulfur release per unit mass of melt. Particularly, $\Delta C_S$ is calculated as:

$$\Delta C_S = (1 - X) (C_{S_{MI}} - C_{S_{glass}}) ,$$  \hspace{1cm} (S-2)

where $X$ is the crystal content of the lava, which is set at 0.02. This is a reasonable and conservative assumption as the 800–1240 AD Fires contain little amount of macrocrysts, <2 vol. % (Caracciolo et al., 2023). $C_{S_{MI}}$ and $C_{S_{glass}}$ are the pre-eruptive and post-erupted sulfur (S) concentrations for a given eruption, respectively, which in this work were chosen as the maximum and minimum S concentrations measured in MIs and in tephra glasses, respectively (cf. Table 1). The volume of the erupted lava unit is expressed as dense rock equivalent ($V_{DRE}$), which reflects the volume of the vesicle-free lava. However, published lava volumes for the 800–1240
AD Fires refer to bulk volumes and do not take into account the porosity of the lava. For this reason, we estimated $V_{\text{DRE}}$ by multiplying the bulk volume ($V$) for a scaling factor that accounts for vesicularity (0.85 in this work). Due to the lack of vesicularty data for the lava units targeted in this study and considering that vesicularity is highly variable across basaltic lava flows (e.g., Cashman and Kauahikaua, 1997), we use a bulk vesicularity of 15%, consistent with findings for the 2014–2015 Holuhraun lava flow (Bonny et al., 2018). This bulk vesicularity is within the range of vesicularity constrained for other basaltic lava flows in Iceland, such as Hafnarhraun in the Reykjanes peninsula (12–37% vesicularity; Nikkola et al., 2019) and Vikrahraun, the 1961 lava flow from Askja (0–30% vesicularity; Blasizzo et al., 2022).

Assessing the daily SO$_2$ emissions of past eruptions is challenging, particularly for those eruptions where the duration is unknown, such as for the 800–1240 AD Fires. We calculated daily SO$_2$ emissions [kg/d] starting from MOR values using the following equation:

$$ \text{Daily SO}_2 \text{ emissions} = \text{MOR}_i \times \rho \times (\Delta C_S \times 2) \times 86,400 . \quad (S-3) $$

Additionally, in Table 1 we included the amount of time required to erupt and emplace a specific volume of magma, which can be used as a proxy for eruption durations. We call this time of lava emplacement. In fact, we cannot rule out that erupted volumes reflect multiple eruptive phases over which the lava was emplaced, as observed during the 2023–2024 Svartsengi eruptions (<48 h) and as suggested by Caracciolo et al. (2023). The time of lava emplacement ($t_m$[days]) of each eruptive unit was calculated by using MOR values calculated by Óskarsson et al. (2024) (Table 1), using the following equation:

$$ t_m = \frac{V_i}{\text{MOR}_i \times 86,400} , \quad (S-4) $$

where $V_i$[m$^3$] and MOR$_i$[m$^3$/s] are the bulk volume and the MOR of a specific lava unit, respectively. The time of lava emplacement for the new 2021–2024 eruptions at Fagradalsfjall and Svartsengi represents the actual duration of each eruption. Óskarsson et al. (2024) calculate mean output rates (MOR) and uncertainty ranges for each eruption targeted in this work. Therefore, the time of lava emplacement and daily SO$_2$ emissions for each eruption was calculated taking into account the uncertainty range in MOR values (Table 1).

### Modelling Sulfur Degassing

Sulfur degassing was modelled using the open source COHS-degassing model Sulfur X (Ding et al., 2023), using the COH model of Newman and Lowenstern (2002) and the S speciation model of O’Neill and Mavrogenes (2022). Oxygen fugacity was set at $\Delta$FMQ = 0 (Novella et al., 2020) and $T = 1200$ °C. Pre-eruptive S concentration was chosen as 1550 ppm based on MI data, whereas H$_2$O and CO$_2$ concentrations were estimated at 0.3 wt. % and 1000 ppm, respectively, in agreement with typical H$_2$O and CO$_2$ concentrations of Icelandic rift basalts (e.g., Bali et al., 2018; Halldórsson et al., 2022). Melt composition was chosen as the mean groundmass glass compositions of the 800–1240 AD Fires. We also tested S degassing of hypothetical scenarios with higher melt H$_2$O contents of 1.5 wt. % and 3 wt. %, unrealistically high for Iceland rift basalts (Ranta et al., 2024). Higher H$_2$O contents push the onset of S degassing to higher pressures.
However, >3 wt. % H₂O contents are required for significant S degassing to occur at known magma storage depths.

Figure S-1    Models of melt S degassing as a function of pressure. Thick black curve indicates realistic H₂O contents for Reykjanes basalts. Dashed grey lines indicate S degassing assuming unrealistically high H₂O contents of 1.5 and 3 wt. %. Modelling was carried out using Sulfur_X (Ding et al., 2023).

The Occurrence of Degassed and Partially Degassed Melt Inclusions
Many melt inclusion compositions from Brennisteinsfjöll, and to a lesser extent, from Svartsengi, are found to be partially to fully degassed (Fig. 1). In the case of Brennisteinsfjöll, in which we have a larger amount of partially and fully degassed MIs, individual crystals contain multiple MIs with variable S contents. Within the same crystal, some MIs have ‘normal’ S contents (>1200 ppm) consistent with fractional crystallisation trends, whereas some are partially (400–1100 ppm S) degassed (Fig. 1). Additionally, some MIs are completely degassed, with S contents similar to groundmass values (<400 ppm) (Fig. 1d). Based on backscattered electron (BSE) images, those MIs that are partially to completely degassed contain very large bubbles (Fig. S-2). Post-entrapment degassing at low-P (e.g., during eruption) could be responsible for the observed partially to completely degassed MIs in Brennisteinsfjöll. The MIs could release S at low pressure, such as when a host
crystal fractures during an eruption, causing the pressure within certain MIs to rapidly decrease to nearly atmospheric. In this scenario, the melt within the MIs degasses sulfur into the shrinkage bubble prior to quenching.

Figure S-2  BSE images of (a–d) plagioclase, (e) clinopyroxene and (f) olivine crystals from Brennisteinsfjöll containing multiple MIs with different S contents. Note that partially and completely degassed MIs contain large bubbles, which probably contained S-bearing mineral precipitates which sequestered a significant amount of S.
Selection of Enriched and Depleted End Member Melt Compositions

End member melt compositions were selected from the MI record preserved in the 800–1240 AD Fires and using K₂O/TiO₂ as a proxy for magma enrichment, which has been proven to be a robust tracer of mantle-derived chemical variability (Halldórsson et al., 2022; Harðardóttir et al., 2022). For the depleted end member melt composition, the MI dataset was filtered by selecting and averaging PEP-corrected MI compositions (n = 3) satisfying the following criteria: K₂O/TiO₂ < 0.1, Mg# > 70 and MgO > 9 wt. %. The resulting melt composition has Mg# = 70.4, 9.5 wt. % MgO and S content of 680 ± 85 ppm. The enriched end member melt composition was chosen by averaging MI compositions (n = 4) satisfying the following criteria: Mg# > 69, MgO > 9 wt. %, K₂O/TiO₂ > 0.3 and TiO₂ > 0.4 wt. %. The resulting composition has Mg# = 70, 9.8 wt. % MgO and S content of 872 ± 166 ppm. Starting from these initial compositions, we modelled fractional crystallisation using Petrolog3 (Danyushevsky and Plechov, 2011) at 2 kbar and at fO₂ corresponding to the FMQ buffer. S partition coefficient between melt and olivine, plagioclase and clinopyroxene were taken from Callegaro et al. (2020) (S partition coefficient of Cpx–melt = 0.05, Olivine–melt = 0.01, Plagioclase–melt = 0.13).

Modelling SCSS

Sulfur concentration at sulfide saturation (SCSS) was modelled along an empirical fractional crystallisation path calculated between Mg# = 75 and Mg# = 40 by regressing the observed MI and glass data with linear functions for each volcanic system. Melt temperature was calculated using the melt-only thermometer (Eq. 14 in Putirka, 2008). SCSS modelling was implemented in PySulfSat (Wieser and Gleeson, 2022), by comparing different SCSS models (Fortin et al., 2015; Smythe et al., 2017; O’Neill and Mavrogenes, 2022) (Fig. S-2). For the modelling parameters, we used Fe³⁺/Fe₄⁰⁺ = 0.1, P = 2 kbar and sulfide Fe/(Fe+Ni+Cu) = 0.65. The different models yield similar results and well within the 1σ uncertainty of the Smythe et al. (2017) model (see Fig. S-3a–d).
Figure S-3  Variation of S contents in groundmass glasses (filled circles) and PEP-corrected MIs (filled triangles) as a function of Mg# in samples from the 800–1240 AD Fires and the 2021 Fagradalsfjall eruption. The purpose of the figure is to compare results from different SCSS models, indicated with different dashed curves.

Figure S-4 [next page]  (a–d) Variation of S contents in groundmass glasses (filled circles) and PEP-corrected MIs (filled triangles) as a function of Mg# [Mg# = 100·Mg/(Mg+Fe<sup>2+</sup>), Fe<sup>2+</sup>/Fe<sup>tot</sup> = 0.9] in samples from the 800–1240 AD Fires and raw MI data from the December 2023, January 2024 and February 2024 Sundhnúkuar eruptions. Data from the 2021 AD Fagradalsfjall eruption are from Halldórsson et al. (2022). Red and blue solid lines indicate fractional crystallisation paths calculated for a geochemically enriched and depleted initial melt compositions, respectively. The black dashed curve indicates SCSS along an empirical fractional crystallisation path calculated after Smythe et al. (2017). (e–h) Measured MI S contents vs. calculated SCSS, coloured after MgO content. SCSS was calculated assuming Fe<sup>3+</sup>/Fe<sup>tot</sup> = 0.1, P = 2 kbar, T = 1220 °C and sulfide Fe/(Fe+Ni+Cu) = 0.65, after the method of Smythe et al. (2017) (cf. Fig. S-3). All SCSS models were implemented in PySulfSat code (Wieser and Gleeson, 2022).
Reykjanes

S (ppm)

Glass or MI Mg#

0 200 400 600 800 1000 1200 1400 1600 1800 2000 2200 2400

Partially to completely degassed MILs

Svartsengi

S (ppm)

Glass or MI Mg#

35 40 45 50 55 60 65 70 75

Glass or MI Mg#

35 40 45 50 55 60 65 70 75

Krysuvík

S (ppm)

Glass or MI Mg#

35 40 45 50 55 60 65 70 75

Glass or MI Mg#

35 40 45 50 55 60 65 70 75

Brendistólsfjöll

S (ppm)

Glass or MI Mg#

35 40 45 50 55 60 65 70 75

Glass or MI Mg#

35 40 45 50 55 60 65 70 75

Hrafnahraun (h128)

Hvíldfellacrusta (KVT)

Kotul-Selvog-Svartsílv lavas (h130, h138, h142)

Hvíldfellacrusta (KVT)

Kotul-Selvog-Svartsílv lavas (h130, h138, h142)

Brendistólsfjöll

S (ppm)

Glass or MI Mg#

35 40 45 50 55 60 65 70 75

Glass or MI Mg#

35 40 45 50 55 60 65 70 75

Hrafnahraun (h128)

Hvíldfellacrusta (KVT)

Kotul-Selvog-Svartsílv lavas (h130, h138, h142)

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Supplementary Tables

The MI and glass dataset published in this work complement the dataset published in Caracciolo et al. (2023) and here includes S and Cl measurements. Re-homogenized melt inclusions are not included in this work. Additionally, the dataset includes new groundmass and MI data from the data from the 2022–2023 Fagradalsfjall eruptions and the 2023–2024 eruptions at Sundhñoakjígar in Svartsengi.

**Table S-1** Overview of samples studied in this work, along with acronyms, lava flows, ages and coordinates. Analysed phases in each sample are indicated with cross marks.

**Table S-2** Groundmass glass dataset.

**Table S-3** Mean compositions of groundmass glasses and glass standards.

**Table S-4** Melt inclusion dataset listing PEP-corrected and raw compositions, along with host mineral compositions and 1σ uncertainties. Note that MI s from the 2023 Fagradalsfjall eruption and the December 2023 and January 2024 Sundhñoakjígar eruptions have not been PEP-corrected.

Tables S-1 to S-4 are available for download (.xlsx) from the online version of this article at http://doi.org/10.7185/geochemlet.2417.

Supplementary Information References


