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This pre-print is not an accepted peer-reviewed manuscript. A version of this pre-print has been submitted to Journal of Petrology for peer-review after a round of reviews.

Dilution drives deep degassing of sulfur in hydrous basaltic magmas

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19 Sulfur (S) is thought to degas at higher pressures from hydrous magmas (e.g., arc and
20 some ocean island basalts) relative to water-poor magmas, from which S degasses at lower
21 pressures (e.g., Kīlauea, mid-ocean ridge basalts). Our modelling of degassing shows this
22 occurs for magmas that are either reduced (i.e., S is present predominantly as H₂S in the
23 vapor and dissolved sulfide in the melt) or oxidised (i.e., dominantly SO₂ in the vapor and
24 dissolved sulfate in the melt). Degassing at even higher pressures occurs for silicate melts
25 containing both dissolved sulfide and sulfate due to the “sulfur solubility minimum”.
26 Additionally degassing of a fictive, inert, and ideal gas species that behaves like a noble gas
27 but with a similar overall solubility function (i.e., similar concentration dissolved in vapor-
28 saturated melt at the same conditions) to S mimics the higher pressure initiation of degassing
29 for hydrous magmas and the overall behavior of S. We show that dilution by additional gas
30 species (i.e., H₂O and CO₂) in the vapor phase is the main driver of relatively high-pressure
31 degassing of S, which can be further enhanced for magmas at the sulfur solubility minimum.
32 Dilution is the same effect that drives: (1) higher pressure degassing of H₂O and CO₂ in melts
33 containing both compared to pure-H₂O or -CO₂; (2) higher pressure degassing of H₂O or He
34 in CO₂-rich systems; and (3) a decrease in the S content in gases during hydrous degassing.
35 This mechanism does not require a direct effect of H₂O (or CO₂) on the solubility function of
36 S, although H₂O does decrease the concentration of S dissolved in the melt at given set of
37 conditions contributing to higher pressure degassing of S. Although our emphasis is on the
38 significant impact of dilution on S degassing, the dilution effect impacts all volatile species,
39 with its magnitude depending on the relative solubility functions and concentrations of
40 volatile components present in the magma.

41

42 **Keywords:** degassing; dilution; magma; sulfur; water, carbon dioxide

Introduction

Sulfur (S) is typically the third most abundant volatile element in undegassed terrestrial magmas after H₂O and CO₂, such that S-bearing species are important constituents of volcanic gasses (e.g., Metrich & Mandeville, 2010; Wallace, 2005; Wallace & Edmonds, 2011). Sulfur has been inferred to begin to degas significantly from mid-ocean ridge (MORB) and some ocean island (OIB, e.g., Hawai‘i, Samoa) basaltic magmas on ascent at much lower pressure (P , typically ~5–30 MPa) than CO₂ and at P ’s comparable to those at which H₂O degasses significantly based on analyses of submarine glasses and melt/fluid inclusions (e.g., Dixon et al., 1991; Labidi et al., 2015; Lerner et al., 2021; Moore & Clague, 1987; Moore & Fabbi, 1971; Moore & Schilling, 1973; Moore & Thomas, 1988; Moussallam et al., 2016; Wallace & Carmichael, 1992; Wallace & Edmonds, 2011; Wieser et al., 2025). These studies have attributed this behaviour to the typically relatively low (and similar) concentrations of S and H₂O (~0.1–0.8 wt.%) and their relatively high (and similar) solubility functions (i.e., the concentration of a volatile species or component that can dissolve in a vapor-saturated melt at a given set of conditions – also described as an equilibrium constant or partition coefficient) in these magmas, especially compared to CO₂.

However, these observations contrast with measurements in some arc and back-arc-basin basalts from submarine lavas (e.g., Brounce et al., 2016; Davis et al., 1991; Nilsson & Peach, 1993) and melt inclusions that have quantified the CO₂ contents of melt-inclusion-hosted bubbles (e.g., Rasmussen et al., 2020), which have low S contents at relatively high P despite similar initial S contents to MORB and Hawai‘i (e.g., ~0.1–0.3 wt.%). Additional observations are found at some OIBs, such as: (1) Raman analyses of fluid inclusions from Mauna Loa (Hawai‘i) that have detected significant amounts of SO₂ in relatively high-density-CO₂ fluid inclusions, which are inferred to have been trapped at high P (Wieser et al., 2025); and (2) the suggestion of deep degassing (~300 MPa) of S based on melt inclusions

from El Hierro (Canary Islands), although these melts are S-rich (up to 0.5 wt% S; Longpré et al., 2017). These observations suggest that there can be significant degassing of S – perhaps even concurrently with CO₂ – at relatively high *P*. Additionally, such deeply degassed S has been invoked as the source of S-bearing gases found in evolved, shallower magmas and thereby to explain the “excess sulfur problem” (e.g., Christopher et al., 2010; Di Muro et al., 2008; Edmonds et al., 2010; Pallister et al., 1992; Roberge et al., 2009; Wallace, 2001) and as a potential trigger for precipitation of sulfide ores in porphyry copper deposits (e.g., Blundy et al., 2015; Hattori & Keith, 2001).

This degassing behaviour of S from (back-)arc and some OIB magmas is consistent with degassing models of water-rich magmas as they ascend, which predict that at the same bulk S content, S begins to degas at much higher *P* from water-rich magmas than from water-poor magmas (e.g., Boulliang & Wood, 2022; Ding et al., 2023; Edmonds & Wallace, 2017; Wallace & Edmonds, 2011; Yip et al., 2022). For Mauna Loa, modelling of degassing can generate higher *P* degassing of S if the initial melt has a higher initial S⁶⁺/S_T (0.3–0.4) than required for Kīlauea (0.1–0.2; Wieser et al., 2025).

Most explanations for the higher than expected *P* of degassing of S in hydrous magmas relate to changes in the partition coefficient of S between melt and vapor such that S transfers from the melt to the vapor at higher *P* in hydrous magmas compared to water-poor magmas, including: (1) a direct effect of H₂O on the partition coefficient between dissolved sulfide and H₂S vapor (i.e., ΔFMQ < +0.5; Ding et al., 2023 – FMQ is the oxygen fugacity (*f*_{O₂}) of the Fayalite-Magnetite-Quartz buffer, and ΔFMQ is the difference in log units between the *f*_{O₂} of the degassing magma and this buffer; e.g., Frost, 1991); (2) the effects of temperature (*T*), *f*_{O₂}, or melt composition on the melt-vapor partitioning of S that are associated with magmas that are more likely to be hydrous (e.g., Métrich et al., 2009; Wallace & Edmonds, 2011; Zajacz et al., 2012); and (3) the lower total solubility function of S at the “sulfur solubility

minimum” ($SS^{\min}$, due to the presence of both dissolved sulfide and sulfate in the melt) that occurs near $\sim\Delta FMQ+1$, which is close to f_{O_2} values of typical arc magmas (e.g., Ding et al., 2023; Hughes et al., 2023; Nilsson & Peach, 1993). Other explanations highlight that the partial degassing of S at high P could be related to the outgassing of other volatiles (e.g., CO_2 or H_2O ; Davis et al., 1991; Dixon & Stolper, 1995; Wallace & Edmonds, 2011). For instance, even for the same partition coefficient of S, for a greater mass fraction of H_2O exsolved, there will be more S partitioned into the vapor (Wallace & Edmonds, 2011).

In this paper, we apply the *VolFe* Python package (Hughes et al., 2025) to model closed- and open-system degassing of a representative Hawaiian basaltic melt spanning a range of bulk H_2O , CO_2 , and S concentrations and initial values of f_{O_2} . Our modelling largely agrees with previous modelling (e.g., Boulliung & Wood, 2022, 2023; Ding et al., 2023; Edmonds & Wallace, 2017; Gaillard & Scaillet, 2009; Wallace & Edmonds, 2011; Yip et al., 2022) in that during closed-system degassing the following are observed: (1) for relatively reduced magmas (initial $\Delta FMQ \lesssim +1$), significant degassing of S occurs at higher P for water-rich relative to water-poor melts; (2) this effect is positively correlated with the H_2O content of the system for a given initial f_{O_2} ; and (3) the magnitude of this effect is enhanced in the vicinity of the $SS^{\min}$. However, we also show that this effect is observed for closed-system degassing of magmas more oxidizing than the $SS^{\min}$ (i.e., for conditions at which the concentration of H_2S is negligible in both melt and coexisting vapor); when H_2S is forced artificially to be absent in the melt and/or vapor, even under reducing conditions; and with increasing bulk CO_2 as well. It also occurs in our modelling for open-system degassing. Finally – and importantly – we show that this effect is also predicted for inert volatile components that are present in the vapor and melt as a single species (e.g., the noble gases). Consequently, the effect of dissolved H_2O on the P at which significant amounts of such a volatile component degasses cannot be attributed dominantly to the concentrations or specific

behaviours of f_{O_2} -sensitive species such as H_2S or to any structural or thermochemical factors unique to S-bearing melts, although these do have some influence.

Based on numerical experiments we performed with *VolFe*, we conclude that these results can be understood via the “dilution effect” (e.g., Anderson et al., 2024, 2025; Botcharnikov et al., 2007; Dixon & Stolper, 1995; Gaillard & Scaillet, 2009; Gonnermann & Mukhopadhyay, 2007; Holloway, 1976; Iacovino et al., 2021; Lowenstern, 2001; Webster & Botcharnikov, 2011). For instance, consider a simple melt-vapor system containing only H_2O at a given P , T , and silicate melt composition. When the system contains both melt and vapor and the vapor is assumed to be pure H_2O , the vapor-saturated melt has a well-defined concentration of dissolved H_2O . If CO_2 is added to the system at constant P and T , some of it must partition into the vapor (and most of it will be, given its typically low solubility function relative to H_2O ; e.g., Dixon et al., 1995). The H_2O in the vapor is thereby *diluted* by the CO_2 in the vapor: i.e., the partial pressure and fugacity of H_2O ($p_{\text{H}_2\text{O}}$ and $f_{\text{H}_2\text{O}}$) must decrease. Consequently (as H_2O dissolution roughly follows Seivert’s law; e.g., Dixon et al., 1995; Stolper, 1982), the melt H_2O concentration must decrease when CO_2 is added to the system (e.g., Botcharnikov et al., 2007; Dixon & Stolper, 1995; Holloway, 1976; Lowenstern, 2001), resulting in transfer (i.e., degassing) of H_2O from melt to vapor. Similarly, starting from a melt + vapor assemblage in a system in which CO_2 is the only volatile component, addition of H_2O dilutes the CO_2 in the vapor, resulting in CO_2 degassing from the melt into vapor.

The dilution effect has been applied to explain changes in the degassing behaviour of H_2O - CO_2 as their concentrations change (e.g., Anderson et al., 2024, 2025; Botcharnikov et al., 2007; Dixon & Stolper, 1995; Holloway, 1976; Iacovino et al., 2021; Lowenstern, 2001) and the greater degree of degassing of He in CO_2 -rich OIBs relative to MORBs (Gonnermann & Mukhopadhyay, 2007). Additionally, it has been invoked to explain the decreasing S content of volcanic gases with increased initial H_2O during degassing (Gaillard & Scaillet,

2009), and now we apply it to explain quantitatively the deep degassing of S in hydrous
basaltic magmas (Dixon & Stolper, 1995).

The behaviour of sulfur during degassing

We use *VolFe* (v1.1.4) to model the behaviour of CHOS+-bearing volatile species during
isothermal, equilibrium, closed- or open-system, decompression-induced degassing (Hughes
et al., 2025). As in Hughes et al. (2023, 2024), all calculations presented in this paper use a
single volatile-free melt composition (i.e., a Hawaiian tholeiite from Brounce et al., 2017) at
a constant T of 1200 °C (Supplementary Table S1). Melt and vapor species considered and
model options used in calculations presented in this paper are detailed in Supplementary
Table S2 (Ardia et al., 2013; Dixon et al., 1995; Frost, 1991; Holland & Powell, 1991;
Hughes et al., 2023, 2024; Kress & Carmichael, 1991; Moussallam et al., 2019; Ohmoto &
Kerrick, 1977; O'Neill, 2021; O'Neill & Mavrogenes, 2022; Pan et al., 1991; Shaw &
Wones, 1964; Shi & Saxena, 1992; Wieser & Gleeson, 2023; Zajacz & Tsay, 2019). Only
melt and vapor phases are considered and hence some calculated melt + vapor compositions
may be metastable with respect to precipitation of an immiscible sulfide and/or anhydrite
phase (or even silicate minerals) if these phases are stable. However, the S content at the
same conditions for sulfide (pure FeS) and anhydrite saturation is calculated to check whether
the calculations are stable or metastable.

Calculations were primarily performed on systems with identical bulk S contents (1000
µg/g) but spanning a range of bulk H₂O content (0.2–5.0 wt.%), bulk CO₂ content (0.1–1.0
wt.%), and initial f_{O_2} values ($0.0 \leq \Delta\text{FMQ} \leq +2.5$). The bulk volatile contents are defined by
total H₂O (i.e., all H dissolved in the melt is reported as the equivalent amount of H₂O); total
CO₂ (i.e., all C dissolved in the melt is reported as the equivalent amount of CO₂), and S (i.e.,

all S dissolved in the melt is reported as the equivalent amount of total S, regardless of its speciation). Although the bulk volatile contents of the melt are referred to using simply H₂O, CO₂, and S throughout the text, they include the contributions of other volatile species present in the melt at equilibrium (e.g., H₂, CO, CH₄, H₂S). The initial f_{O_2} value for a particular calculation is given by ΔFMQ_0 , which we define as the difference in $\log_{10} f_{O_2}$ between the FMQ buffer at P_{sat}^v for the T of the calculation, where P_{sat}^v is the P of vapor-saturation for the melt composition of interest given its volatile concentrations (Hughes et al., 2023, 2024).

Degassing paths with varying bulk water and carbon dioxide

The behaviour of water and carbon dioxide

Solid curves in Figure 1 show *VolFe*-calculated, closed-system, isothermal, equilibrium (albeit potentially metastable) degassing paths from P_{sat}^v to 0.1 MPa for melts initially containing 1000 µg/g bulk CO₂ + 1000 µg/g bulk S at $\Delta FMQ_0=0$ with a bulk H₂O range from 0.2 to 5.0 wt.%. The P at which degassing begins upon decompression (i.e., P_{sat}^v) increases with increasing bulk H₂O because the sum of the partial pressures of all vapor species must add up to the total P (e.g., Dalton, 1802; Hughes et al., 2024; Verhoogen, 1949). All volatiles begin degassing at P_{sat}^v : i.e., all volatiles will be present in the saturating vapor for $P \leq P_{sat}^v$, albeit in potentially small quantities (e.g., Métrich et al., 2009; Métrich & Wallace, 2009; Verhoogen J, 1949; Wallace et al., 2015). To compare changes in the P of degassing of different volatiles across different initial conditions, we compare the P at which 10% of each volatile has degassed (i.e., the dissolved melt content of a particular volatile i is 10% its initial, undegassed value), which we term $P_{10\%}^i$ (i.e., circles in all figures). Hence, higher P (i.e., deeper) degassing results in an increase in $P_{10\%}^i$.

Regardless of the bulk H₂O and S, CO₂ is always the first volatile to degas significantly in Figure 1 (i.e., $P_{10\%}^{\text{CO}_2} > P_{10\%}^{\text{H}_2\text{O}}$ and $P_{10\%}^{\text{S}}$: compare the P of the filled circles on the solid curves) because CO₂ is the most insoluble of these species (e.g., Dixon et al. 1995). For 0.2 and 1.0 wt.% bulk H₂O, CO₂ degasses roughly linearly with decreasing P from P_{sat}^v to 0.1 MPa (i.e., melt CO₂ decreases roughly linearly with decreasing P along the red and purple solid curves in Figure 1b). For 3.0 and 5.0 wt.% bulk H₂O, CO₂ degassing also begins roughly linearly with a similar slope to the 0.2 and 1.0 wt.% bulk H₂O cases, which is similar to the modelling results on MORB for 0–2 wt.% H₂O and 500 µg/g CO₂ in Dixon et al., (1995). However, the trends are offset to higher P by approximately the P_{sat}^v values for melts containing 3 and 5 wt.% bulk H₂O only (i.e., systems in which bulk CO₂ and S are 0 µg/g). At $\geq 95\%$ degassing of the bulk CO₂, the rate of CO₂ degassing with respect to P decreases dramatically (i.e., solid black and blue curves in Figure 1b curve upward dramatically at lower P).

Degassing of H₂O has a strongly concave-down shape for all bulk H₂O values (Figure 1a). Significant degassing of H₂O occurs at a similar P to a melt containing only H₂O of the same bulk concentration would begin degassing: i.e., $P_{10\%}^{\text{H}_2\text{O}}$ and P_{sat}^v for the equivalent pure-H₂O system (grey curve) are very similar in Figure 1a. This significant *increase* in the degassing rate of H₂O also occurs at a similar P to the *decreasing* rate of CO₂ degassing with respect to P (Figure 1a and b). Additionally, there is a long, near-vertical tail at $P > P_{10\%}^{\text{H}_2\text{O}}$ corresponding to low degassing rates with respect to P whilst most of the CO₂ degasses (Figure 1a). This behaviour can also be seen when the bulk CO₂ content is increased to 1 wt.% (solid vs. dash-dot curves in Figure 2a).

This behaviour of H₂O-CO₂ degassing is due to the dilution effect, as described in the Introduction (e.g., Anderson et al., 2024, 2025; Botcharnikov et al., 2007; Dixon & Stolper, 1995; Holloway, 1976; Iacovino et al., 2021; Lowenstern, 2001). Dilution by H₂O has a large

effect on CO₂ degassing, resulting in CO₂ degassing at higher P than if it were the only volatile in the system (Figure 1b). For example, CO₂ degasses at increasingly higher P with increasing bulk H₂O (i.e., $P_{10\%}^{\text{CO}_2}$ is a strong function of bulk H₂O, Figure 1b; e.g., Dixon & Stolper, 1995; Yip et al., 2022). These results reflect the typically high concentration of H₂O relative to CO₂ in natural magmas and the low solubility function of CO₂ relative to H₂O in most natural melts (e.g., Dixon et al., 1995; Dixon & Stolper, 1995). This combination means that the CO₂ degassing paths are essentially simply offset to higher P from the pure-CO₂ curve by roughly the P_{sat}^v of pure-H₂O.

The complementary dilution effect of CO₂ on H₂O degassing is more subtle (i.e., $P_{10\%}^{\text{H}_2\text{O}}$ is similar to P_{sat}^v for pure-H₂O; e.g., Dixon et al., 1995) reflecting the low concentration of CO₂ (Figure 1a). Nevertheless, the presence of CO₂ does result in a long tail of high P degassing of H₂O (from P_{sat}^v to $\sim P_{10\%}^{\text{H}_2\text{O}}$; Figure 1a) due to CO₂ being highly insoluble. However, higher initial CO₂ concentrations (e.g., ≥ 1 wt.%), have more substantial effects on the degassing of H₂O (Figure 2a; e.g., Anderson et al., 2024, 2025; Dixon & Stolper, 1995), highlighting the importance of relative bulk concentrations on the quantitative effects of dilution.

The behaviour of sulfur

For 1000 $\mu\text{g/g}$ bulk CO₂, as for H₂O, the rate of S degassing with respect to P is small initially, leading to the high- P , near-vertical tails at ~ 1000 $\mu\text{g/g}$ S (Figure 1c). The rate of S degassing increases at a similar (although slightly higher) P relative to that at which the rate of H₂O degassing with respect to decreasing P increases dramatically, leading to strong downward curvatures of the degassing curves (Figure 1c). By design, the $P_{10\%}$ degassing for H₂O and S are all close to the position on their respective degassing curves at which the rate of degassing increases rapidly (i.e., 10% was chosen for this effect, compare the P 's of the solid coloured circles in Figure 1a and c). It is interesting that $P_{10\%}^{\text{H}_2\text{O}}$ and $P_{10\%}^{\text{S}}$ are similar to

each other for all of the bulk H₂O concentrations shown, ranging from 0.2 to 5.0 wt.%. Although there are subtle differences, $P_{10\%}^S$ also increases with increasing bulk H₂O when degassing is open- rather than closed-system (solid vs. dotted curves and open vs. closed symbols in Figure 1c). Additionally, increasing the bulk CO₂ content also increases $P_{10\%}^S$ substantially (Figure 2c; e.g., Yip et al., 2022). All degassing paths shown in Figure 1 and Figure 2 (all of which started degassing at $\Delta FMQ_0=0$) are undersaturated with respect to sulfide (assuming pure FeS) and anhydrite for the parameterisations of sulfide content at sulfide saturation and sulfate content at anhydrite saturation used in our calculations (Supplementary Material Section 3.1).

A key feature of Figure 1 is the increase in $P_{10\%}$ for *all* volatiles with increasing bulk H₂O of the melt at constant bulk CO₂ and S. For H₂O itself, the higher the melt H₂O, the greater the depth at which H₂O significantly degasses. In the case that H₂O is the only volatile in the system, the P that H₂O begins to degas (P_{sat}^v) is simply the P on the solubility curve for pure-H₂O corresponding to the bulk H₂O content (Figure 1a; e.g., Dixon & Stolper, 1995). The increase of $P_{10\%}^S$ with increasing bulk H₂O is particularly dramatic (Figure 1c): the same melt with only 1000 µg/g S at $\Delta FMQ_0=0$ would begin degassing at just ~0.5 MPa (Hughes et al., 2023). The fact that $P_{10\%}^S$ also increases with increasing bulk CO₂ highlights that this behaviour is not unique to the effect of H₂O (Figure 2c).

The VolFe-calculated increase in $P_{10\%}^S$ with increasing bulk H₂O is in overall agreement with the independent modelling of Yip et al. (2022) and Ding et al. (2023). We regard this agreement as important validation of *Sulfur_X* (Ding et al., 2023) and *VolFe* given that they were developed independently and with different thermodynamic formulations, computational approaches, and approximations (see Figure S2 for a direct comparison of the results of *VolFe* and *Sulfur_X* output). *EVo* (Liggins, 2023; Liggins et al., 2020, 2022), that was used in Yip et al. (2022), uses a similar modelling approach to *VolFe*.

266

267 *The behaviour of sulfur during degassing with variable initial oxygen fugacity*

268 The speciation of H, C, and S both in silicate melt and vapor vary with f_{O_2} and this
269 results in variability of degassing paths when calculated over a sufficiently large range in
270 ΔFMQ_0 (e.g., Gaillard et al., 2022; Holloway & Blank, 1994; Lo et al., 2021). Some of these
271 effects are illustrated in Figure 3 by repeating the closed-system calculations from Figure 1 at
272 a single bulk H₂O (3 wt.% H₂O + 1000 µg/g CO₂ + 1000 µg/g S) over a range of ΔFMQ_0
273 from 0.0 to +2.5 that spans abundant terrestrial magmas (e.g., Cottrell et al., 2021). All
274 calculations shown in Figure 3 are undersaturated with respect to sulfide (assuming pure FeS)
275 and anhydrite for the parameterisations of sulfide content at sulfide saturation and sulfate
276 content at anhydrite saturation used (Supplementary Material Section 3.1). For $\Delta FMQ_0 \geq 0$,
277 the H₂O and CO₂ degassing curves are essentially independent of ΔFMQ_0 (Figure 3a and b)
278 because H₂O and CO₂ are the dominant calculated H- and C-bearing vapor species and H₂O_T
279 and CO_{2,T} the dominant melt species (i.e., oxidised H- and C-bearing species in the melt: OH⁻
280 and H₂O_{mol} or CO_{2,mol} and CO₃²⁻, respectively; e.g., Armstrong et al., 2015; Hirschmann et
281 al., 2012; Hughes et al., 2024). In contrast, the speciation of S in vapor and melt vary
282 significantly over this f_{O_2} range (Figure 3g–i), resulting in observable variations in S
283 degassing behaviour as a function of the initial f_{O_2} (Figure 3d and e).

284

285 Speciation of sulfur at the pressure of vapor saturation

286 The variation in ΔFMQ_0 correlates with systematic changes in the speciation of S in the
287 melt and vapor at P_{sat}^v that can be understood by the chemical equilibria among S-bearing
288 species containing S of different oxidation states (e.g., Hughes et al., 2024). For example, the
289 melt S⁶⁺/S_T at P_{sat}^v varies from ~0 at $\Delta FMQ_0=0$ (i.e., dissolved S in the melt is almost entirely

sulfide: note that “sulfide” in this statement includes both H_2S plus “other” S^{2-} species – e.g., FeS , MgS , etc. – collectively referred to as “ *S^{2-} ”; see Hughes et al., 2024) to ~ 1 at $\Delta\text{FMQ}_0 = +2.5$ (i.e., almost entirely sulfate; Figure 3i). This reflects the sigmoidal relationship between S speciation in melt and f_{O_2} (see eq. 20 in Hughes et al., 2024 – there are some minor mistakes in this equation in the original equation and the correct version is given in Supplementary Material Section 2: e.g., Baker & Moretti, 2011; Baumgartner et al., 2017; Fincham & Richardson, 1954; Gaillard et al., 2011, 2013, 2015; Gaillard & Scaillet, 2009; Hughes et al., 2023; Moretti, 2021; Moretti & Ottonello, 2003, 2005).

The gradual transition in S speciation in the melt from dissolved sulfide to sulfate with increasing ΔFMQ_0 leads to a maximum in P_{sat}^v (i.e., the “minimum” – because P increases downwards – in the curve defined by the stars in Figure 3c) that corresponds to the SS^{min} (Hughes et al., 2023, 2024). At constant total dissolved S in the melt, T , and major element melt composition, the SS^{min} occurs at $\text{S}^{6+}/\text{S}_\text{T} \approx 0.75$ because when the vapor is SO_2 -dominated, the *S^{2-} concentration in the melt decreases with increasing f_{O_2} , whilst the S^{6+} concentration in the melt increases with increasing f_{O_2} (e.g., Baker & Moretti, 2011; Cicconi et al., 2020; Ding et al., 2023; Fincham & Richardson, 1954; Hughes et al., 2023; Moretti et al., 2003; Moretti & Ottonello, 2005; Papale et al., 2022). The SS^{min} occurs at *exactly* $\text{S}^{6+}/\text{S}_\text{T} = 0.75$ in the S-O system (i.e., it contains no H and C) when the vapor consists of purely ideal SO_2 (i.e., no S_2 ; Hughes et al., 2023). The non-ideality of the vapor, presence of other vapor species (e.g., S_2 , H_2O , CO_2 , H_2S , etc.), presence of other S-bearing melt species (e.g., H_2S), and variations in $f_{\text{H}_2\text{O}}$ mean the SS^{min} occurs at $\text{S}^{6+}/\text{S}_\text{T} \approx 0.75$ in the C-H-S-O system (Hughes et al., 2024).

The total solubility function of S^{2-} ($\text{*S}^{2-} + \text{H}_2\text{S}$) does not vary significantly when the dominant S-bearing vapor species is H_2S (e.g., Ding et al., 2023; Hughes et al., 2024) and the fraction of S dissolved as H_2S in the melt ($\text{S}_{\text{H}_2\text{S}}/\text{S}_\text{T}$) at P_{sat}^v decreases with increasing ΔFMQ_0

(Figure 3h: note this is currently a unique result of *VolFe* as other tools do not include both S^{2-} and H_2S ; e.g., Hughes et al., 2025; Iacovino et al., 2026). The ratio of dissolved H_2S to dissolved S^{2-} is essentially unchanging at P_{sat}^v because melt H_2O is nearly constant such that $f_{\text{H}_2\text{O}}$ is essentially not varying (see eq. 19 in Hughes et al., 2024). However, as melt $\text{S}^{6+}/\text{S}_\text{T}$ increases with increasing ΔFMQ_0 , $(\text{S}^{2-} + \text{S}_{\text{H}_2\text{S}})/\text{S}_\text{T}$ is decreasing such that $\text{S}_{\text{H}_2\text{S}}/\text{S}_\text{T}$ also decreases. The calculated equilibrium vapor composition also varies significantly with increasing ΔFMQ_0 , with increasing proportions of SO_2 relative to H_2S at P_{sat}^v , and the transition from H_2S to SO_2 in the vapor phase with increasing f_{O_2} happens at lower f_{O_2} than the SS^{min} (Figure 3g: see homogeneous vapor equilibria equations 7 and 9 in Hughes et al., 2024).

Behaviour of sulfur during depressurisation

From any given ΔFMQ_0 , the *VolFe*-calculated f_{O_2} -value varies with progressive depressurization and degassing (Figure 3c), which is reflected in the melt $\text{Fe}^{3+}/\text{Fe}_\text{T}$ (Figure 3f). For the f_{O_2} range considered here, H_2O and CO_2 degassing cause a small amount of oxidation (e.g., Benaroya & Herd, 2026; Brachmann et al., 2026; Burgisser & Scaillet, 2007; Candela, 1986; Carmichael & Ghiorso, 1986; Gaillard et al., 2015; Gaillard & Scaillet, 2014; Herd & Benaroya, 2025; Mathez, 1984), which is modified by S degassing. Sulfur degassing leads to overall reduction for melts at lower f_{O_2} where dissolved sulfide (S^{2-}) degasses to SO_2 (S^{4+}) because there is a +6 change in the redox state of S (red curves in Figure 3c; e.g., Anderson & Wright, 1972; Brounce et al., 2017; Burgisser & Scaillet, 2007; Cottrell & Kelley, 2011; de Moor et al., 2013; Gaillard et al., 2011, 2015; Hughes et al., 2023; Métrich et al., 2009; Moussallam et al., 2014, 2016; O'Neill & Mavrogenes, 2022; Taracsák et al., 2026). Degassing of dissolved sulfide (either as S^{2-} or H_2S) to H_2S in the vapor (S^{2-}) is neutral in its effect on f_{O_2} (e.g., de Moor et al., 2013; Ding et al., 2023; Taracsák et al., 2026,

for S^{2-}). Overall oxidation occurs due to S degassing at higher f_{O_2} where dissolved sulfate (S^{6+}) degasses to SO_2 (S^{4+}) in the vapor due to the -2 change in the redox state of S (blue curves in Figure 3c; e.g., Boulliang & Wood, 2022, 2023; de Moor et al., 2013; Farsang & Zajacz, 2024; Gaillard et al., 2015; Hughes et al., 2023; Métrich et al., 2009; O'Neill & Mavrogenes, 2022). The boundary separating S-degassing induced reduction and oxidation is the f_{O_2} of the $SS^{\min}$ (e.g., de Moor et al., 2013; Hughes et al., 2023; O'Neill & Mavrogenes, 2022), but changes in oxygen fugacity also reflect the effects of C- and H-degassing.

These f_{O_2} variations as degassing proceeds lead to systematic changes in the S speciation of the melt and vapor (Figure 3g–i) due to the same chemical equilibria described for the changes at P_{sat}^v in the previous section. However, H_2O begins degassing significantly at ~ 100 MPa (Figure 3a, shown by the dashed line in Figure 3c) causing f_{H_2O} to decrease significantly from this P . This H_2O degassing at $P < 100$ MPa has a minor effect on the melt S^{6+}/S_T (see eq. 20 in Hughes et al., 2024) but causes S_{H_2S}/S_T to decrease significantly (Figure 3h; see eq. 19 in Hughes et al., 2024) and the proportion of SO_2 in the vapor to increase rapidly such that it is the dominant S-bearing gaseous species for all f_{O_2} near the surface (Figure 3g; e.g., Ding et al., 2023; Farsang & Zajacz, 2024; further explanation in Supplementary Material Section 2.3). These systematic changes in sulfur speciation in the melt and vapor also lead to systematic changes in sulfur isotopes during degassing (e.g., Taracsák et al., 2026) because the magnitude and sign of the isotope fractionation factors are highly sensitive to speciation.

Based on our modelling, the extent of S degassing at any given value of $P \leq P_{\text{sat}}^v$ increases with increasing ΔFMQ_0 (particularly visible by the variation in $P_{10\%}^S$) from 0 to +1.4 ($\sim SS^{\min}$) when the S dissolved in the melt is mostly sulfide-dominated ($S^{6+}/S_T \lesssim 0.75$: i.e., see the downward pointing arrow in Figure 3d; e.g., Ding et al., 2023; Gaillard & Scaillet, 2009; Hughes et al., 2023). The extent of S degassing and $P_{10\%}^S$ then decreases as ΔFMQ_0 increases further to +2.5 when the melt is sulfate-dominated ($S^{6+}/S_T \gtrsim 0.75$: see the

upward pointing arrow in Figure 3e; e.g., Boulliang & Wood, 2022, 2023; Hughes et al., 2023; Yip et al., 2022). The higher $P_{10\%}^S$ for a melt that begins degassing at an f_{O_2} near the $SS^{\min}$ relative to the $P_{10\%}^S$ of the same melt at higher and lower values of f_{O_2} reflects the lower total S solubility function at the $SS^{\min}$ relative to melts that begin degassing at higher and lower f_{O_2} (e.g., Backnaes & Deubener, 2011; Baker & Moretti, 2011; Carroll & Rutherford, 1985; Cicconi et al., 2020; Ding et al., 2023; Fincham & Richardson, 1954; Hughes et al., 2023, 2024; Katsura & Nagashima, 1974; Lesne et al., 2015; Matjuschkin et al., 2016; Moretti et al., 2003; Moretti & Ottonello, 2005; Nash et al., 2019; O'Neill & Mavrogenes, 2022; Papale et al., 2022). All other things being equal (e.g., bulk H_2O , CO_2 , and S in Figure 3), $P_{10\%}^S$ increases (and thus S has degassed more at any given $P \leq P_{sat}^v$) if the total S solubility function is lower (as it is at the $SS^{\min}$; Figure 3c). Hence, f_{O_2} -dependent S speciation in the melt and vapor affects S degassing through its effects on the total S solubility function as a function of f_{O_2} . However, at a given ΔFMQ_0 , $P_{10\%}^S$ increases systematically with increasing bulk H_2O as was observed at $\Delta FMQ_0=0.0$ (see Figure 1c for $\Delta FMQ_0=0.0$ and Figure S4c and f for $\Delta FMQ_0=+1.4$ and $+2.5$, respectively).

Potential drivers of the behaviour of sulfur during degassing

The significant quantities of H_2S in the melt (Figure 3h) and vapor (Figure 3g) under relatively reducing conditions (e.g., $\Delta FMQ_0 < +1$) could suggest that H_2S plays an important role in the increase in $P_{10\%}^S$ with increasing bulk H_2O shown in Figure 1c. Ding et al. (2023), whilst not including an H_2S species in the melt, attributed the higher P degassing of S with increasing bulk H_2O (i.e., the modelled increase in $P_{10\%}^S$) to a lowering of the S solubility function by reaction of H_2O in the vapor with anhydrous sulfide species in the melt (e.g., FeS) to generate H_2S molecules that exsolve into the vapor and a direct effect of dissolved H_2O on this partition coefficient. However, we observe an increase in $P_{10\%}^S$ in hydrous melts

when: (1) H₂S is artificially forced to be completely insoluble in the melt (i.e., H₂S is present in the coexisting vapor but not in the melt); (2) H₂S is assumed to be completely absent from the system (i.e., no H₂S is allowed to stabilise in either the melt or vapor); and (3) whether or not there is a direct-effect of H₂O on the sulfide capacity (e.g., Moretti & Ottonello, 2005; O'Neill, 2021). See Supplementary Material Section 3.5 for more details on all of these numerical experiments.

Additionally, under oxidising conditions (i.e., $\Delta FMQ_0 > +1.4$), $P_{10\%}^S$ increases for increasing bulk H₂O at a comparable magnitude to the reduced cases (Figure 1c and Figure S4c and f) when: (1) H₂S is a minor species in the melt (Figure 3h) and vapor (Figure 3g) for the parameters used in these calculations (Table S2); and (2) regardless of whether there is or is not a direct effect of H₂O on the sulfate capacity (see Supplementary Material Section 3.5: e.g., Boulliung & Wood, 2022; Moretti & Ottonello, 2005). That the effect of bulk H₂O of increasing $P_{10\%}^S$ is present in the (near) absence of H₂S in the system under oxidizing conditions supports our conclusion that H₂S does not play a major role in the *VolFe*-modelled effects of dissolved H₂O on the higher *P* degassing of S from basaltic magmas.

These observations demonstrate that in the context of *VolFe*: neither: (1) the presence or absence of H₂S species in the melt and/or vapor; nor (2) a direct influence of H₂O on the S solubility function (e.g., a H₂O compositional term in the sulfide and/or sulfate capacity); nor (3) the transition from sulfide to sulfate as the dominant dissolved S-bearing melt species with increasing f_{O_2} , are likely to be the main drivers of the higher *P* degassing of S during ascent of hydrous magmas. These factors are predicted by *VolFe* to have effects on the details of S degassing, including higher *P* S degassing for melts that begin degassing in the vicinity of the SS^{min} and minor changes in the *P* of degassing depending on whether H₂S is a melt and/or vapor species (as shown in the previous section and Supplementary Material Section 3.5). Finally, a similar increase in $P_{10\%}^S$ occurs with increasing bulk CO₂ even at low bulk

H₂O (Figure 2c). Hence, our modelling suggests that something else must be responsible for the first-order result that, for a given anhydrous major element melt composition, melts with higher bulk H₂O (or CO₂) degas S at higher P than melts with less dissolved H₂O (or CO₂) over the range of ΔFMQ_0 values we have explored (which are most relevant to common and abundant terrestrial magmas) As we anticipated in the Introduction and describe in detail in the following sections, we conclude that a “dilution effect” – driven by the presence of H₂O and/or CO₂ – is primarily responsible for the higher P degassing of S from natural, hydrous melts.

The behaviour of an inert, ideal volatile species during degassing

As illustrated in the previous section, the interactions between homogeneous and heterogeneous equilibria involving melt and vapor in S±H±C-bearing magmas are complex. As a consequence, the details of degassing behaviour can be hard to anticipate fully without an internally consistent thermodynamic model that incorporates the large number of potentially relevant independent interspecies interactions. In this section we consider the degassing behaviour of a fictive volatile component – referred to as “ χ ” – that does not have these complications (χ is referred to as “X” in Hughes et al., 2025), but to avoid confusion with mole fraction we use χ here). By definition, χ is an ideal volatile component that: (1) is present in the same form in melt and vapor; (2) does not react with other species in melt or vapor to form additional species; and (3) has a solubility function in melt unaffected by variations in f_{O_2} or H₂O. As a tangible example, the noble gases would closely approximate the χ component (e.g., Carroll & Stolper, 1993; Iacono-Marziano et al., 2010). In the following calculations, the molecular weight of χ is chosen as 40 (i.e., it can be viewed as similar to Ar). We compare the *VolFe*-calculated degassing behaviour of such a component to

that of S – especially the degree to which such a model χ component displays an increase in the P of significant degassing with increasing H₂O content of the system.

We define the equilibrium constant (or solubility function) for χ (K_χ) as:

$$K_\chi = \frac{w_\chi^m}{p_\chi}, \quad (1)$$

where w_χ^m is the concentration of χ in the melt ($\mu\text{g/g}$) and p_χ is the partial pressure of χ in the vapor (MPa). For simplicity we choose K_χ to be independent of all other parameters, including P , T , and the concentrations of all other species in the melt and vapor (especially H₂O and CO₂). We initially choose a value of 320 $\mu\text{g/g/MPa}$ for K_χ so that the χ component mimics quantitatively the behaviour of S for the calculation containing bulk concentrations of 3 wt.% H₂O + 1000 $\mu\text{g/g}$ CO₂ at $\Delta\text{FMQ}_0=0$.

In Figure 4 we show the effects of replacing S – with its full complexity resulting from the presences of multiple S-bearing species in melt and vapor – by χ , with its single species in both the melt and vapor and its strictly Henrian solubility function. The degassing curves in black for S (solid) and χ (dashed) in Figure 4c are nearly identical, which is to some degree by design; i.e., the value of 320 $\mu\text{g/g/MPa}$ for K_χ was chosen for this purpose such that $P_{10\%}^S \approx P_{10\%}^\chi$. However, the excellent agreement between the *shapes* of these two degassing curves is not by design. The details of the shapes of the two black degassing curves in Figure 4c reflect the thermodynamic functions and interactions built into *VolFe*: i.e., the complex speciation in the melt and vapor of S; and in contrast, the simple, Henrian behaviour of the non-reacting χ species. The strong similarity of the shapes of the S and χ degassing curves in Figure 4c suggests that the degassing behavior of S in these calculations is not strongly dependent on the details of S speciation in the melt or vapor. Instead it depends primarily on the overall solubility function of S in the melt, which is very similar to that chosen for χ in Figure 4c. Moreover, changing bulk H₂O up to 5.0 wt.% or down to 0.2–1.0 wt.% deepens or

shallows the degassing curves similarly for χ and S – again, with no adjustable parameters (i.e., $K_\chi = 320 \mu\text{g/g/MPa}$ for all three dashed curves in Figure 4c). Although the shapes of the S and χ curves for the 0.2–1.0 and 5.0 wt.% bulk H₂O curves are still similar, they are no longer essentially identical as they are for the 3.0 wt.% bulk H₂O curves. The progressive overall increases in P of S and χ degassing with increasing bulk H₂O are also very similar (i.e., $P_{10\%}^S$ and $P_{10\%}^\chi$ are similar to each other for each bulk H₂O content). The solid and dashed degassing curves for H₂O (Figure 4a) and CO₂ (Figure 4b) are essentially indistinguishable, reflecting the similarity in the behaviour of S and χ .

Figure 5a shows how tuning K_χ (vertical dashed lines in Figure 5b) can enable the degassing behaviour of χ to mimic the behaviour of S when ΔFMQ_0 is varied. In contrast to χ , the solubility function of S in the melt along its degassing curve is not constant (Figure 5b), reflecting the fully complex thermodynamic characterization of heterogeneous and homogeneous equilibria among all S-bearing species in melt and vapor used by *VolFe*. Although the K_χ values are constant (by assumption) and the calculated “ K_S ” values are not (Figure 5b), the behavior of χ and S are still essentially indistinguishable (Figure 5a). These comparisons of the degassing curves of S and χ demonstrate that an inert χ component mimics the degassing of S if it is assigned a similar solubility function to total S (Figure 4 and Figure 5). The major element melt composition can strongly influence the S solubility function through the compositional dependencies of the sulfide and sulfate capacities (e.g., Boulliung & Wood, 2022, 2023; Moretti & Ottonello, 2005; O’Neill, 2021; O’Neill & Mavrogenes, 2002, 2022); consequently, although we anticipate that the χ degassing curves will mimic the S degassing curves for other melt compositions as well, the equilibrium constant of χ used in the model would also have to vary with melt composition in proportion to the variations in the total S solubility function.

In summary, increasing the bulk melt H₂O content causes deeper degassing (i.e., increases $P_{10\%}$) even for an ideal and inert volatile component with only a single vapor species and melt species (Figure 4c). This replicates well the dependence of S degassing on variable bulk H₂O and initial f_{O_2} despite the absence of the complexity of melt and vapor speciation characteristics of S. This suggests that such complexities do not play significant roles in the variability in the depth of significant degassing of S in magmas spanning relatively large ranges of bulk H₂O and initial f_{O_2} . In the next section, we describe how the so-called “dilution effect” (e.g., Anderson et al., 2024, 2025; Botcharnikov et al., 2007; Dixon & Stolper, 1995; Gonnermann & Mukhopadhyay, 2007; Holloway, 1976; Iacovino et al., 2021; Lowenstern, 2001; Webster & Botcharnikov, 2011) can provide a straightforward explanation of the similar behavior of S and χ during degassing and guidance into the expected degassing behavior of systems in which multiple volatile components are actively and simultaneously degassing from melts on ascent of magmas.

Dilution-driven degassing

It has long been known that the solubility function of S in vapor-saturated silicate melts is a strong function of f_{O_2} , as f_{O_2} affects the S^{6+}/S^{2-} ratio of silicate melt over the range relevant to most terrestrial magmas (e.g., Carroll & Rutherford, 1985; Fincham & Richardson, 1954; Scaillet et al., 1998; Wallace & Carmichael, 1994) and the SO₂/H₂S ratio of the vapor (e.g., Symonds et al., 1994; Whitney, 1984). These changes in melt S^{6+}/S^{2-} and vapor SO₂/H₂S result in shifts in the dominant heterogeneous equilibria that control the total S contents of vapor-saturated melts (e.g., Fincham & Richardson, 1954; Hughes et al., 2023; Moretti & Ottonello, 2003). It may thus appear at first somewhat surprising that the behaviour of the fictive volatile species χ – which does not have multiple molecular or

heterovalent species and is assumed by definition to have no chemical interactions with water in the melt or vapor – reproduces so well the effects of bulk H_2O and f_{O_2} on the degassing curves of S when χ is assigned a partitioning between vapor and melt similar to that of the total S under similar conditions (Figure 4 and Figure 5). In this section, we describe how this behaviour predicted by *VolFe* – and in particular the increased P of χ degassing from the melt as the H_2O content of the melt increases and the degree to which it matches the details of S degassing – can be relatively simply understood with reference to the phenomenon of “dilution” (e.g., Anderson et al., 2024, 2025; Botcharnikov et al., 2007; Dixon & Stolper, 1995; Gonnermann & Mukhopadhyay, 2007; Holloway, 1976; Iacovino et al., 2021; Lowenstern, 2001; Webster & Botcharnikov, 2011) .

Dilution-driven degassing by isobaric addition of an insoluble volatile species to the vapor

Suppose we start with a silicate melt and a multicomponent vapor in equilibrium at constant P and T . For this simple example, we assume that the vapor is an ideal gas and that each of the i volatile species dissolved in the melt are ideal species (i.e., eq. 1 applies for all of the i species). Starting from this equilibrium state, suppose we instantaneously add a finite amount of an additional, perfectly insoluble component to the system at constant P and T . The new component will partition completely into vapor as it is insoluble but since P is constant, the partial pressures of all other species in the vapor must decrease proportionately due to dilution with the newly introduced insoluble volatile component. However, the instantaneous decrease in the p_i for each of the i soluble species results in the melt and vapor being out of equilibrium: i.e., w_i^m/p_i has increased for each vapor species due to the instantaneous decrease in p_i but the unchanged value of w_i^m for each melt species. As a result, this initial value of w_i^m is higher than the equilibrium value given the new, lower value of p_i . In other words, the addition of the new component to the vapor means the melt is now

oversaturated with respect to all the initial components in the vapor. In response, some of each of the i soluble volatile component must transfer from the melt to the vapor (i.e., degas), decreasing w_i^m and increasing p_i until the equilibrium value of K_i is re-achieved. The p_i of the instantaneously added completely insoluble component will decrease due to dilution resulting from the transfer of dissolved volatiles from the melt to the vapor, resulting in further adjustment of the concentrations of these components in the melt and vapor. Eventually the system will reach a new equilibrium state that is degassed in all volatiles relative to the initial state, despite the fact that the equilibrium constants (i.e., the value of K_i) for each species has not changed.

A simplified illustration of this process is shown in Figure 6 for a system with two ideal volatile components: “Y” is soluble in the melt (i.e., $K_Y > 0$ $\mu\text{g/g/MPa}$) whilst “Z” is insoluble (i.e., $K_Z = 0$ $\mu\text{g/g/MPa}$; note that this cannot be the case in real systems). Initially, there is 10 $\mu\text{g/g}$ Y dissolved in the melt coexisting with a pure-Y vapor at 100 MPa and 1200 °C (Figure 6a). At constant P and T , Z is gradually added to the system such that the vapor achieves 50:50 mol.% Y and Z, causing the volume of vapor to increase because the number of moles of gas species in the vapor increases (Figure 6b). To achieve equilibrium given that K_Y remains constant, Y must degas leaving only 5 $\mu\text{g/g}$ Y in the melt (Figure 6b). This particular process could apply to pure- CO_2 gas streaming through a column of water-bearing magma at low P where CO_2 is not very soluble; such a process would progressively remove H_2O from the melt, and it could eventually nearly entirely dehydrate the magma, assuming no changes in T (e.g., Anderson et al., 1989; Scaillet, 2022).

The central role of dilution by water degassing in the deepening of sulfur degassing

In contrast to the simple example of the effects of dilution illustrated in Figure 6 and described in the previous section, for closed-system degassing the bulk composition of the

system is held constant. Hence, dilution of volatile components in the vapor by addition of a new volatile component to the system (or more of a volatile that is already present) and the resultant degassing of other volatile components in the melt as described in the previous section do not apply in detail to a closed-system (nor is addition of a *completely* insoluble volatile component as previously described realistic). However, decompression-driven degassing of more volatile components and/or of components present in the system at high concentrations relative to other volatile species (e.g., H₂O relative to all other volatile components as considered here and in some cases CO₂) will have the same overall effect of diluting all other components in the vapor (i.e., increasing their $P_{10\%}$). This dilution in a closed-system drives “additional” degassing of all other species relative to what would occur if the other volatiles were not present in the undegassed melt (Dixon & Stolper, 1995). This has been used to explain the behaviour of mixed H₂O-CO₂ degassing (e.g., Anderson et al., 2024, 2025; Botcharnikov et al., 2007; Dixon & Stolper, 1995; Holloway, 1976; Iacovino et al., 2021; Lowenstern, 2001) and increased He degassing in CO₂-rich OIBs compared with MORBs (e.g., Gonnermann & Mukhopadhyay, 2007). In this section, by comparing *VolFe*-modeled degassing of S to that of the inert and ideal species χ , we conclude that the presence of H₂O is typically the principal driver of the increase in P of degassing of S during closed- and open-system degassing of hydrous magmas (Figure 4c; Dixon & Stolper, 1995) via the significant dilution of S in the vapor by the degassing of H₂O as the magma decompresses (Gaillard & Scaillet, 2009), although CO₂ can be important at high bulk CO₂.

We can do a thought experiment using a simplified version of the calculations with bulk concentrations of 3 wt.% H₂O + 1000 $\mu\text{g/g}$ CO₂ + 1000 $\mu\text{g/g}$ χ at $\Delta\text{FMQ}_0=0$ to explore the dilution effect during closed-system degassing (i.e., black dashed curves in Figure 4 are reproduced in Figure 7a–c). For these conditions, the melt becomes vapor saturated at 263.7 MPa (i.e., P_{sat}^v = black star) at which point there is no vapor (Figure 7d). With further closed-

system degassing to 260.0 MPa, the concentrations of H₂O and χ in the melt are essentially the same as at P_{sat}^v because of their high solubility functions, but the CO₂ content of the melt has decreased to 978 $\mu\text{g/g}$ (Figure 7a–c and e). There is a small amount of coexisting vapor (0.003 wt.%; its volume relative to the melt is exaggerated in Figure 7e to be visible) with a composition of ~69 mol.% CO₂, ~27 mol.% H₂O, and ~1 mol.% χ (their relative molar proportions are shown by their relative area in the vapor in Figure 7e).

We now instantaneously decompress the melt + vapor to 96.2 MPa, at which point 10% of the dissolved χ would have degassed under equilibrium conditions. However, we prevent CO₂ and χ from degassing such that the concentration of CO₂ and χ in the melt and number of moles of CO₂ and χ in the vapor stay as they were at 260.0 MPa (Figure 7b, c, and f). Only H₂O is allowed to degas (i.e., its concentration in the melt decreases and the number of moles of H₂O in the vapor increases), which we do in a simplified way by setting the H₂O content of the melt to that of the equilibrium value for the true degassing calculation (i.e., 2.89 wt.%, the open black diamond in Figure 7a) and calculating the number of moles of H₂O transferred to the vapor. One way to think of this is as if there were a semi-permeable membrane present between the melt and vapor that lets H₂O degas, but not CO₂ or χ . Something like this might be approximated in nature when degassing occurs during high magma ascent rates. In such cases, H₂O nearly approaches equilibrium between the melt and vapor (due to its high diffusivity in silicate melt, which controls the rate of transfer of H₂O from melt to vapor bubbles), whereas CO₂ could be largely retained in the melt due to its slower diffusivity relative to H₂O (e.g., Zhang & Ni, 2010). Such behaviour was observed in the experiments of Pichavant et al. (2013) and Le Gall & Pichavant (2016).

This results in a melt containing 2.89 wt.% H₂O, ~978 $\mu\text{g/g}$ CO₂, and ~1000 $\mu\text{g/g}$ χ , with a vapor composition of nearly pure-H₂O and negligible CO₂ ($\sim 10^{-3}$ mol.%) and χ ($\sim 10^{-4}$

mol.%) (Figure 7a–c and f). Note that the system is actually vapor-undersaturated because the sum of the partial pressures in the vapor does not equal the total P (H_2O would need to degas more than in the equilibrium degassing calculation to compensate for the lack of CO_2 and χ degassing) but for the purpose of this illustration, this is not quantitatively important. The amounts of CO_2 and χ in the vapor have been highly diluted by the degassed H_2O , causing the melt and vapor to be highly out of equilibrium for CO_2 and χ (Figure 7b, c, and f). For instance, the equilibrium constant for χ (K_χ) is $320 \mu\text{g/g/MPa}$ but the ratio of w_χ^m to p_χ is $>80,000 \mu\text{g/g/MPa}$! Hence, for CO_2 and χ to achieve equilibrium between the melt and vapor they must degas, lowering their melt-vapor ratios until they reach their equilibrium constants (eq. 1, Figure 7g). This highlights how dilution can occur without the external addition of a volatile, but instead by degassing of a volatile already present in the system.

This analysis of how the transfer of an abundant volatile component during degassing can drive enhanced transfer of other dissolved species from the melt to the vapor highlights that the dilution effect can be quite dramatic. For instance, based on our modeling using *VoFe*, increasing the bulk H_2O from 0.2 to 5.0 wt.% with $1000 \mu\text{g/g CO}_2 + 1000 \mu\text{g/g S}$ at $\Delta\text{FMQ}_0=0$ increases $P_{10\%}$ from ~ 181 to ~ 343 MPa for CO_2 and ~ 4 to ~ 206 MPa for S (Figure 1b and c); or increasing the bulk CO_2 from 0.1 to 1.0 wt.% with 0.2 wt.% $\text{H}_2\text{O} + 1000 \mu\text{g/g S}$ at $\Delta\text{FMQ}_0=0$ increases $P_{10\%}^S$ from ~ 4 to ~ 187 MPa (Figure 2c). The effect of dilution by one volatile (e.g., H_2O) on the degassing behaviour of another volatile (e.g., CO_2 and S) depends on their relative equilibrium constants (e.g., eq. 1) and their relative concentrations of the volatiles (Dixon & Stolper, 1995). All things being equal, for either: (1) a smaller equilibrium constant, or (2) a higher concentration of a volatile, the deeper its $P_{10\%}$ and the larger its dilution effect on other volatiles (see Supplementary Material Section 3.6 for more details). At $\Delta\text{FMQ}_0=0$, H_2O is the most soluble species, followed by S, and CO_2 is the most insoluble

(i.e., H₂O has the largest equilibrium constant, and CO₂ the smallest). Water has a large dilution effect on S (and CO₂) in hydrous magmas because of the high concentration of H₂O relative to S (and CO₂: 0.1 vs. up to 5 wt.% H₂O). If the equilibrium constant for S or its typical concentration in melts were higher or lower, the impact of dilution by H₂O would be different and S could even become a more important diluent (Figure S5–7).

The long-tail of higher *P* degassing of S (up to ~10% of S for the degassing curves in Figure 2c) is driven by dilution of S by CO₂, because total S has a larger effective equilibrium constant compared to that of CO₂. Although H₂O is an important diluent in igneous processes, CO₂ concentrations on the order of a weight percent (or higher) in some basaltic parental magmas mean it can also be an important driver of deepened S degassing under certain situations, even surpassing the influence of H₂O depending on the specific bulk concentrations of H₂O and CO₂ (Figure 2c; e.g., Anderson et al., 2024, 2025). With growing observational evidence for high bulk CO₂ contents in arc basalts and OIBs (e.g., Barth et al., 2024; Blundy et al., 2010; Burton et al., 2023; Dayton et al., 2024; Dixon et al., 1997; Huang et al., 2026; Rasmussen et al., 2020), the influence of dilution by CO₂ on the behaviour of S degassing maybe common place. Note that as a general rule, volatiles that have high solubility functions (i.e., that are highly soluble in the melt, such as Cl and Br; e.g., Gorjovsky et al., 2026; Rusiecka & Wood, 2026; Thomas & Wood, 2022) will be less affected by dilution than those that are relatively insoluble in the melt (e.g., Figure S7).

It should be emphasized that *f*_{O₂} *does* affect the speciation of S in both the melt and vapor. Thus, the analogy between the behavior of S and that of a simplified inert and ideal “ χ ” component during H₂O-dominated dilution degassing has its limits. In particular, care should be taken when comparing processes across large ranges of *f*_{O₂}, over which S-speciation in the melt and vapor can change significantly. For example, as emphasized earlier, the concentration of total S in vapor-saturated basaltic melts has a minimum near the

f_{O_2} of the SS^{min} , and the presence of H_2S in the melt and/or vapor and its effects on the total S solubility function are also f_{O_2} -dependent (e.g., Figure 3). However, as discussed previously, these effects are mostly secondary relative to the effect of dilution by H_2O on the effects of degassing of S from hydrous magmas. The dilution effect we have described here is inherent to any degassing models during closed-system degassing that include mass balance between the melt and vapor and requires that the total P is the sum of all the partial pressures.

Further impacts of dilution-driven degassing in natural magmas

It is beyond the scope of this paper to delve into all possible petrological and volcanological observations that could be related to the dilution effect of H_2O (and CO_2) on S (and CO_2) degassing. We comment here briefly on how dilution by H_2O may contribute to understanding differences in the behaviour of S and CO_2 during degassing of magmas in different tectonic settings and to interpreting the C/S ratios of volcanic gases.

Extent of dilution-driven degassing of sulfur by water across tectonic settings

The bulk H_2O content of magmas varies markedly across different tectonic settings (e.g., Wallace et al., 2015). The S contents of submarine MORBs suggest that little if any S degassing occurs prior to their eruption on the sea floor (e.g., Moore & Fabbi, 1971; Wallace & Carmichael, 1992; Wallace & Edmonds, 2011), unless they reach shallow water depths (e.g., <200 m on the Reykjanes Ridge; Moore & Schilling, 1973). This is consistent with their low H_2O content (~0.2 wt.%; e.g., Wallace et al., 2015), which results in little dilution of S by H_2O degassing (e.g., red curve for 0.2 wt.% bulk H_2O in Figure 1c), leading to the conclusion that S degassing is at most a minor phenomenon for most submarine MORB magmas. Similarly, magmas at Kīlauea, Mauna Kea, and Samoa are thought to degas S at

relatively shallow depths (e.g., Dixon et al., 1991; Gerlach & Thomas, 1986; Labidi et al., 2015; Lerner et al., 2021; Moore & Clague, 1987; Moore & Fabbi, 1971; Moore & Schilling, 1973; Moore & Thomas, 1988; Moussallam et al., 2016; Wallace & Carmichael, 1992; Wallace & Edmonds, 2011), which is consistent with their low H₂O contents (~0.3–0.8 wt.%; e.g. Seaman et al., 2004; Workman et al., 2006), again causing some – but relatively little – dilution driven S degassing. However, hydrous OIBs (such as El Hierro, Canary Islands, at ~3 wt.% H₂O) display higher *P* degassing of S due to their elevated bulk H₂O (e.g., Longpré et al., 2017). Additionally, water-poor magmas closer to the SS^{min} (e.g., Mauna Loa; Wieser et al., 2025) display degassing at higher *P* than magmas with higher or lower initial *f*_{O₂} (e.g., Kīlauea) due to the lower solubility function of total S at the SS^{min}.

In contrast, arc magmas are hydrous (average ~4 wt.% and potentially up to 7 wt.%; e.g., Plank et al., 2013; Wallace et al., 2015), which causes large amounts of dilution of S by H₂O and higher pressure degassing of S in these magmas (e.g., Figure 1c and Figure S4c and f). This transfers S from the melt to the vapor, which can be released at higher *P* from ascending basaltic magmas. If this vapor decouples from the ascending magma (i.e., approximating open-system degassing), the vapor can migrate to shallower, more evolved magmas, providing additional S which could then contribute to understanding the “excess sulfur problem” (e.g., Christopher et al., 2010; Di Muro et al., 2008; Edmonds et al., 2010; Pallister et al., 1992; Roberge et al., 2009; Wallace, 2001). Similarly, this S derived from higher *P* degassing of basaltic magmas may contribute to triggering extensive precipitation of sulfide ores in porphyry copper deposits (e.g., Blundy et al., 2015; Hattori & Keith, 2001). These effects are amplified by these magmas often having an *f*_{O₂} near the SS^{min}, decreasing the solubility function of S and increasing even further the potential impact of deeply degassed S on shallower magmas and eruptive processes (e.g., Ding et al., 2023; Hughes et al., 2023; Nilsson & Peach, 1993). However, the formation of sulfide and/or sulfate phases, as well as

changes in melt composition due to crystallisation, may enhance or reduce the magnitude of the increase in P of S degassing depending on the amount of S available to be stored in the melt and the melts capacity to do so.

Influence of dilution-driven degassing of carbon dioxide by water on vapor C/S ratios

Higher P degassing of S and CO₂ from water-rich arc and OIB magmas would also influence the C/S ratio of gases measured at volcanic vents, which is commonly used to monitor volcanoes because of its link to the depth of vapor segregation from the magma (i.e., magma depth; Aiuppa et al., 2007, 2017; Burton et al., 2007; de Moor et al., 2016; Ding et al., 2025; Giggenbach, 1996; Iacovino, 2015; Kern et al., 2022). In volcanoes without significant hydrothermal systems that can scrub S, CO₂-rich gases (i.e., high C/S ratios) are assumed to reflect gases separating from magmas that are degassing at higher P because of the relatively low solubility function of CO₂ compared to S (e.g., Aiuppa et al., 2007; Kern et al., 2022). Likewise, the high solubility function of S relative to CO₂ has typically been taken to mean a magma would need to reach relatively low P before it would degas significant S and generate vapors with low C/S (e.g., Aiuppa et al., 2007; Kern et al., 2022). Together, these lines of reasoning have led to the expectation that C/S ratios in volcanic gases would decrease as magmas degas during ascent (e.g., Aiuppa et al., 2007; Kern et al., 2022). Similarly, the C/S ratio of fluid inclusions can be used as a barometer (e.g., Wieser et al., 2025).

Figure 8 demonstrates that the large dilution effect of H₂O results in C/S ratios that – at any given P during closed- or open-system degassing – are also highly dependent on the bulk H₂O content of the magma. However, in this case it is the dilution effect of H₂O on CO₂ rather than on S, that is driving these variations. Although the mole fraction of total S in the vapor during degassing does vary depending on bulk H₂O, the changes in total C in the vapor

exert considerable control on the shape of the C/S curves shown in Figure 8 (more details in Supplementary Material Section S3.7). Lower C/S ratios at a given P are characteristic of the vapor if the bulk H₂O content of the magma is higher, reflecting the more effective “driving” of CO₂ from melt to vapor via dilution (i.e., $P_{10\%}^C > P_{10\%}^S$; Figure 8a). Therefore, knowledge of the initial H₂O content of the melt is required to estimate accurate depths of magmatic degassing from the C/S ratio of the gas (e.g., Ding et al., 2025; Hughes et al., 2025). For example, a C/S ratio of 20 could be produced by closed system degassing from a magma at ~14 MPa with 0.2 wt.% bulk H₂O or ~271 MPa for 5 wt.% bulk H₂O at $\Delta FMQ_0=0$ (Figure 8a). The initial value of f_{O_2} (i.e., ΔFMQ_0) can have comparably large effects on the C/S ratio of the vapor as does the bulk H₂O content: for 3 wt.% H₂O, a C/S ratio of 20 is produced at ~158 MPa for $\Delta FMQ_0=0$, not at all at the $SS^{\min}$, and ~250 MPa for $\Delta FMQ_0=2.5$ (Figure 8c). This highlights the potential complexities for interpreting C/S ratio in volcanic gases and fluid inclusions as barometers. It should also be noted that different degassing tools currently give very different results for the same conditions for the vapor phase; this discrepancy must also be resolved to enable accurate barometry from volcanic gases and fluid inclusions (Figure S2d and h; e.g., Ding et al., 2025; Hughes et al., 2025; Iacovino et al., 2026).

The model C/S curves for 3 and 5 wt.% H₂O are strongly concave up and nearly vertical at $P \lesssim 100$ MPa and 200 MPa, respectively (the approximate P 's at which substantial degassing of H₂O begins to degas, Figure 1a and Figure 8a and b). In contrast, for 0.2 and 1.0 wt.% H₂O, the degassing curves are concave down for most of this shallow P range (Figure 8b). As a result, the C/S ratio during shallow degassing of water-rich magmas is not expected to vary much during their final ascent, in contrast to relatively water-poor magmas (Figure 8b).

Magmas are thought to stall near the depth of substantial degassing of H₂O due to the increased magma viscosity triggered by dehydration-driven crystallisation and increased melt

viscosity (e.g., Cashman & Blundy, 2000; Ding et al., 2025; Rasmussen et al., 2022). Thus, as a consequence of the effects of dilution by H₂O on the degassing of CO₂, our calculations suggest that shallow magma ascent after potential stalling for water-rich magmas may be essentially “invisible” using the C/S ratio, potentially reducing its effectiveness at monitoring magma ascent prior to eruption. For example, for a bulk composition containing 5 wt.% H₂O, the C/S ratio of the vapor only changes from ~1.3 to ~0.7 over the last 100 MPa to the surface (Figure 8b); this would be a subtle signal to resolve depending on how noisy the data are. Moreover, the absolute change in C/S will depend on the bulk volatile contents, initial f_{O_2} , melt compositions and T (e.g., Figure 8). Additionally, although we have not analysed them quantitatively, the influences of sulfide and/or sulfate precipitation, as well as changes to melt composition due to crystallisation and therefore its capacity to dissolve sulfur and CO₂, will also influence the C/S ratio.

Summary

In water-poor magmas, S usually begins to degas significantly at shallow levels based on observations of MORBs and some OIBs; its known partitioning between melt and vapor; and its typically low concentrations in magmas (e.g., Dixon et al., 1991; Labidi et al., 2015; Lerner et al., 2021; Moore & Clague, 1987; Moore & Fabbi, 1971; Moore & Schilling, 1973; Moore & Thomas, 1988; Moussallam et al., 2016; Wallace & Carmichael, 1992; Wallace & Edmonds, 2011). However, hydrous magmas are thought to degas S at much greater depths based on observations from arc volcanoes, experiments, and numerical modelling of degassing (e.g., Boulliang & Wood, 2022; Brounce et al., 2016; Davis et al., 1991; Ding et al., 2023; Edmonds & Wallace, 2017; Nilsson & Peach, 1993; Rasmussen et al., 2020; Wallace & Edmonds, 2011; Yip et al., 2022). Moreover, at a given initial f_{O_2} and amount of

dissolved S, the magnitude of the increase in P of significant degassing of S is strongly and positively correlated with the amount of dissolved H₂O based on modelling. The higher P degassing of S from hydrous mafic magmas is likely important in igneous petrology, volcanology, and ore genesis in that it provides a mechanism for deep mobilization of S into the vapor phase (e.g., Christopher et al., 2010; Di Muro et al., 2008; Edmonds et al., 2010; Hattori & Keith, 2001; Pallister et al., 1992; Roberge et al., 2009; Wallace, 2001).

Explanations of this large effect of H₂O on the depth of S degassing have mostly focussed on changes in the partitioning of S in the melt and vapor related to hydrous magmas that are also often oxidised (e.g., Ding et al., 2023; Métrich et al., 2009; Nilsson & Peach, 1993; Wallace & Edmonds, 2011; Zajacz et al., 2012). However, the influence of the degassing of other volatiles such as CO₂ and H₂O (e.g., Davis et al., 1991; Dixon & Stolper, 1995; Wallace & Edmonds, 2011) and the effects of the dilution of S in the vapor by H₂O as it degasses (Gaillard & Scaillet, 2009) have also been implicated.

In this paper, we use the Python package *VolFe* (Hughes et al., 2025) to explore the effects of varying the bulk H₂O content and f_{O_2} of a basaltic melt on the degassing behavior of S. We find that an inert and ideal volatile species also displayed higher P degassing with increasing bulk H₂O, demonstrating that the complex chemical interactions between H₂O and S in melt and vapor are not the main drivers of higher P degassing S from water-rich melts.

We propose that the increased P of S degassing that correlates with increases in the bulk melt H₂O content is dominantly an example of the “dilution” effect during degassing (e.g., Anderson et al., 2024, 2025; Botcharnikov et al., 2007; Dixon & Stolper, 1995; Gaillard & Scaillet, 2009; Gonnermann & Mukhopadhyay, 2007; Holloway, 1976; Iacovino et al., 2021; Lowenstern, 2001). In its simplest terms for this case, transfer of H₂O from melt to vapor during decompression “dilutes” all other components in the vapor (i.e., decreases their thermodynamic activity and fugacity in the vapor; e.g., Gaillard & Scaillet, 2009). As a

result, the vapor becomes undersaturated with respect to the melt in S, C, and any other volatiles in the system, and melt-vapor equilibrium is only re-achieved by transfer of S, C, etc. from the melt into the vapor (i.e., degassing), increasing their activities and fugacities until the vapor and the melt are in equilibrium. High bulk CO₂ concentrations (≥ 1 wt.%) have a similar effect and also drive higher P degassing of S. The sulfur solubility minimum additionally increases the P of degassing of S due to the lower total solubility function of S (e.g., Ding et al., 2023; Hughes et al., 2023; Nilsson & Peach, 1993).

Based on our numerical experiments, the details of the dilution process depend strongly both on (1) the equilibrium (or solubility) constant of the reaction describing equilibrium between melt and vapor for the volatile component of interest relative to that of H₂O; and (2) and the relative concentrations of the volatile component of interest and of H₂O – or any other diluent of potential interest (Dixon & Stolper, 1995). For systems with significantly different absolute and relative concentrations of H₂O, CO₂, and S; f_{O_2} ; major element melt composition; and additional volatile components (e.g., rare gases, Cl, etc.), *VolFe* and/or other degassing tools (e.g., Iacovino et al., 2026) could be readily modified to anticipate the consequences of dilution in a broader petrological and volcanological context.

Data availability statement

The *VolFe* Python package is freely available on GitHub (<https://github.com/eryhughes/VolFe>) and a version has been archived in Zenodo (Hughes et al., 2025b). The Jupyter Notebook to execute the calculations using *VolFe* and *Sulfur_X* and reproduce the graphs in this paper and all model outputs presented in this study are included as Supplementary Material.

827 **Funding**

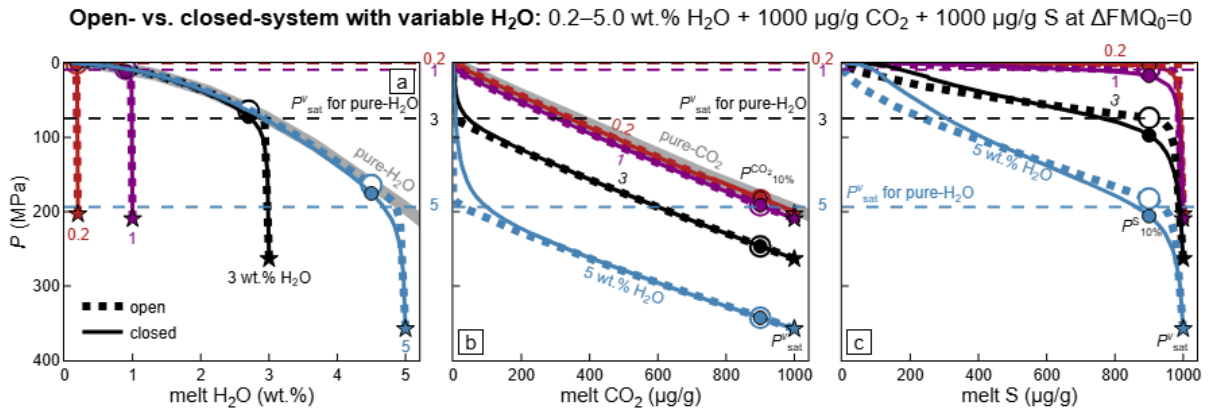
828 This work was partly supported by the New Zealand Ministry of Business, Innovation and
829 Employment (MBIE) through the Strategic Science Investment Fund – Hazards and Risk
830 Management [contract C05X1702, supporting ECH].

831

832 **Acknowledgements**

833 We thank an anonymous reviewer, Bruno Scaillet, and Terry Plank for their thoughtful and
834 constructive comments that greatly improved the paper, and Mary Reid for their editorial
835 handling of the paper.

836 Figures



837

838 **Figure 1.** Effect of varying bulk H₂O on the melt volatile content during equilibrium,
839 isothermal, decompression-induced, closed- and open-system degassing. The initial conditions
840 are $T = 1200$ °C and bulk concentrations of variable H₂O + 1000 µg/g CO₂ + 1000 µg/g S at
841 $\Delta FMQ_0=0$, where curve colour indicates bulk H₂O: 0.2 (red), 1.0 (purple), 3.0 (black), and 5.0
842 (blue) wt.% (labelled on each curve); and curve style indicates degassing style: closed (solid)
843 and open (dotted). Concentration dissolved in the melt of (a) H₂O, (b) CO₂, and (c) S. Symbols
844 indicate the location of P_{sat}^v (star) or $P_{10\%}^i$ (circle), which are labelled in (b) and (c): filled
845 symbols are for closed-system and open symbols for open-system calculations. The closed-
846 system degassing curves for pure-H₂O and CO₂ are shown as a thick grey curves in (a) and (b),
847 respectively (not shown in (c): the same melt containing only 1000 µg/g S would begin
848 degassing at ~0.5 MPa, but this is not shown for clarity; Hughes et al., 2023), and the thin-
849 horizontal-dashed lines in all panels show the P where pure-H₂O would begin to degas (i.e.,
850 P_{sat}^v for pure-H₂O with the same bulk H₂O, labelled in between panels and using the same line
851 colour).

852 **Alt text:** Graphs showing the concentration of water, carbon dioxide, and sulfur dissolved in
853 the melt decreasing with decreasing pressure during open- and closed-system degassing for
854 four different initial water concentrations of 0.2, 1, 3, and 5 weight percent. The depth at which

855 ten percent of water and sulfur has degassed is similar to the depth at which the same initial
856 concentration of water in the pure-water system would have degassed.

857

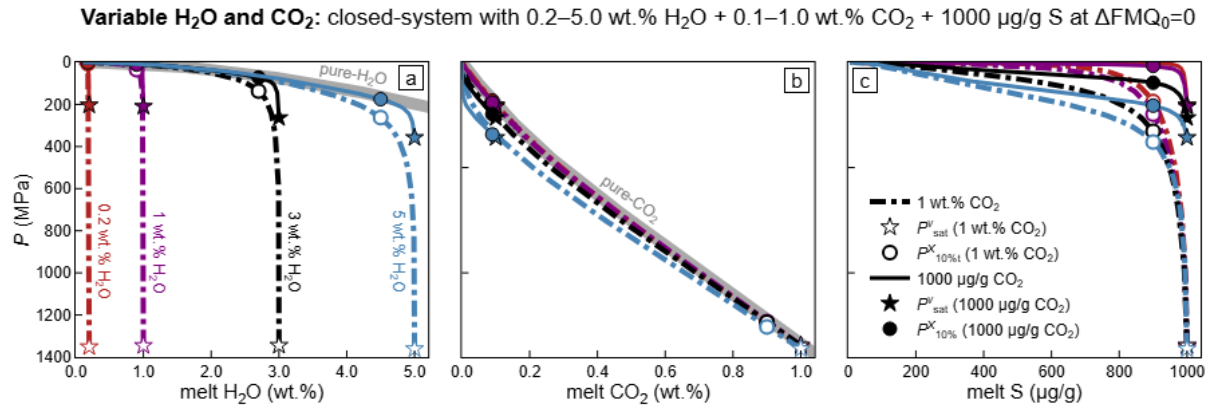
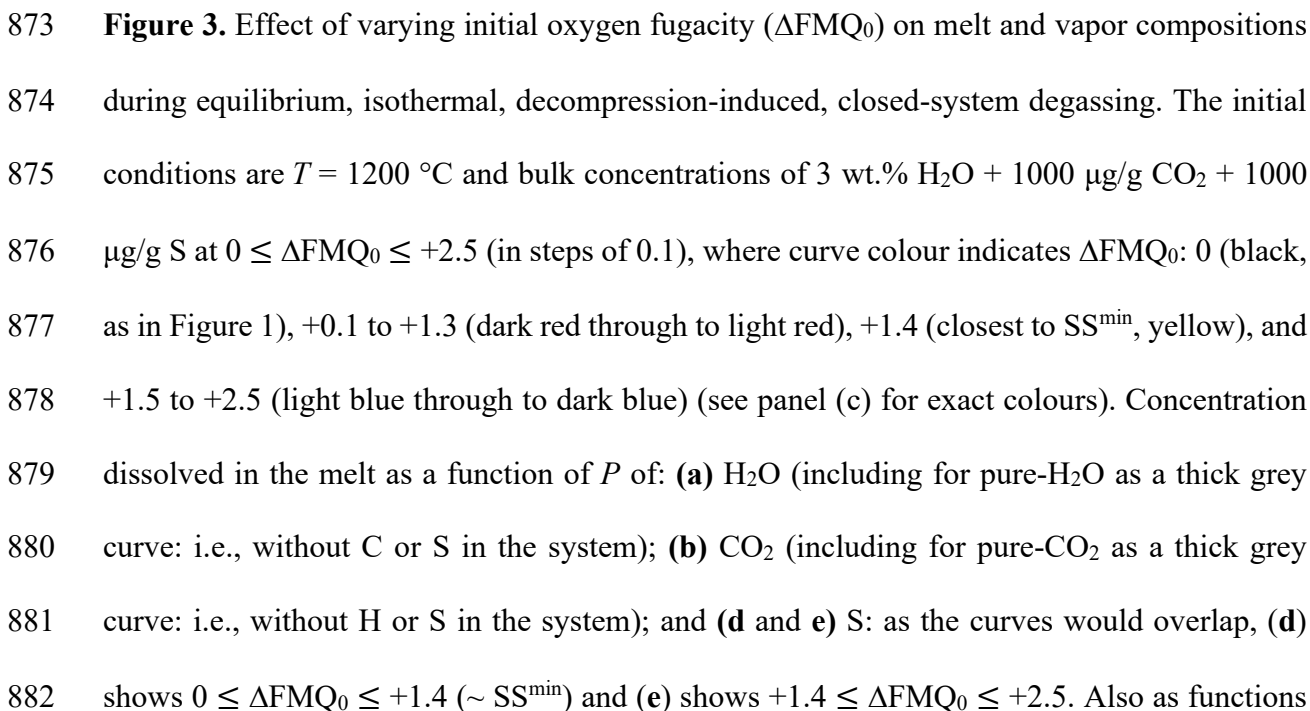


Figure 2. Effect of varying bulk H₂O and CO₂ on the melt volatile content during equilibrium, isothermal, decompression-induced, closed-system degassing. The initial conditions are $T = 1200$ °C and bulk concentrations of variable H₂O + variable CO₂ + 1000 µg/g S at $\Delta FMQ_0=0$. Concentration dissolved in the melt of (a) H₂O, (b) CO₂, and (c) S. Colour, style, and shapes of curves, lines, and symbols as in Figure 1 for 1000 µg/g bulk CO₂ (i.e., solid curves and filled symbols) and pure-H₂O and CO₂ (thick grey curves), with the addition that dot-dashed curves and open symbols indicate 1 wt.% bulk CO₂.

Alt text: Graphs showing the decreasing concentrations of water, carbon dioxide, and sulfur dissolved in the melt with decreasing pressure during closed-system degassing for four different initial water concentrations of 0.2, 1, 3, and 5 weight percent and two different initial carbon dioxide concentrations of 0.1 and 1 weight percent. The depth at which ten percent of water and sulfur has degassed increases for 1 weight percent bulk carbon dioxide compared to 0.1 weight percent.

872



883 of P : (c) oxygen fugacity, where the high- P end of each curve corresponds to ΔFMQ_0 (i.e., at
884 P_{sat}^v , stars); and $P_{10\%}^S$ is shown by circles (corresponds to the black vertical arrows in d and e)
885 – the horizontal black-dashed line shows for comparison the P_{sat}^v for a system with 3 wt.%
886 pure- H_2O (i.e., no C or S) would begin degassing; (f) $\text{Fe}^{3+}/\text{Fe}_T$ in the melt; (g) $\text{SO}_2/(\text{SO}_2+\text{H}_2\text{S})$
887 in the vapor; (h) the proportion of S dissolved as H_2S relative to total S ($\text{S}_{\text{H}_2\text{S}}/\text{S}_T$) in the melt;
888 and (i) S^{6+}/S_T in the melt. The direction of increasing ΔFMQ_0 in panels (c–f) is shown by a
889 black, thin arrow and labelled ‘ $\uparrow\Delta\text{FMQ}_0$ ’.

890 **Alt text:** Graphs showing the melt and vapor compositions during degassing for a melt initially
891 containing 3 weight percent water, 1000 micro-grams per gram carbon dioxide, and 1000
892 micro-grams per gram sulfur at initial oxygen fugacities ranging from 0 to plus 2.5 delta F M
893 Q. All values are against pressure. Panel A shows water concentration in the melt. Panel B
894 shows carbon dioxide concentration in the melt. Panel C shows oxygen fugacity. Panel D
895 shows sulfur concentration in the melt for initial oxygen fugacities from 0 to plus 1.4 delta F
896 M Q, which is at the sulfur solubility minimum, whilst plus 1.4 to plus 2.5 are shown in Panel
897 E. Panel F shows the iron 3 plus over total iron ratio in the melt. Panel G shows the sulfur
898 dioxide over sulfur dioxide plus hydrogen sulfide ratio in the vapor. Panel H shows the sulfur
899 as hydrogen sulfide over total sulfur ratio in the melt. Panel I shows the sulfur 6 plus over total
900 sulfur ratio in the melt.

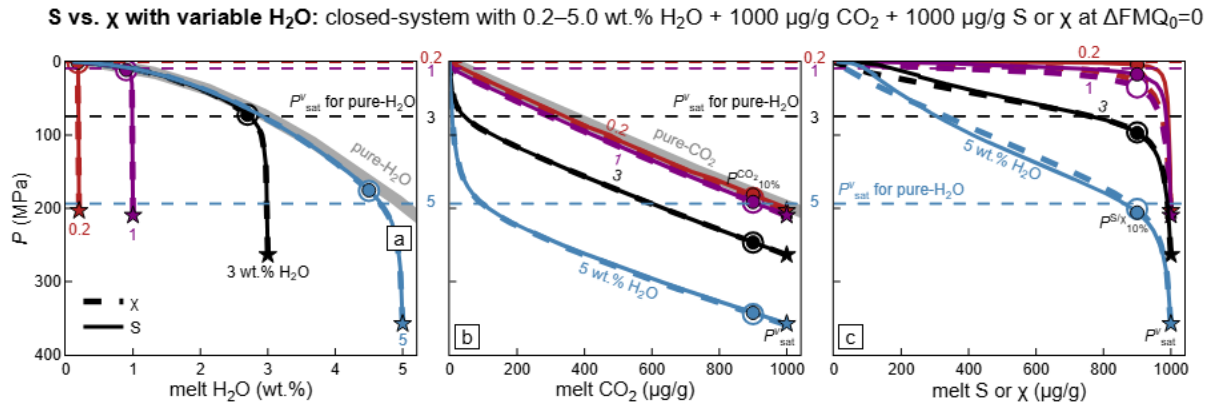
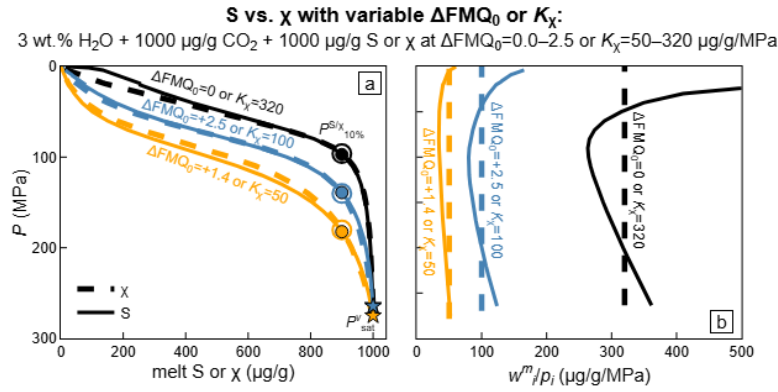


Figure 4. Comparison of the effect of including S or χ and varying bulk H₂O on the melt volatile content during equilibrium, isothermal, decompression-induced, closed-system degassing. The initial conditions are $T = 1200$ °C and bulk concentrations of variable H₂O + 1000 $\mu\text{g/g}$ CO₂ + 1000 $\mu\text{g/g}$ S or χ at $\Delta\text{FMQ}_0=0$. Concentration dissolved in the melt of (a) H₂O, (b) CO₂, and (c) S or χ . Colour, style, and shapes of curves, lines, and symbols as in Figure 1, with the addition that dashed curves and open symbols indicate the behavior of χ -bearing systems, while solid curves and filled systems indicate the behavior of S-bearing systems (reproduced from Figure 1).

Alt text: Graphs showing the concentration of water, carbon dioxide, and sulfur or chi dissolved in the melt decreasing with decreasing pressure during closed-system degassing for three different initial water concentrations of 0.2, 1, 3, and 5 weight percent. The depth at which ten percent of water, sulfur, and chi has degassed is similar to the depth at which ten percent water in the pure-water system would have degassed. The behaviour of sulfur and chi is very similar.

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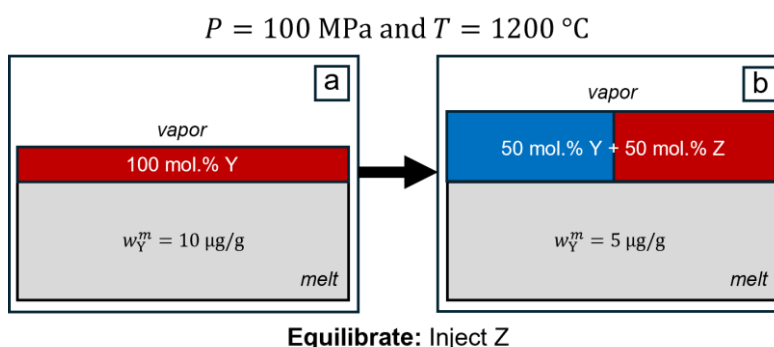
918

919 **Figure 5.** The effect of varying ΔFMQ_0 or K_χ during equilibrium, isothermal, decompression-
 920 induced, closed-system degassing. The initial conditions are $T = 1200^\circ\text{C}$ and bulk
 921 concentrations of 3 wt.% H_2O + 1000 $\mu\text{g/g}$ CO_2 + 1000 $\mu\text{g/g}$ S or χ at either variable ΔFMQ_0
 922 for S or variable K_χ for χ ($\Delta FMQ_0=0$), where curve colour indicates ΔFMQ_0 or K_χ ($\mu\text{g/g/MPa}$):
 923 $\Delta FMQ_0=0$ or $K_\chi=320$ (black), $\Delta FMQ_0+1.40$ ($\sim SS^{\min}$) or $K_\chi=50$ (yellow), and $\Delta FMQ_0+2.5$ or
 924 $K_\chi=100$ (blue); and curve style indicates S (solid) or χ (dash). **(a)** Concentration in the melt of
 925 S or χ ; and **(b)** w^m_i/p_i vs. P , which is an equilibrium constant (K_i , eq. 1) – for S, p_i is the sum of
 926 p_{SO_2} and $p_{\text{H}_2\text{S}}$.

927 **Alt text:** Graphs comparing the behaviour of sulfur at different initial oxygen fugacities and
 928 chi with different equilibrium constants that have been chosen to match the behaviour of sulfur
 929 during degassing. Panel A shows the concentration of sulfur and chi decreasing very similarly
 930 with decreasing pressure. Panel B shows the equilibrium constants for sulfur and chi with
 931 pressure.

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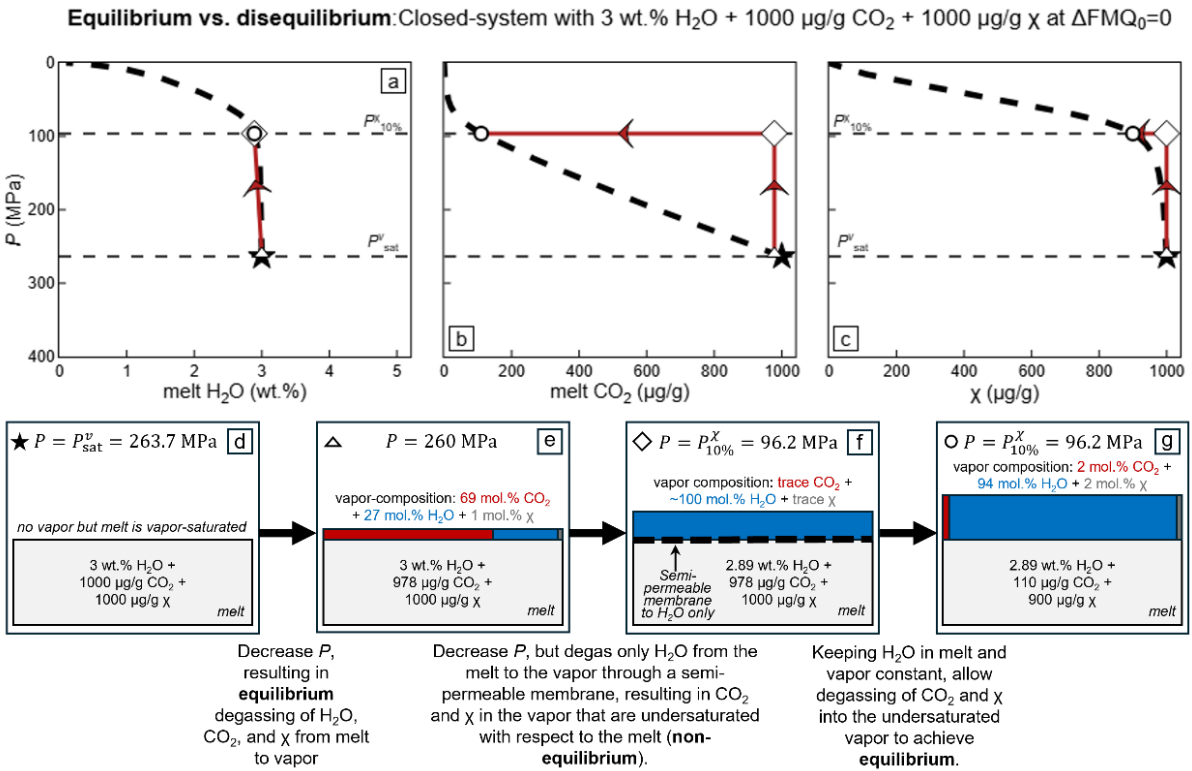


934

935 **Figure 6.** Schematic illustration of dilution driven degassing of a vapor-saturated silicate melt
 936 by isobaric, isothermal addition of an insoluble volatile species into the system. **(a)** Initially, at
 937 $P = 100 \text{ MPa}$ and $T = 1200 \text{ }^{\circ}\text{C}$, the vapor-saturated melt contains $10 \text{ } \mu\text{g/g}$ of Y because the
 938 vapor is pure Y (which is an ideal gas) and the equilibrium constant (see eq. 1) for Y (K_Y) is
 939 $0.1 \text{ } \mu\text{g/g/MPa}$. Component Z, which is assumed to be insoluble in the silicate melt, is added to
 940 the vapor in **(a)** at constant $P = 100 \text{ MPa}$ until the vapor is 50 mol.% Y and 50 mol.% Z, whilst
 941 Y is equilibrating continuously between the melt and vapor. **(b)** The vapor is now a 50:50
 942 mol.% mix of Y and Z and, to maintain equilibrium, Y has degassed until there is $5 \text{ } \mu\text{g/g}$ in the
 943 melt. The vapor volume has increased to maintain constant P as the number of moles in the
 944 vapor has increased.

945 **Alt text:** Schematic illustration of changes in melt and vapor composition at constant pressure
 946 and temperature when species Z is injected to a system only containing species Y. Panel A
 947 shows the initial system containing 10 micro grams per gram of species Y dissolved in the melt
 948 and a vapor of 100 mole percent species Y. Panel B shows the system after species Z has been
 949 added, which now contains 5 micro grams per gram of species Y dissolved in the melt and a
 950 vapor containing 50 mole percent of species Y and 50 mole percent of species Z.

951



953

954 **Figure 7.** Degassing curves and schematic illustrations of the thought experiment described in

955 the text illustrating the effect of dilution by H₂O on CO₂ and χ during isothermal,

956 decompression-induced, closed-system degassing for initial conditions of T = 1200 °C and 3

957 wt.% H₂O-eq + 1000 µg/g CO₂-eq + 1000 µg/g χ at ΔFMQ₀=0. The thick, black-dashed curves

958 for **(a)** H₂O, **(b)** CO₂, and **(c)** χ show their concentrations during equilibrium, closed-system

959 degassing (reproduced from Figure 4). The schematic illustrations in **(d–g)** show (1) the

960 evolution after each step in the thought experiment described in the text of the concentrations

961 in the melt of H₂O, CO₂, and χ (indicated in the light grey boxes labelled “melt”); and (2) the

962 mol.% in the vapor of CO₂ (red), H₂O (blue), and χ (dark grey) written above the coloured

963 boxes and by the areas of these coloured boxes, the sums of which are schematically indicative

964 of the total volume of vapor present in the system (the size of the melt and vapor boxes are not

965 accurate). Panel **(d)** shows the system at P = P_{sat} = 263.7 MPa (filled black star in **a–c**):

966 although the melt is vapor-saturated at this point, there is no vapor present. Panel **(e)** shows the

967 system after the system has decompressed from 263.7 MPa to $P = 260.0$ MPa (the open triangle
968 in **a–c**), having undergone equilibrium degassing of H_2O , CO_2 , and χ (i.e., to the open triangle
969 plots on the dashed black curve). Panel **(f)** shows the effects of decompressing the system from
970 $P = 260.0$ MPa to $P = 96.2$ MPa ($P_{10\%}^\chi$) on the vertical red path from the open triangle to the
971 open diamond in **a–c**. During this decompression, H_2O is allowed to degas, through a semi-
972 permeable membrane with no degassing of CO_2 or χ . Finally, in panel **(g)** the semi-permeable
973 between melt and vapor is removed at constant P , allowing equilibration of the melt and vapor
974 by degassing of CO_2 and χ without further H_2O degassing (open circle in **a–c**).

975 **Alt text:** Graphs and schematic illustration of changes in the melt and vapor composition
976 during decompression-induced degassing. Panel **A** shows the concentration of water dissolved
977 in the melt decreasing with decreasing pressure. Panel **B** shows the concentration of carbon
978 dioxide dissolved in the melt decreasing with decreasing pressure. Panel **C** shows the
979 concentration of chi dissolved in the melt with decreasing with decreasing pressure. Panel **D**
980 shows the initial system at 263.7 MPa with all volatiles dissolved in the melt. Panel **E** shows
981 the system at 260 MPa with a small amount of vapor after equilibrium degassing. Panel **F**
982 shows the system at 96.2 MPa where only water has degassed into the vapor but the
983 concentrations of carbon dioxide and χ are artificially retained in the melt (i.e., by separation
984 of the melt and vapor in this step by a membrane semi-permeable to CO_2 and χ) and are
985 therefore highly diluted in the vapor. Panel **G** shows the system at 96.2 MPa at equilibrium,
986 where carbon dioxide and chi have degassed from the melt to the vapor.

Open- vs. closed-system with variable H₂O and ΔFMQ_0 : 0.2–5.0 H₂O + 1000 $\mu\text{g/g}$ CO₂ + 1000 $\mu\text{g/g}$ S at $\Delta FMQ_0=0.0\text{--}2.50$

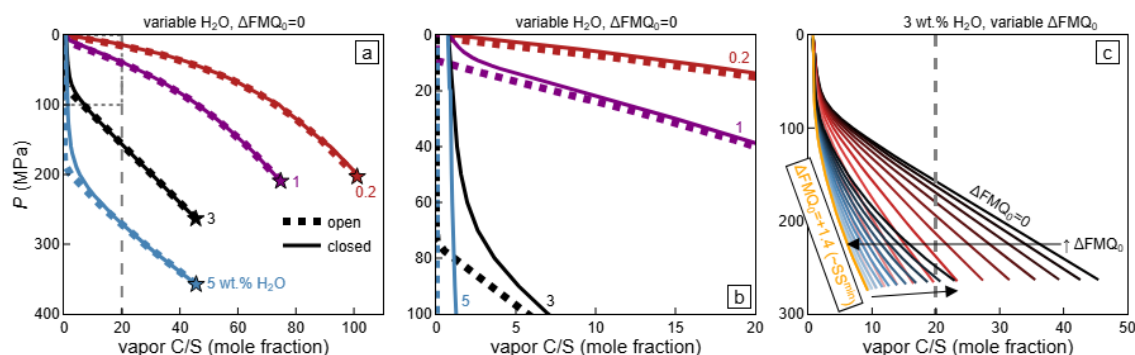


Figure 8. Effect of varying bulk H₂O and initial oxygen fugacity (ΔFMQ_0) on the C/S ratio of vapor during equilibrium, isothermal, decompression-induced, closed- and open-system degassing. The initial conditions are $T = 1200$ °C and bulk concentrations of variable H₂O + 1000 $\mu\text{g/g}$ CO₂ + 1000 $\mu\text{g/g}$ S at variable ΔFMQ_0 : (a) and (b) show the effects of variable H₂O at $\Delta FMQ_0=0$ for open- and closed-system degassing (where b is an enlargement of the low- P area shown by the dotted box in a), whilst (c) shows C/S ratios of degassing melt with has 3 wt.% H₂O + 1000 $\mu\text{g/g}$ CO₂ + 1000 $\mu\text{g/g}$ S and $0 \leq \Delta FMQ_0 \leq +2.5$ for closed-system degassing. Curve colours and styles as in Figure 1 for a and b, and Figure 3 for c, with the addition of the vertical grey dashed line in (a) and (c) for C/S = 20.

Alt text: Graphs showing decreasing carbon to sulfur ratio in the vapor with decreasing pressure for different initial water concentrations in the melt for open- and closed-system degassing in panel A and B, whereas the initial oxygen fugacity varies at 3 weight percent water in panel C. Panel B is a zoomed in version of panel A to highlight the behaviour at low pressures.

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