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*Geochemistry, Geophysics, Geosystems*

Supporting Information for

**Magmatic Controls on Volcanic Sulfur Emissions at the Iceland Hotspot**

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## Introduction

This file contains an overview of the non-S volatiles and the Supplementary Figures S1-S5. Supplementary Tables S1-S8 are provided as separate Excel-files.

### S1. H<sub>2</sub>O, CO<sub>2</sub>, F and Cl concentrations of Icelandic melt inclusions

Water contents of Icelandic melt inclusions vary from <0.1 wt.% in trace element-depleted rift basalts to up to 6.5 wt.% in silicic melt inclusions from Hekla. Overall, H<sub>2</sub>O concentrations increase with increasing SiO<sub>2</sub> (Fig. 3a) and are lowest in basaltic MIs (0-1.5 wt.%), moderate in intermediate MIs (0.7-4 wt.%) and highest, but also highly variable in silicic melts (0.1 to 6.5 wt.%). Among basalts, highest H<sub>2</sub>O contents are seen in flank zone eruptions of Surtsey 1963-67 ( $1.2 \pm 0.1$  wt.%,  $1\sigma$ ) and Fimmvörðuháls 2010 ( $0.9 \pm 0.1$  wt.%) in the SIVZ and in the Berserkjahraun eruption of Ljósufjöll ( $0.7 \pm 0.3$  wt.%) in SNVZ. Rift basalts tend to have lower MI H<sub>2</sub>O contents, for example  $0.36 \pm 0.14$  wt.% in Holuhraun 2014-15, and  $0.23 \pm 0.02$  wt.% in Fagradalsfjall 2021. The lowest MI H<sub>2</sub>O contents are observed in highly trace-element depleted, high-MgO (> 9 wt.%) basalts such as Miðfell ( $0.07 \pm 0.01$  wt.%) in the WRZ and Heilagsdalsfjall of Fremrinámar in the NRZ ( $0.05 \pm 0.01$  wt.%). Silicic MI H<sub>2</sub>O contents from the Askja 1875 eruption vary from 0.6 to 2.7 wt.% (excluding outliers) with an average of  $1.6 \pm 0.7$  ‰. Compared to Askja, 2-3 times higher average MI H<sub>2</sub>O contents are found in the silicic Hekla eruptions 1104 ( $3.1 \pm 1.2$  wt.%), H3 ( $3.5 \pm 1.2$  wt.%) and H4 ( $5.7 \pm 0.6$  wt.%), and the rhyolitic Öraefajökull 1362 ( $3.1 \pm 0.6$  wt.%) eruption. Of intermediate Hekla eruptions, the andesitic 1845 and the 1991 basaltic andesite eruptions have MI H<sub>2</sub>O contents of  $2.1 \pm$  wt.% and  $1.2 \pm 0.4$  wt.%, respectively.

The CO<sub>2</sub> concentrations in Icelandic MIs vary between 0 and 5960 ppm (Fig. 3b), but rarely exceed 3000 ppm. Only basaltic MIs show elevated CO<sub>2</sub> contents, whereas, as a rule, intermediate and silicic MIs have low CO<sub>2</sub> contents (< 50 ppm) that are indistinguishable from matrix glasses. It should be noted that in bubble-bearing melt inclusions, vapor bubbles may host close to 100% of the total MI CO<sub>2</sub> content, but only a few studies have measured and quantified the CO<sub>2</sub> content in the bubbles (Hartley et al., 2014; Neave et al., 2014; Bali et al., 2018). Thus, apparent CO<sub>2</sub> systematics are likely to be affected by sporadic underestimation of true MI CO<sub>2</sub> contents.

Fluorine contents of basaltic MIs range from < 50 ppm in primitive (> 10 wt.% MgO) rift zone basalts to 2850 ppm in the SNVZ Berserkjahraun eruption (Fig. 4c). Apart from Berserkjahraun, all F concentrations in basaltic MIs are below 1000 ppm. Intermediate MIs have between 980-2090 ppm F. Silicic MIs have highly variable F contents from 100 to 2200 ppm. The silicic Askja 1875 MIs lack

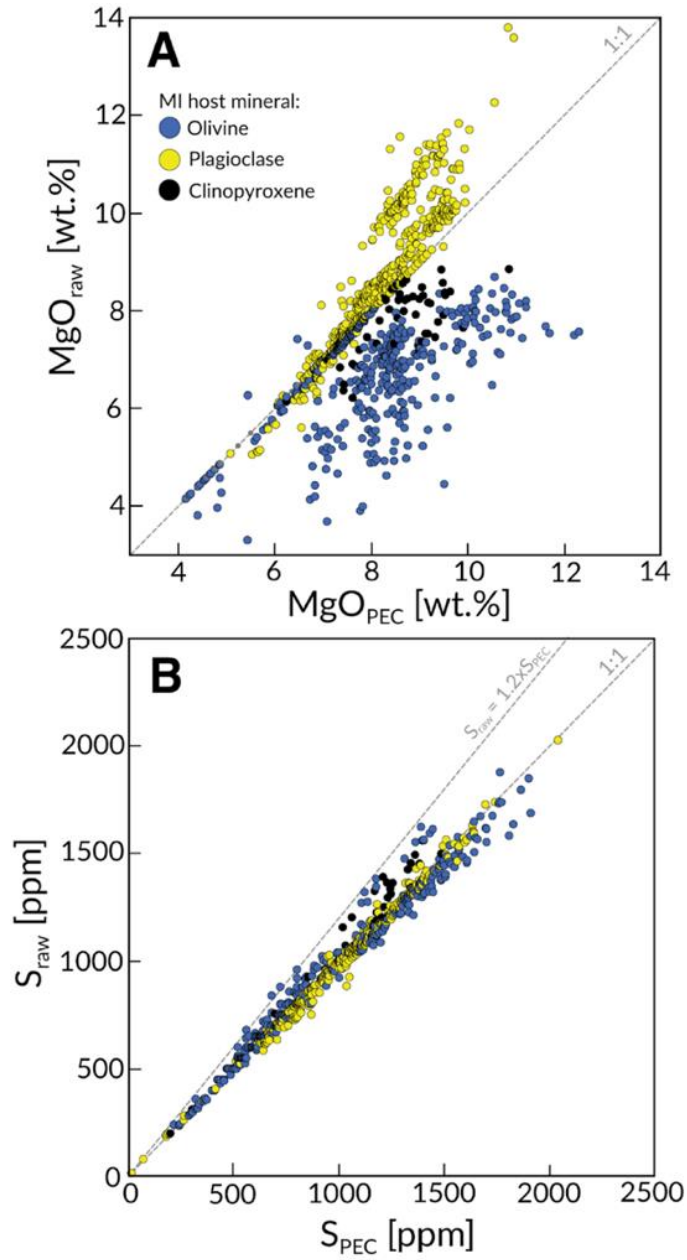
the high F contents present in the silicic eruptions of Hekla and Öraefajökull with a range of 200-600 ppm. On average, there is no significant difference between matrix glass and MIs in either average F contents or variability. Fluorine concentrations in plagioclase-hosted MIs are not considered here and are excluded from Figure 3, as they commonly show anomalously high concentrations that reflect the crystallization process rather than the pre-eruptive melt (Neave et al., 2017).

Basaltic MI Cl concentrations vary between 3 and 1900 ppm (Fig. 3d), excluding a single Fimmvörðuháls MI with 2500 ppm. Intermediate MIs (only available from Hekla) have a rather limited Cl range of 210-790 ppm. Silicic MIs have the highest, but most variable Cl contents of 0-2400 ppm.

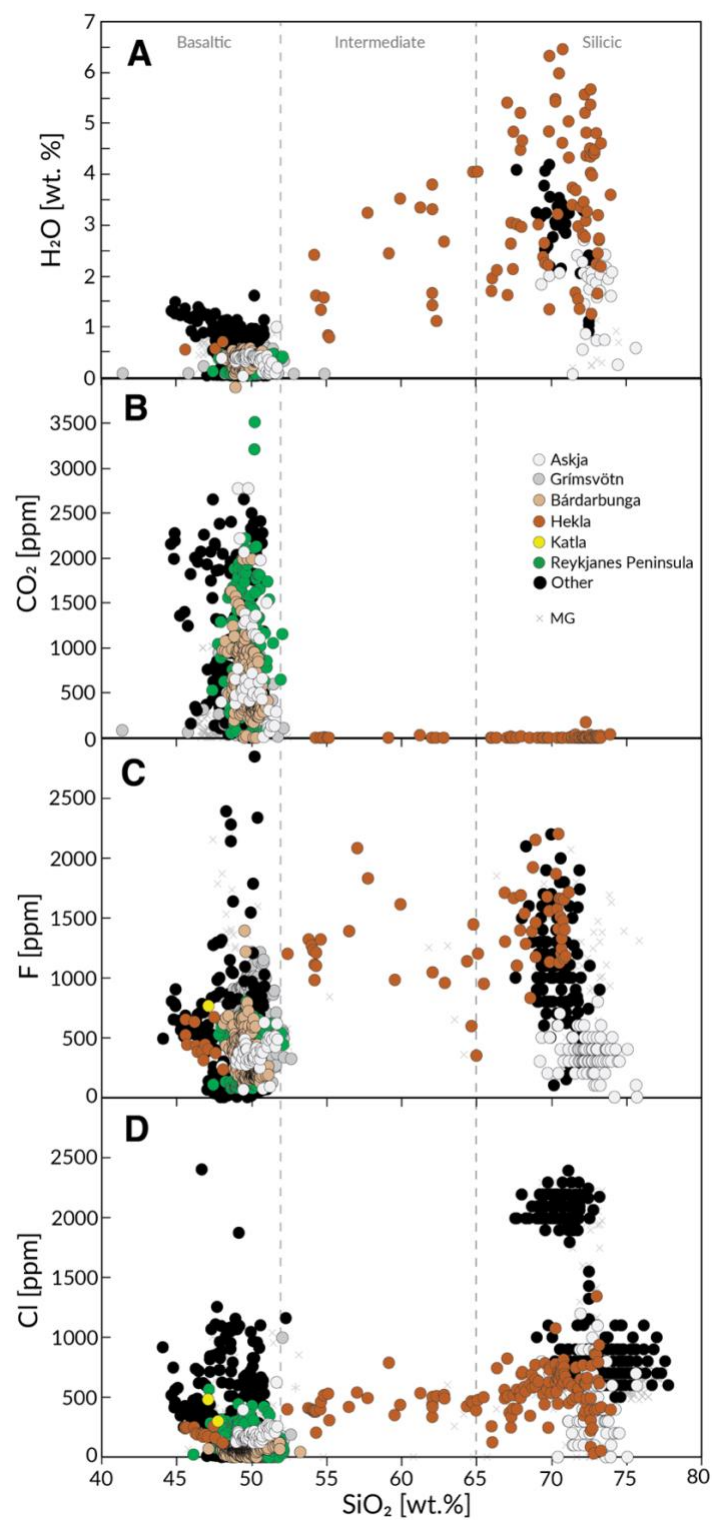
## **S2. Effects of crustal magma evolution on H<sub>2</sub>O, CO<sub>2</sub>, F and Cl**

Because of the low solubility of CO<sub>2</sub> at crustal pressures and inferred high CO<sub>2</sub> contents of primary melts, virtually all Icelandic basalts saturate a CO<sub>2</sub>-rich vapor phase already at great depth, with only a few possible exceptions (e.g., Hauri et al., 2018; Miller et al., 2019; Matthews et al., 2021; Ranta et al., 2023). As a result, the highest CO<sub>2</sub> contents are typically found in primitive MIs, but are negligible in intermediate and silicic rocks (Fig. 4b). At the other end of the solubility spectrum, H<sub>2</sub>O, Cl and F remain undersaturated in basaltic melts at typical Icelandic magma storage pressures, leading to increasing concentrations with decreasing MgO observed in basaltic pillow rim glasses (Nichols et al., 2002; Halldórsson et al., 2016b) and MIs. For silicic rocks, the H<sub>2</sub>O-F-Cl systematics are more complicated. Silicic MIs of single eruptions can have highly variable H<sub>2</sub>O-F-Cl contents due to the chained effects of a complicated magmatic history involving fluid exsolution, brine assimilation, recharge, gas fluxing and crustal assimilation that may occur in long-lived (>100 ka) silicic magma mushes in Iceland (Gunnarsson et al., 1998; Schattel et al., 2014; Padilla et al., 2016; Ranta et al., 2021). These effects are smaller at off-rift settings, where silicic magma genesis may approximate closed system fractional crystallization paths (Martin & Sigmarsson, 2007; Schattel et al., 2014), resulting in higher concentrations and smaller variability of H<sub>2</sub>O, F and Cl. Opposing behavior is sometimes observed between H<sub>2</sub>O and the halogens; for example, silicic Hekla MIs have the highest H<sub>2</sub>O concentrations in Iceland, but are the most Cl poor (Fig. 4a, c; Ranta et al., 2021). In summary, the volatile evolution of silicic magmas appears more complicated and is affected by more processes than that of basalts.

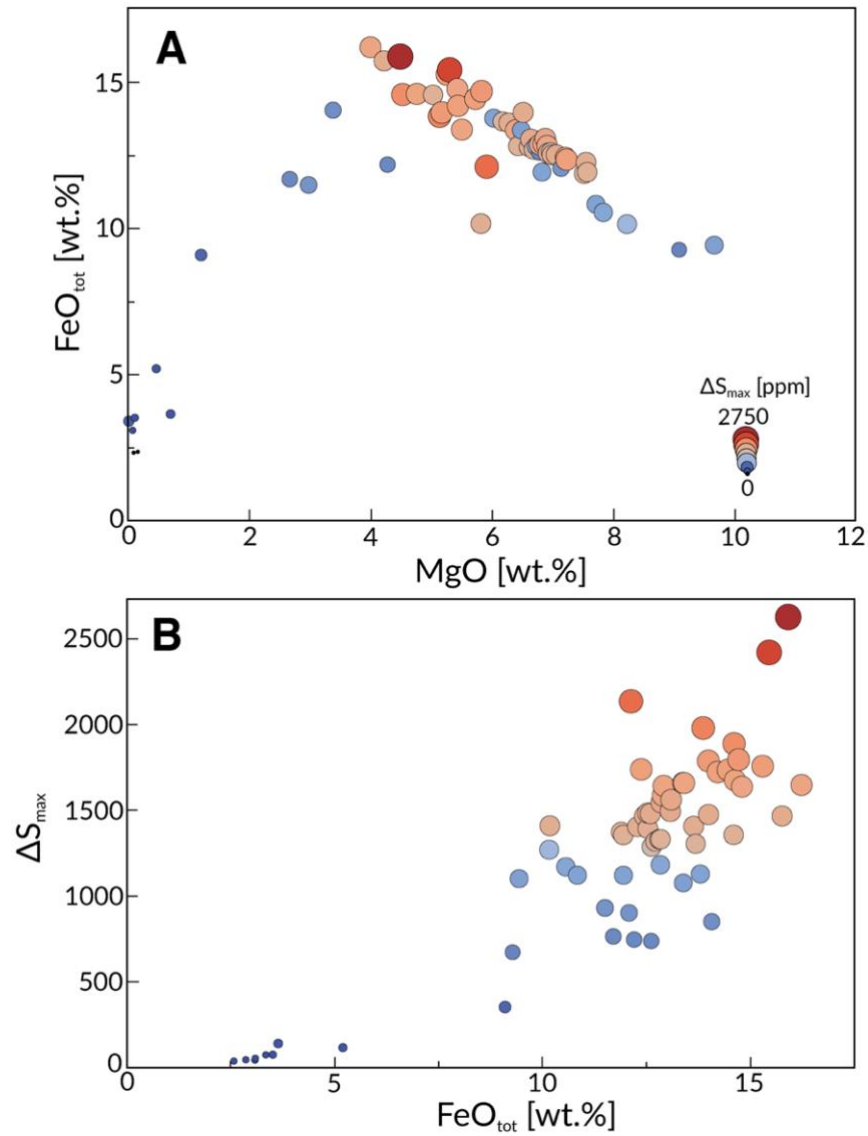
## Supplementary Figures



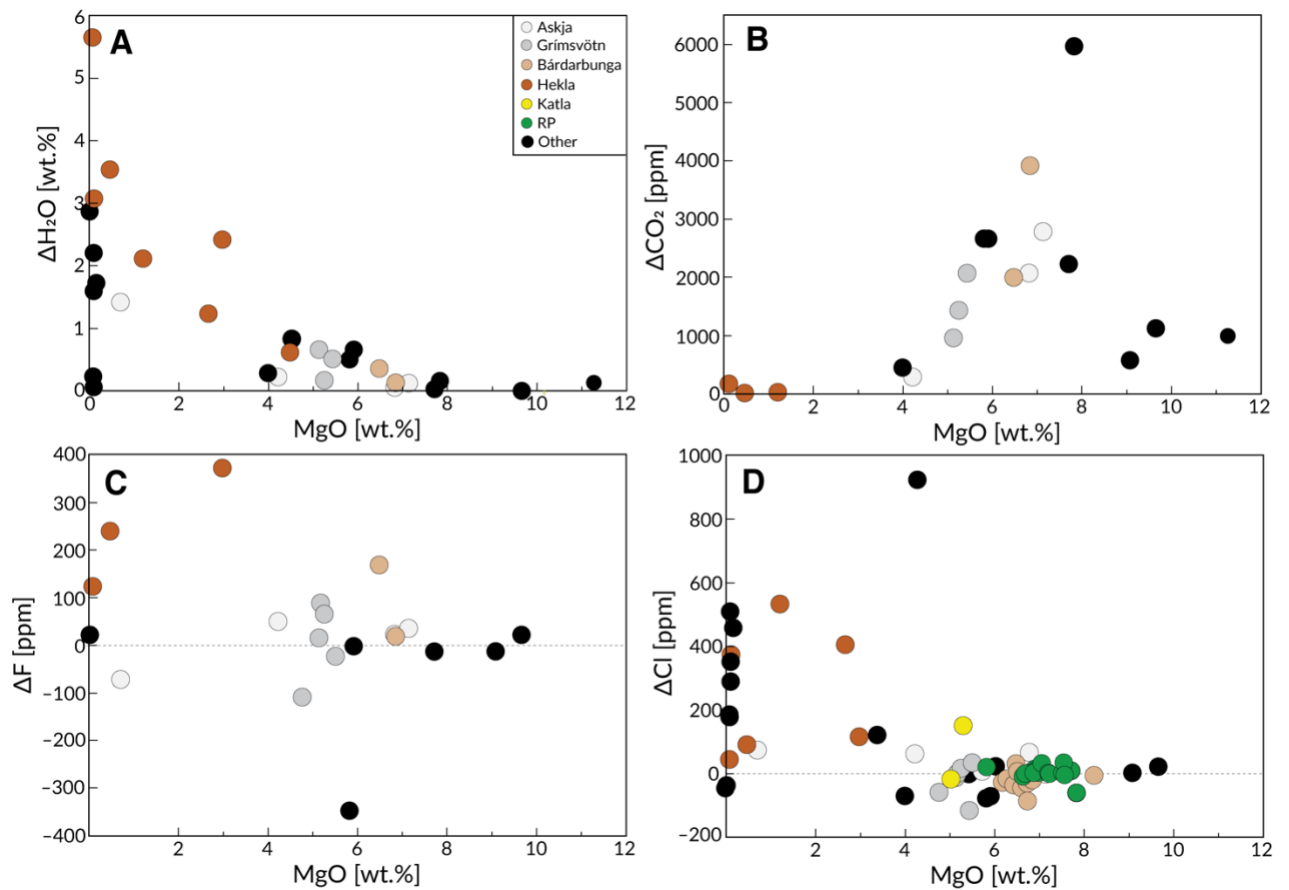
**Figure S1.** Effects of post-entrapment processes on (a) MgO, and (b) S concentrations of melt inclusions. Subscripts 'raw' and 'PEC' refer to uncorrected raw data and PEP-corrected data, respectively.



**Figure S2.** Melt inclusion volatile contents.  $\text{SiO}_2$  plotted against (a)  $\text{H}_2\text{O}$ , (b)  $\text{CO}_2$ , (c) F and (d) Cl. Data from the Iceland Melt Inclusion Catalogue (IMIC; Table S1)



**Figure S3.** Iron and sulfur. (a)  $\text{FeO}_{\text{tot}}$  vs.  $\text{MgO}$ . (b)  $\Delta S_{\text{max}}$  vs.  $\text{FeO}_{\text{tot}}$ .  $\text{FeO}$  content of melts is one of the main controls on sulfur solubility (Smythe et al., 2017), resulting in a broad correlation between  $\Delta S_{\text{max}}$  and  $\text{FeO}_{\text{tot}}$ . However, large variability ( $> 100\%$ ) in  $\Delta S_{\text{max}}$  is observed at a given  $\text{FeO}_{\text{tot}}$ , demonstrating the juxtaposed effects of pressure, temperature and  $f\text{O}_2$  on the SCSS, as well as the likely presence of sulfide-undersaturated melts at higher  $\text{MgO}$ . Circles represent individual eruptions and are shaded from blue to red and increase in size with increasing  $\Delta S_{\text{max}}$ .



**Figure S4.** Application of the petrological method for (a)  $\text{H}_2\text{O}$ , (b)  $\text{CO}_2$ , (c) Cl and (d) F vs. MgO. The volatile emission estimates for these volatiles are not considered to be as accurate as for sulfur (see Section 5)