

On the oxidation state of arc magmas

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Supplementary Information

The Supplementary Information includes:

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Supplementary Text

a) Detailed sample description and processing

The sample from Yasur (TAN 6), is a scoria lapilli described in Métrich *et al.* (2011) and attributed to a sub-Plinian eruption dated to 1400–800 yr BP (Nairn *et al.*, 1988). The phase assemblage consists of a few per cent of plagioclase, clinopyroxene and olivine phenocrysts and Fe-Ti oxide microlites in a highly vesicular glassy matrix (Métrich *et al.*, 2011). The sample from Aoba (also called Ambae) consists of lapilli from the 2017-2018 plinian eruption (Moussallam *et al.*, 2019). The phase assemblage consists of rare phenocrysts of plagioclase, olivine, and clinopyroxene, and microlites of the same phases and spinel in a glassy matrix (Moussallam *et al.*, 2019). The sample from Ambrym is described in Moussallam *et al.* (2021) and consists of lapilli from an intra-caldera, lava-fountain eruption. The phase assemblage includes olivine, plagioclase and clinopyroxene phenocrysts and glomerocrysts, sometimes associated with magnetite in a glassy vesicular matrix (Moussallam *et al.*, 2021). The sample from Shinmoedake is a lapilli-sized tephra for the 2011 eruption.

It comes from a mixed andesitic pumice from the Shinmoedake cone of the Kirishima volcanic group. The sampled layer corresponds to Unit 2 of (Miyabuchi *et al.*, 2013). The bulk composition is andesitic, containing phenocrysts of plagioclase, clinopyroxene, orthopyroxene, olivine, magnetite and ilmenite (Suzuki *et al.*, 2013). The Aso sample is a coarse black scoria fall deposit from the Kishimadake cone, that erupted 3.7 ka ago (Miyabuchi and Watanabe, 1997; Hirata *et al.*, 2020) and is further described in Kawaguchi *et al.* (2021). It is identified as the mafic end member of all Holocene Aso eruptions. The sample from Okmok is described in Peccia *et al.* (2023) and consists of scoria from ash fall and flow deposits from the 43 BCE Okmok II caldera-forming eruption. The phase assemblage includes plagioclase, clinopyroxene, orthopyroxene, titanomagnetite, and, in the flow deposit, rare olivine (Peccia *et al.*, 2023). The San Cristóbal sample (SC11D) was collected from the avalanche block layer located 600 m south of the San Cristóbal crater rim, belongs to a sequence of Holocene eruptions consisting of unconsolidated layers of homogeneous coarse scoriae and bombs of basaltic andesite composition covering a thick avalanche block layer (Robidoux *et al.*, 2017). The deposit was made of medium-to-coarse unconsolidated lapilli-sized scoriae. It contains ~50vol% phenocrysts of plagioclase, olivine, rare clinopyroxenes and highly vesiculated scoria from a XVIII century sub-plinian eruption (BA deposit: Havlicek *et al.*, 2000; Hazlett, 1977). The phase assemblage consists of phenocrysts of plagioclase, clinopyroxene, orthopyroxene and Fe-Ti oxides included into vesicular fragments. The sample from Fuego (VF18-AC4, provided by Terry Plank) is from the Fuego 2018 eruption and was collected from a rooftop 14.5 km away from the volcanic crater during the eruption (Shi *et al.*, 2021). It is a tephra fall deposit consisting of ash, lapilli and bombs with olivine, clinopyroxene and plagioclase in a trachy-andesite matrix (Taracsák *et al.*, 2023). The sample from Cotopaxi is described in Pistolesi *et al.* (2021) and originates from the upper part of the Layer 2, emitted during the 1744 eruption. The bulk composition is slightly more evolved than andesitic. Tephra matrix is mostly glassy with less than 5% microlites and 20% of phenocrysts, primarily plagioclase (90%), pyroxenes (8%), and olivine (<2%). The sample from Villarrica is described in Moussallam *et al.* (2023) and consists of agglutinate scoria, from the March 2015 lava fountain eruption. The phase assemblage consists of 68% matrix glass (highly vesiculated), 28% plagioclase, 2% olivine, 1%

clinopyroxene, and 0.2% chromian spinel (Moussallam *et al.*, 2023). Sample from Stromboli originate from several different eruptions major and paroxysmal. In chronological order, the 5 May 2003 paroxysmal eruption (ST345), the 24 of November 2009 major eruption, and both July and August 2019 paroxysmal eruptions (STG and SHE750, respectively). Each tephra shows mingling of two magma batches, a dark microlite-rich “high porphyric” (HP) black scoria from the shallower magma chamber and a light glassy “low porphyric” (LP) golden pumice from the deeper magma chamber. HP patches contain euhedral plagioclases, pyroxenes and olivines and LP patches are less crystalline and with smaller crystals but with more primitive olivines.

Crystal-hosted melt inclusions were found by lightly crushing, sieving, magnetically separating and hand-picking under a binocular microscope in the 0.5-2 mm size and 0.25-0.5 mm size fractions. Crystals were doubly polished using a corundum mat (to avoid carbon contamination) to expose the melt inclusion on two parallel sides. Melt inclusions from Yasur, Shinmoedake, Aso, Fuego, San Cristóbal, Cotopaxi, Villarrica and Stromboli were newly prepared for this study. Melt inclusions from Aoba/Ambae included both new preparations and existing samples from Moussallam *et al.* (2019). Melt inclusions from Ambrym were existing samples from Moussallam *et al.* (2021). Melt inclusions from Okmok were existing samples from Peccia *et al.* (2023). When using existing sets of melt inclusions, host crystals were removed from their existing mount and polished on the reverse side, parallel to the first polish, to obtain double-polished wafers, except for the Okmok samples where melt inclusions (large enough to be analysed by XANES without host contamination) were re-polished but only single-exposed. The newly polished side was the one used for XANES analyses. Melt inclusions were analysed first by FTIR, then by XANES and finally by EPMA. Photomicrographs of all MI are provided in Figure S-1.

b) Fourier Transform Infrared Spectroscopy (FTIR)

Volatile (H₂O, CO₂) contents in MIs and matrix glasses were determined using an N₂ purged Thermo Nicolet iN10mx FTIR at Lamont-Doherty Earth Observatory (LDEO). Depending on the sizes of the MIs and glasses, various aperture sizes (>25x25µm) were applied to maximize the FTIR signal without host crystal

contamination. For each measurement, 256 high resolution ($2\text{--}4\text{cm}^{-1}$) scans were applied from 400cm^{-1} to 8000cm^{-1} to include both near- and mid IR peaks for water. The background was measured prior to each transect and subsequently subtracted from each spectrum. Thickness of the doubly polished wafers were measured using a digital micrometer (Mitutoyo Digimatic Indicator). Baseline fitting and H_2O and CO_2 concentrations were determined using PyIRoGlass (<https://github.com/sarahshi/PyIRoGlass>; Shi *et al.*, 2024).

c) Electron probe microanalyses (EPMA)

The major element compositions of matrix glasses, MIs and their host crystals were measured with a Cameca SxFiveTactis electron microprobe at the Laboratoire Magmas et Volcans in Clermont-Ferrand during a single session (except for Okmok for which we report previous published EMPA from Peccia *et al.*, 2023). For crystalline phases (olivine and plagioclase), beam conditions of 15kV and 15 nA were employed ($2\mu\text{m}$ beam size), with variable counting time ranging from 10 to 30s on peak as a function of element concentration to reduce detection limit (with off peak counting times similar to on peak ones). Errors in wt.% (one standard deviations based on 15 repeats on San Carlos olivine) are ± 0.19 for SiO_2 , ± 0.01 for Al_2O_3 , ± 0.09 for FeO , ± 0.04 for MnO , ± 0.01 for CaO , ± 0.12 for MgO , and ± 0.02 for NiO . For glass analyses, the beam current was decreased to 8 nA and the beam defocused to $10\mu\text{m}$. Sodium was analysed first to limit the effects of Na loss. Counting times of 10 s minimum were used (with off peak counting times similar to on peak ones). Error in wt.% (one standard deviations based on 23 repeats on standard glass VG-A99) are ± 0.18 for SiO_2 , ± 0.05 for Na_2O , ± 0.04 for K_2O , ± 0.19 for Al_2O_3 , ± 0.10 for CaO , ± 0.16 for FeO , ± 0.09 for MgO , ± 0.06 for TiO_2 , ± 0.04 for MnO , and ± 0.04 for P_2O_5 . The instrument was calibrated on natural and synthetic mineral standards and glasses: wollastonite (Si and Ca in glasses and plagioclase), San Carlos Olivine (Si in olivine), MnTiO_3 (Ti, Mn), orthoclase (K and Al in minerals), standard glass VG2 (Al in glasses), fayalite (Fe), forsterite (Mg), NiO (Ni), albite (Na) and apatite (P). Beam conditions for volatile measurements were 15kV, 40 nA and a $10\mu\text{m}$ defocused beam with a total counting time of 60s for Cl and 120s for S (sum of two spectrometers); S peak positions were carefully checked prior to analyses. Error (one standard deviations based on 10 repeats

on standard glasses Alvine and KE12) were 30 ppm and 48 ppm for S (alvine) and Cl (KE12) respectively. Standard repeat analyses are given in the accompanying supplementary EPMA standards spreadsheet (.xlsx), available for download from the online version of this article at <https://doi.org/10.7185/geochemlet.2611>.

d) Validity of the fractional crystallisation correction approach

While the calculated oxidation state of melt inclusions at MgO = 6 wt.% is model-dependent—and the use of Petrolog3, which assumes anhydrous conditions at 1 atm (conditions far removed from natural systems, especially for water-rich arc magmas), can be criticized—the emerging trend is robust and model-independent. This can be demonstrated using a fundamentally different approach.

We compiled all existing literature data for the Mariana arc (Kelley and Cottrell, 2012; Brounce *et al.*, 2014, 2016) and focused on melt inclusions with sulfur contents between 1350 and 1650 ppm. By narrowing the sulfur content range, we minimized the influence of sulfur degassing and targeted the least degassed portion of the dataset. Plotting sulfur content against oxidation state (Fig. S7A) reveals an empirical linear relationship ($R^2 = 0.6$), which we used to model the effects of fractional crystallization in our dataset.

The results of this empirical approach align closely with those of the original model (Fig. S7B). Using the empirical method, the same trend persists: melt inclusions with higher sulfur contents are consistently more oxidized than those with lower sulfur contents (Fig. S8). Quantitatively, melt inclusions with $S > 2000$ ppm exhibit an average oxidation state of QFM +1.0 (± 0.2 ; 1σ), while those with $S < 100$ ppm average QFM +0.2 (± 0.5 ; 1σ).

The agreement between two entirely independent modeling approaches—each with distinct assumptions and methodologies—lends strong confidence to the validity of our findings.

e) Sulfur degassing and oxidation state in arc magmas

Three gas–melt equilibrium, closed-system, degassing calculations were tested against the observed data trend. The first model is a modified version of the gas–melt equilibrium model by Gaillard and Scaillet (2009)

using the CO₂-H₂O solubility model of Iacono-Marziano *et al.* (2012). The second model is the D-Compress model by Burgisser *et al.* (2015). The third model is the Sulfur_X model by Ding *et al.* (2023).

In all three cases, two runs were performed. The first run was performed using the average composition of all MIs in our dataset at MgO = 6 wt.% (53.59±2.23 wt.% SiO₂; 0.89±0.29 wt.% TiO₂; 15.61±1.16 wt.% Al₂O₃; 9.12±1.77 wt.% FeO_T; 0.14±0.05 wt.% MnO; 6.00 wt.% MgO; 10.39±1.00 wt.% CaO; 2.70±0.36 wt.% Na₂O; 1.61±0.55 wt.% K₂O; 0.19±0.09 wt.% P₂O₅; errors given as 1 standard deviation) and the highest volatile contents measured in the dataset (2500 ppm S; 2000 ppm CO₂; 5 wt.% H₂O) as starting conditions (with no excess fluid). Temperature was set at 1150°C (as determined from Petrolog3 calculations), initial oxidation state was one log unit above the QFM buffer (calculated at 1 bar) corresponding to a Fe³⁺/ΣFe of 0.24. We note that the absence of correlation between S and FeO content of the magmas points to no role of sulfide saturation in influencing the sulfur content of the melts (Fig. S-3). The second run was performed using the average composition of all MIs with 6 <MgO< 7 wt.% in the Muth and Wallace 2021 dataset of melt inclusion from scoria cones in the Lassen area (California, USA): 49.08±0.93 wt.% SiO₂; 1.07±0.22 wt.% TiO₂; 18.34±1.36 wt.% Al₂O₃; 7.92±1.52 wt.% FeO_T; 0.14±0.03 wt.% MnO; 5.83±0.47 wt.% MgO; 9.88±0.90 wt.% CaO; 3.53±0.28 wt.% Na₂O; 0.70±0.18 wt.% K₂O; 0.23±0.08 wt.% P₂O₅ and the highest volatile contents measured in that dataset: 2493 ppm S; 3.02 wt.% H₂O as starting conditions. As no CO₂ measurement is reported, the D-Compress run assumes 5757 ppm CO₂, and the Sulfur_X run assumes 5775 ppm CO₂. The starting pressure was set to 560 MPa, with no excess fluid, a temperature of 1150°C and the initial oxidation state was 1.5 log unit above the QFM buffer (calculated at 1 bar), as measured by Muth and Wallace (2021) for the most sulfur-rich and most oxidised melt inclusion in that dataset. All model outputs are reported in the additional supplementary model outputs spreadsheet (.xlsx), available for download from the online version of this article at <https://doi.org/10.7185/geochemlet.2611>.

In Run 1, all three models predict some degree of melt reduction associated with sulfur degassing (Fig. S-9). The Sulfur_X model aligns well with the most oxidized MIs in our dataset, particularly matching the trend defined by the Aoba/Ambae MIs. The D-Compress model corresponds well with some of the most

reduced MIs in our dataset but unfortunately concludes its calculations with predicted sulfur content remaining in the melt that are too high (600 ppm). The Gaillard and Scaillet (2009) model best reproduce the full magnitude of the observed change in oxidation state with sulfur content and matches best the trend defined by the Stromboli MIs. The observation that most of the change in oxidation state occurs during the last 500 ppm of sulfur degassing is well reproduced by the Gaillard and Scaillet (2009) and Sulfur_X models. Our modelling approach is intentionally limited to a first-order analysis, as additional magmatic processes, such as crustal assimilation and magma mixing, may have variably influenced oxidation states across different localities.

In Run 2 (Fig. S-9), the Gaillard and Scaillet (2009) model still predicts a reduction of the melt associated with sulfur degassing, reproducing, to a first order, the trend observed in the Lassen data and in the entire dataset. The D-Compress model also still predicts a reduction of the melt associated with sulfur degassing, with a trend aligned with the melt inclusions datasets but still concludes its calculations (at 1 bar) with a remaining sulfur content in the melt that is too high (790 ppm). The Sulfur_X model by contrast predicts that under these starting conditions sulfur degassing should be associated with oxidation of the melt, which is not consistent with the melt inclusions datasets.

Figure 3 in the main text shows that the oxidation state of arc magmas remains unchanged in the 2500 to 500 ppm S range, with most of the recorded change in oxidation state occurring between 500 and 0 ppm S. This is distinct from the trend observed in hotspot volcanoes (Fig. 3D). Two processes might, in part, explain this observation.

First, at greater pressure, H₂S is favored in the gas phase over SO₂, as a result S degassing from the melt can be redox-neutral as S²⁻_(melt) goes to S²⁻_(gas) (in the form of H₂S). At lower pressure however, SO₂ is strongly favored over H₂S in the gas phase, S²⁻_(melt) goes to S⁴⁺_(gas) (in the form of SO₂) liberating six electrons that can reduce Fe in the melt (Gaillard *et al.*, 2011). Degassing at shallower pressure (i.e., once less S remains in the melt) should hence be associated with a stronger reduction of the melt with S degassing.

It is worth noting that if sulfur was initially predominantly dissolved as S⁶⁺ in the melt, degassing as either SO₂ or H₂S in the gas phase would be expected to oxidize the remaining melt (by taking two and eight

electrons from the melt respectively, thus oxidizing Fe in the melt), a process which we do not observe in the current dataset.

The second process is the degassing of water. While most of the water dissolved in the melt will degas as H₂O in the gas phase — a redox-neutral reaction where H₂O_(Melt) converts to H₂O_(gas) — some of the water will produce H₂ in the gas phase via a reaction that could be written as H₂O_(Melt) + 2FeO_(Melt) = H_{2(gas)} + Fe₂O_{3(melt)}. Thus, the degassing of water should be associated with an oxidation of the melt (e.g., Candela, 1986; Burgisser and Scaillet, 2007; Humphreys *et al.*, 2015). In arc magmas, which are more water-rich than their hotspot counterparts, this effect is likely significant, counteracting to a large extent the reduction due to sulfur degassing.

Supplementary Tables

Table S-1 Acquisition parameters used during Fe K-edge XANES analysis.

Energy range (eV)	Step size (eV)	Dwell time (s)
For moderate attenuation (0.1 mm Al plate)		
Conditions:		
7020.0–7100.0	5	1
7101.0–7104.0	1	1
7104.1–7109.9	0.5	5
7110.0–7117.9	0.1	5
7118.0–7119.4	0.1	1
7119.5–7127.0	0.5	1
7128.0–7144.0	1	1
7148.0–7408.0	5	1
7410.0–7500.0	10	1
For high attenuation (0.25 mm Al plate)		
Conditions:		
7000.0–7100.0	10	2
7101.0–7104.0	1	2
7104.1–7109.9	0.5	15
7110.0–7117.9	0.1	15
7118.0–7119.4	0.1	3
7119.5–7127.0	0.5	2
7128.0–7144.0	1	2
7148.0–7408.0	5	2
7410.0–7500.0	10	2

Table S-2 $\text{Fe}^{3+}/\Sigma\text{Fe}$ (1: calculated using Zhang *et al.*, 2018 Mössbauer value and 2: calculated using Berry *et al.*, 2018 Mössbauer value), ΔQFM (1: calculated using the $\text{Fe}^{3+}/\Sigma\text{Fe}$ 1 ratio and the equation of Kress and Carmichael, 1991 and 2: calculated using the $\text{Fe}^{3+}/\Sigma\text{Fe}$ 2 ratio and the equation of Kress and Carmichael, 1991) for all melt inclusions analysed together with volatile contents.

Table S-3 Major element compositions and melt inclusion and bubble dimensions. Error in wt.% on melt inclusions major elements (one standard deviations based on 23 repeats on A99 standard glass) are ± 0.18 for SiO_2 , ± 0.05 for Na_2O , ± 0.04 for K_2O , ± 0.19 for Al_2O_3 , ± 0.10 for CaO , ± 0.16 for FeO , ± 0.09 for MgO , ± 0.06 for TiO_2 , ± 0.04 for MnO , and ± 0.04 for P_2O_5 . Data for Okmok are from Peccia *et al.* (2023).

Table S-4 Mineral host major element compositions. Error in wt.% on host mineral phase major elements (one standard deviations based on 15 repeats on San Carlos olivine) are ± 0.19 for SiO_2 , ± 0.01 for Al_2O_3 , ± 0.09 for FeO , ± 0.04 for MnO , ± 0.01 for CaO , ± 0.12 for MgO , and ± 0.02 for NiO . Data for Okmok are from Peccia *et al.* (2023).

Table S-5 Average, standard deviation, maximum and minimum values and number of analyses for $\text{Fe}^{3+}/\Sigma\text{Fe}$, ΔQFM , volatile contents and major element compositions of all glasses analysed at each volcano. ¹ and ² are as described in Table S-2 caption.

Table S-6 $\text{Fe}^{3+}/\Sigma\text{Fe}$ (1: calculated using Zhang *et al.*, 2018 Mössbauer value and :2 calculated using Berry *et al.*, 2018 Mössbauer value), volatile contents and major element compositions of all glasses analysed at each volcano.

Table S-7 $\text{Fe}^{3+}/\Sigma\text{Fe}$, ΔQFM , volatile contents and major element compositions of Melt inclusions after reverse fractional crystallisation calculations to $\text{MgO}=6$ wt.%.

Tables S-2 to S-7 (.xlsx) are available for download from the online version of this article at <https://doi.org/10.7185/geochemlet.2611>.

Supplementary Figures

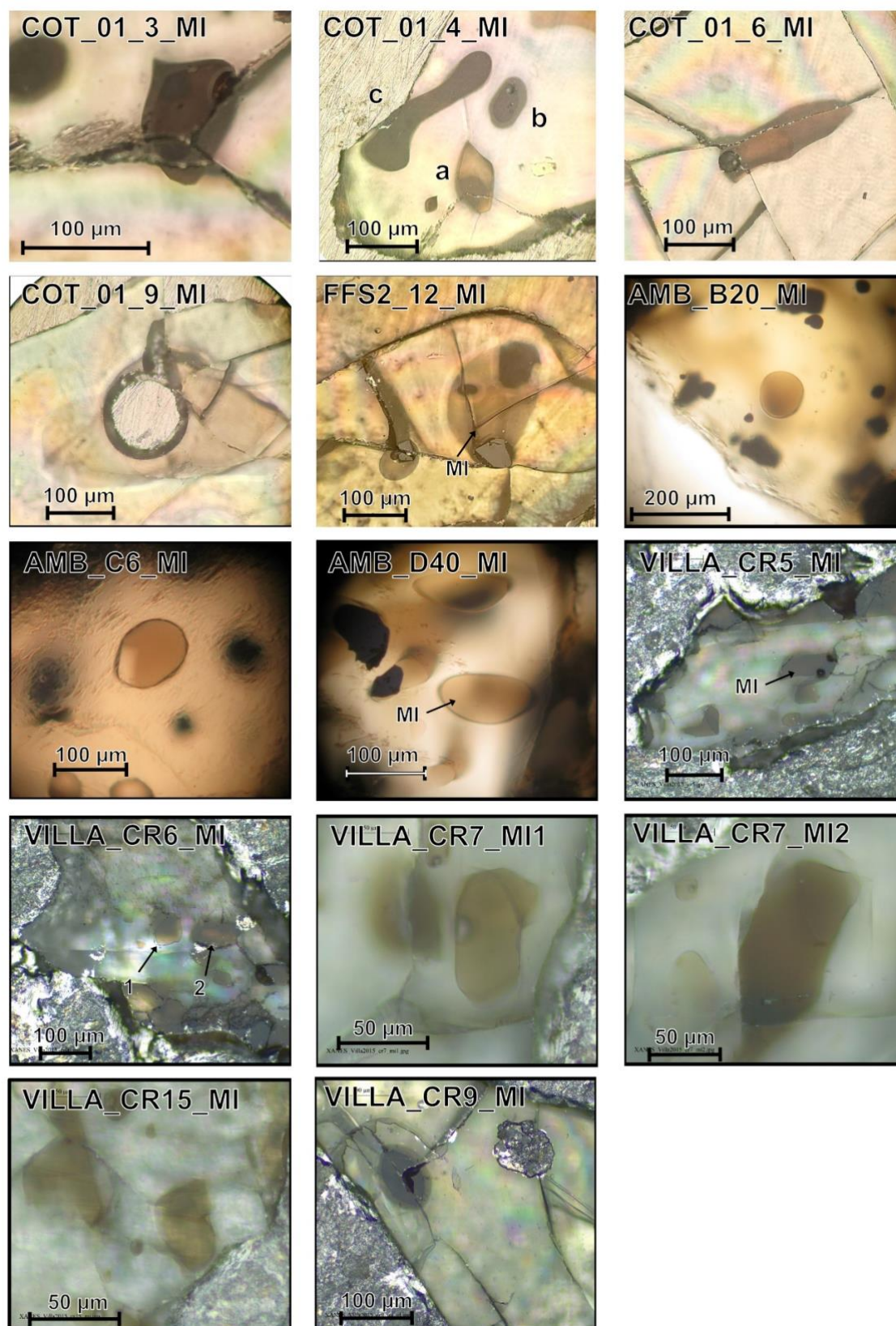


Figure S-1 Transmitted and reflected light microphotographs of all melt inclusions analysed.

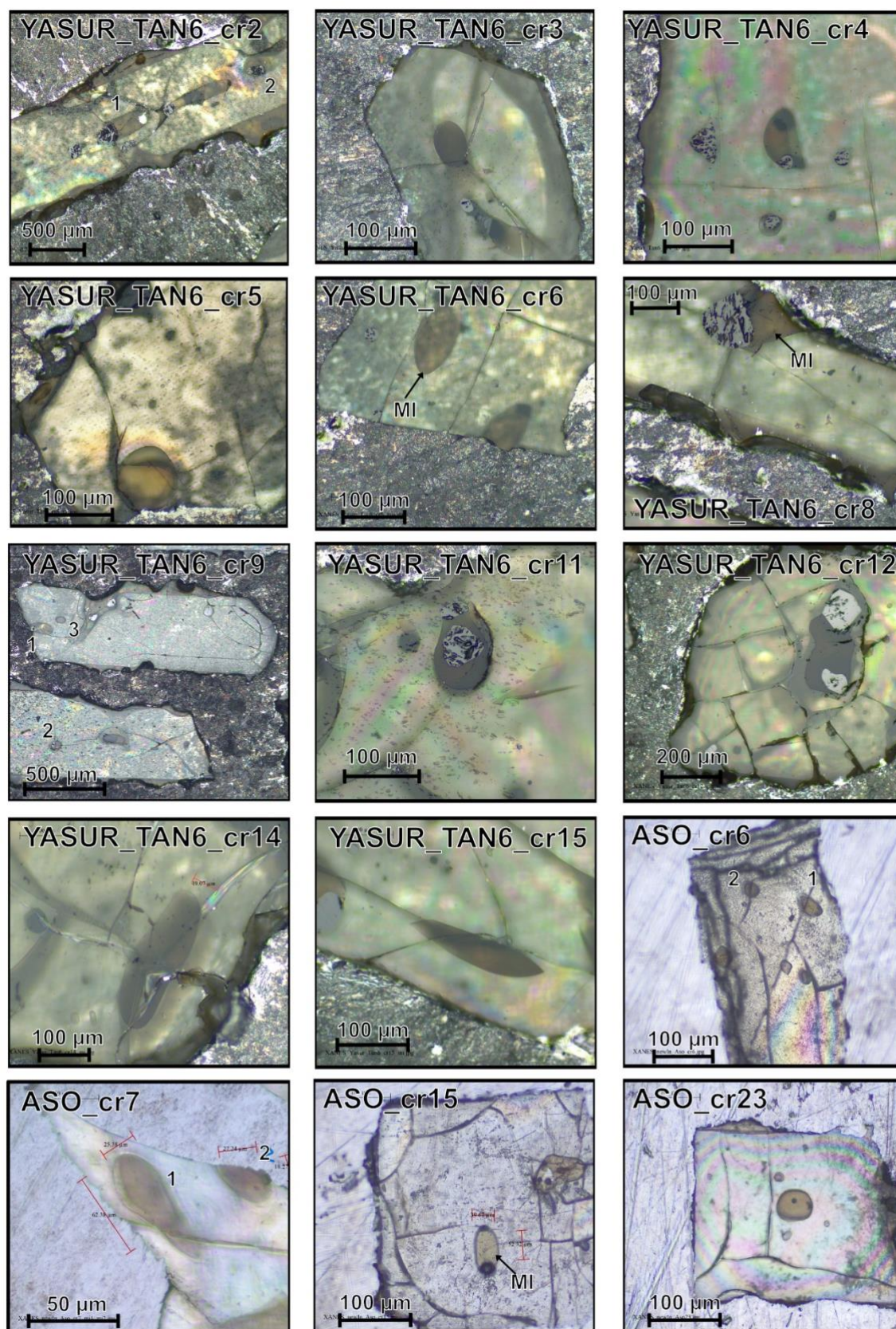


Figure S-1 continued.

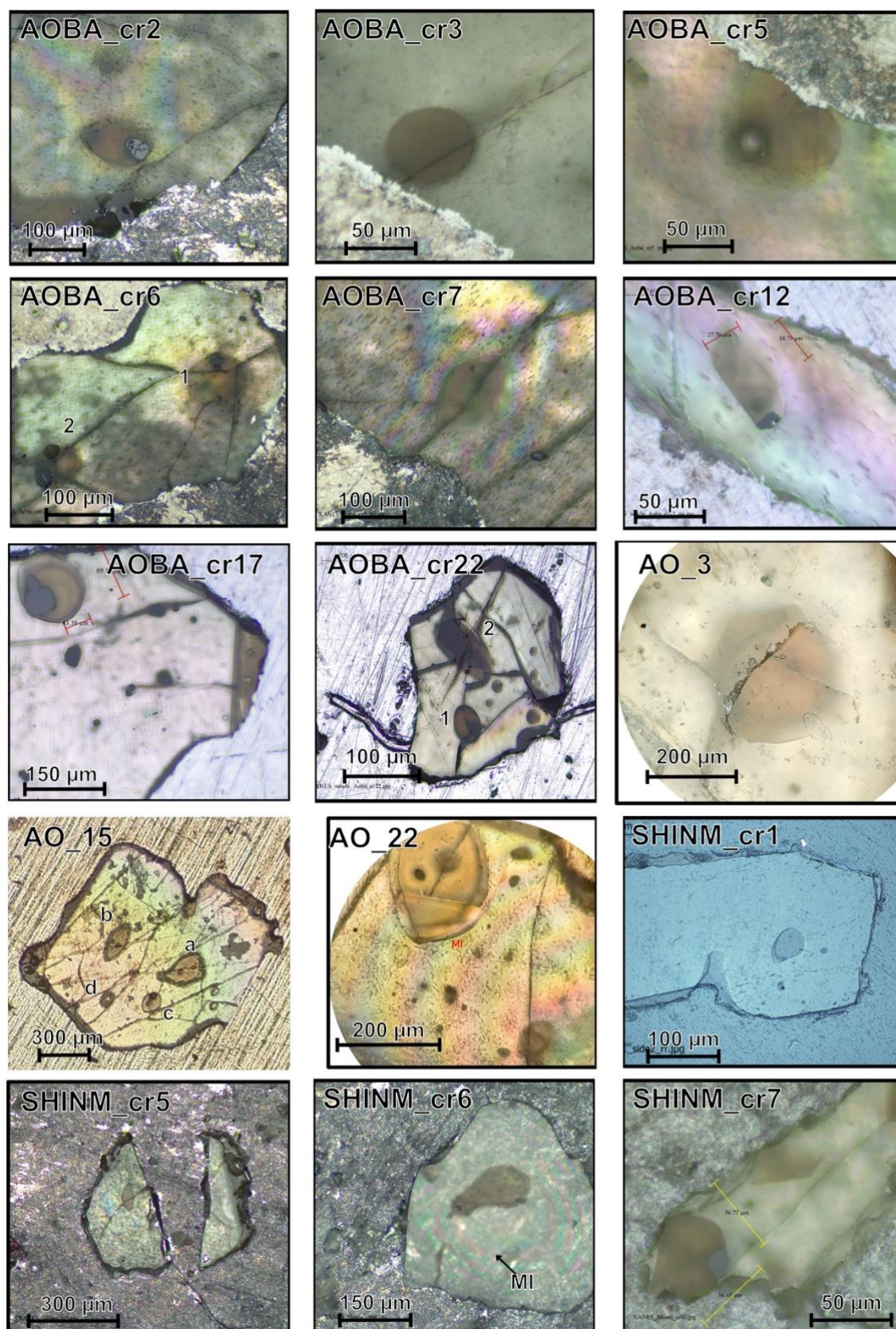


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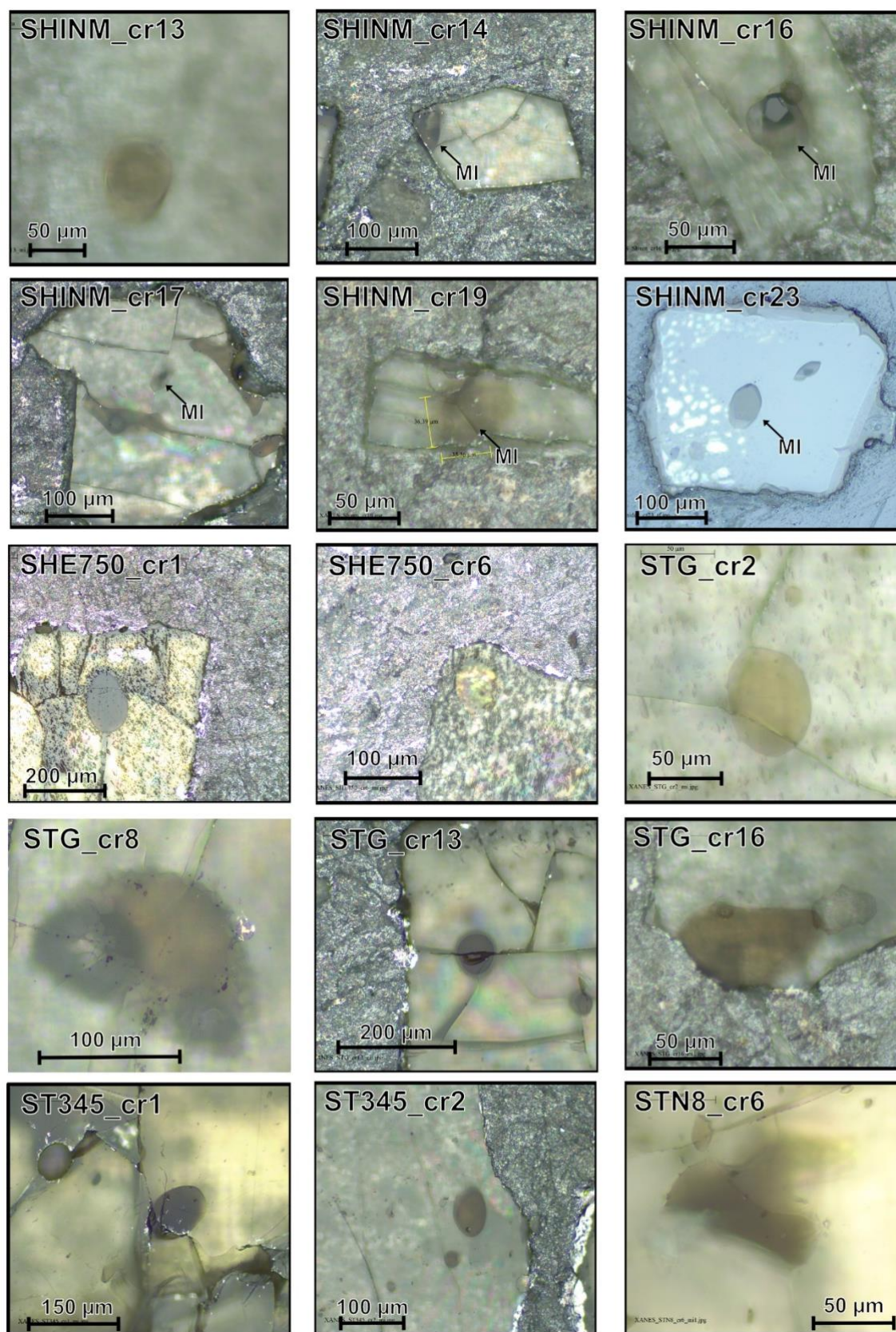


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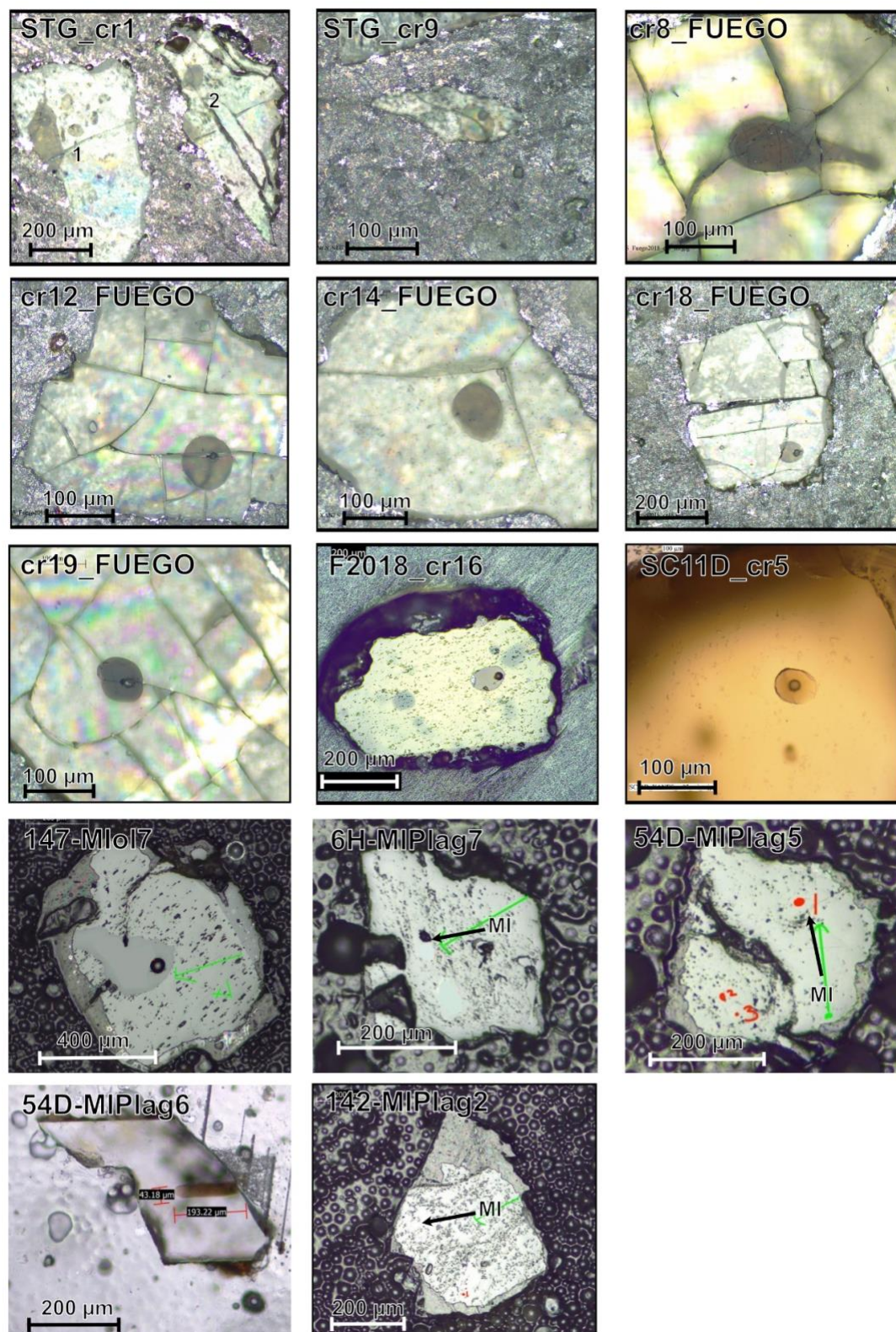


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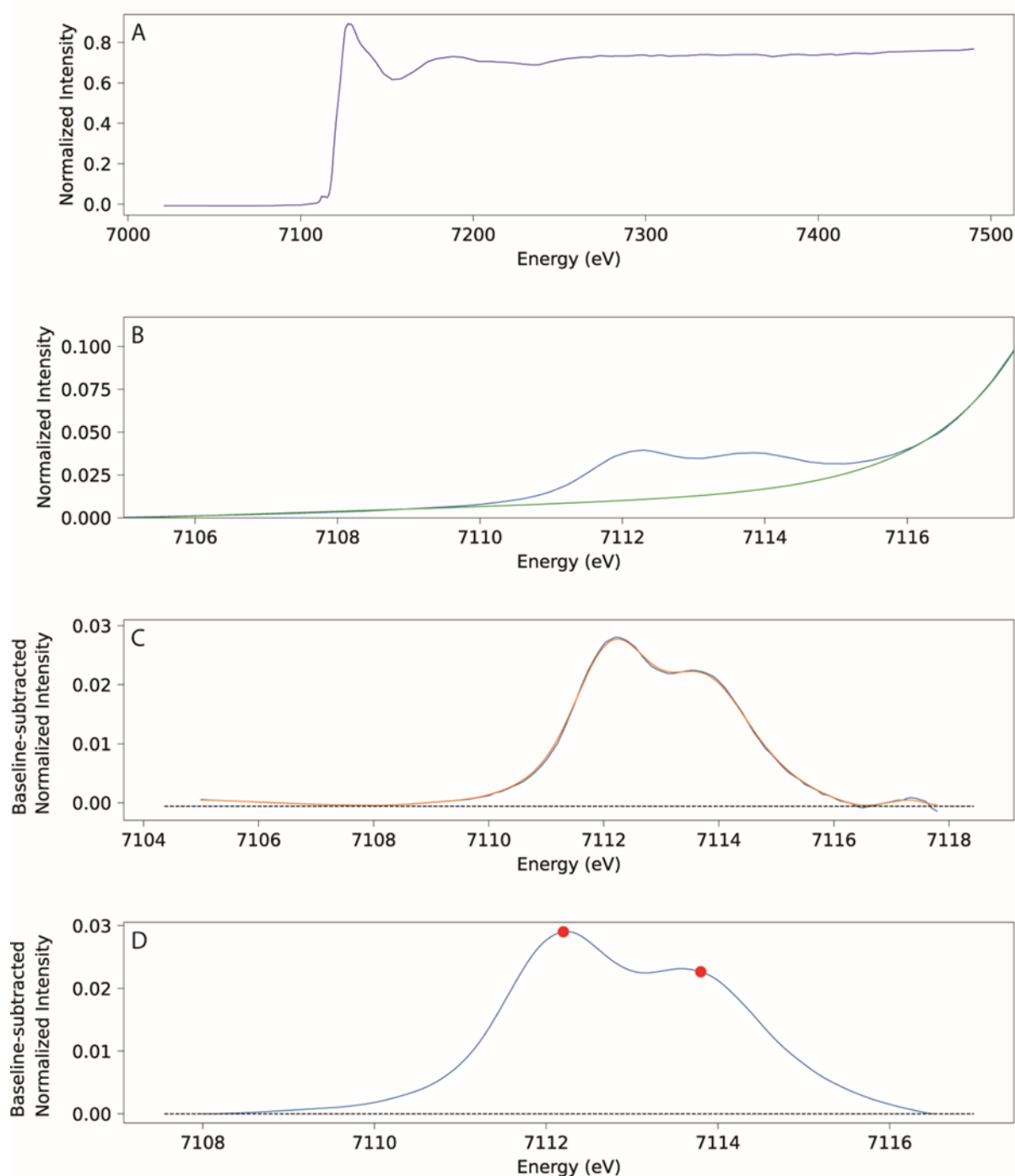


Figure S-2 Example of XANES spectra processing, on a spectrum obtained from a natural melt inclusion from Villarrica volcano. A. Edge-step normalized spectra. B. Pre-edge region fitting; green line shows the baseline absorption of the main Fe absorption edge (linear +DHO functions). C. Baseline-subtracted spectrum (blue), modelled as a combination of two gaussian functions (orange). D. Final pre-edge, the ratio of the baseline-subtracted normalized intensities at 7112.2 and 7113.8 eV (red dots) is extracted.

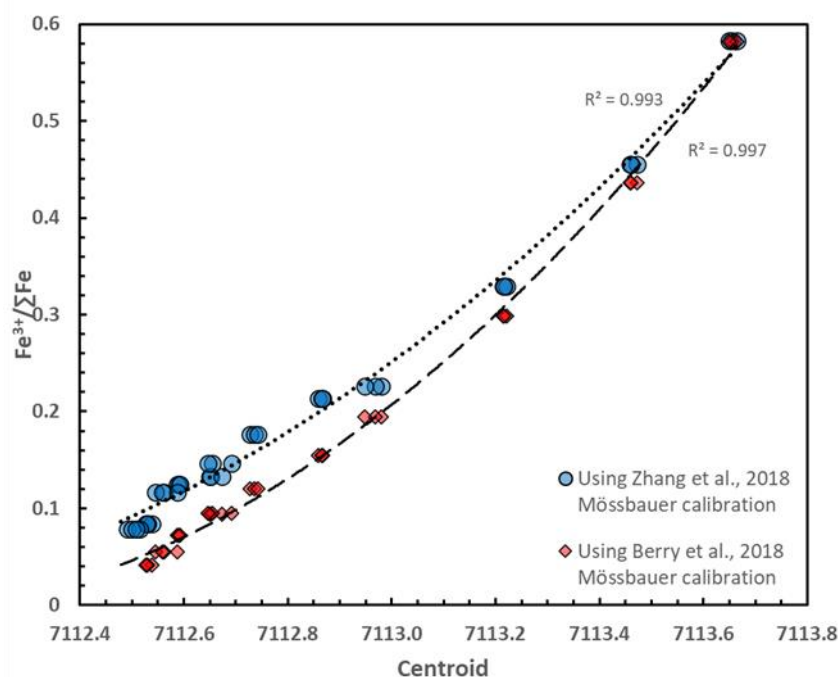


Figure S-3 Calibration lines based on the centroid energy position determined by XANES compared with $\text{Fe}^{3+}/\Sigma\text{Fe}$ ratios determined by Mössbauer spectroscopy in basaltic standard glasses from the Smithsonian NMNH (Mössbauer $\text{Fe}^{3+}/\Sigma\text{Fe}$ values from Zhang *et al.*, 2018 and Berry *et al.*, 2018 are shown). Note that the R^2 values on the 2nd degree polynomial fits are lower than when using the peak height ratio.

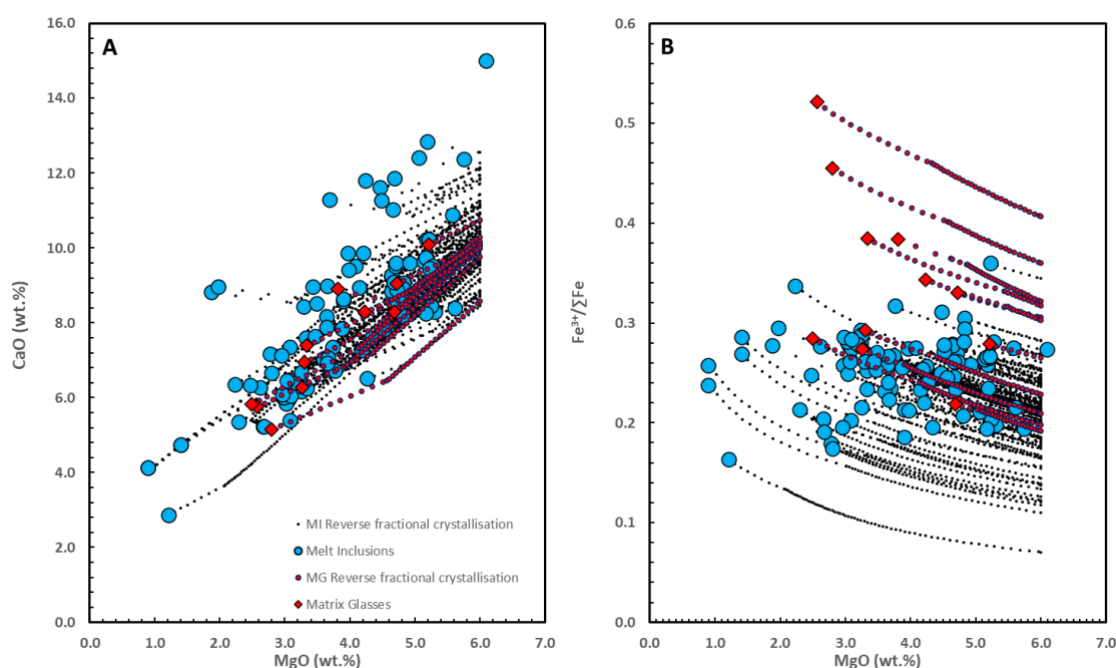


Figure S-4 The effect of pre-entrapment crystallization modelled on the composition (A) and oxidation state (B) of the melt inclusions and averages of matrix glasses. Reverse fractional crystallisation calculations were performed until the melt reaches a value of $\text{MgO}=6\text{wt.}\%$ using Petrolog3 (Danyushevsky and Plechov, 2011).

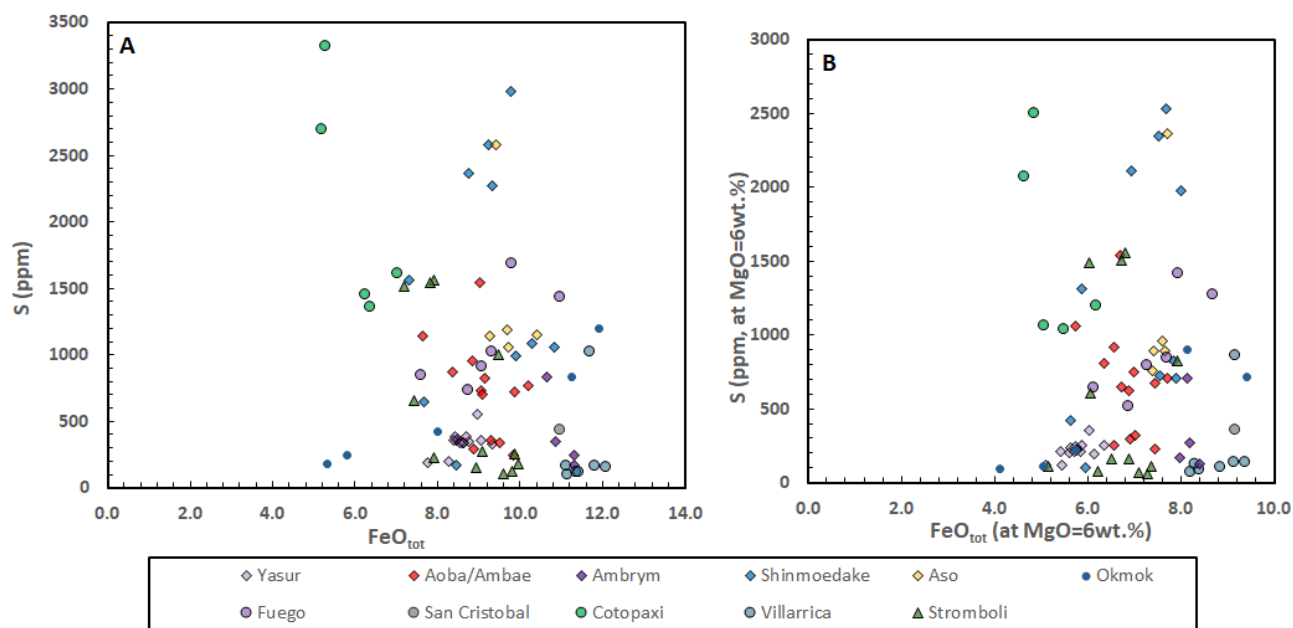


Figure S-5 Sulfur content versus FeO (total) in all melt inclusions. **A.** Raw data. **B.** Data after reverse fractional crystallization calculations to MgO=6wt.%.

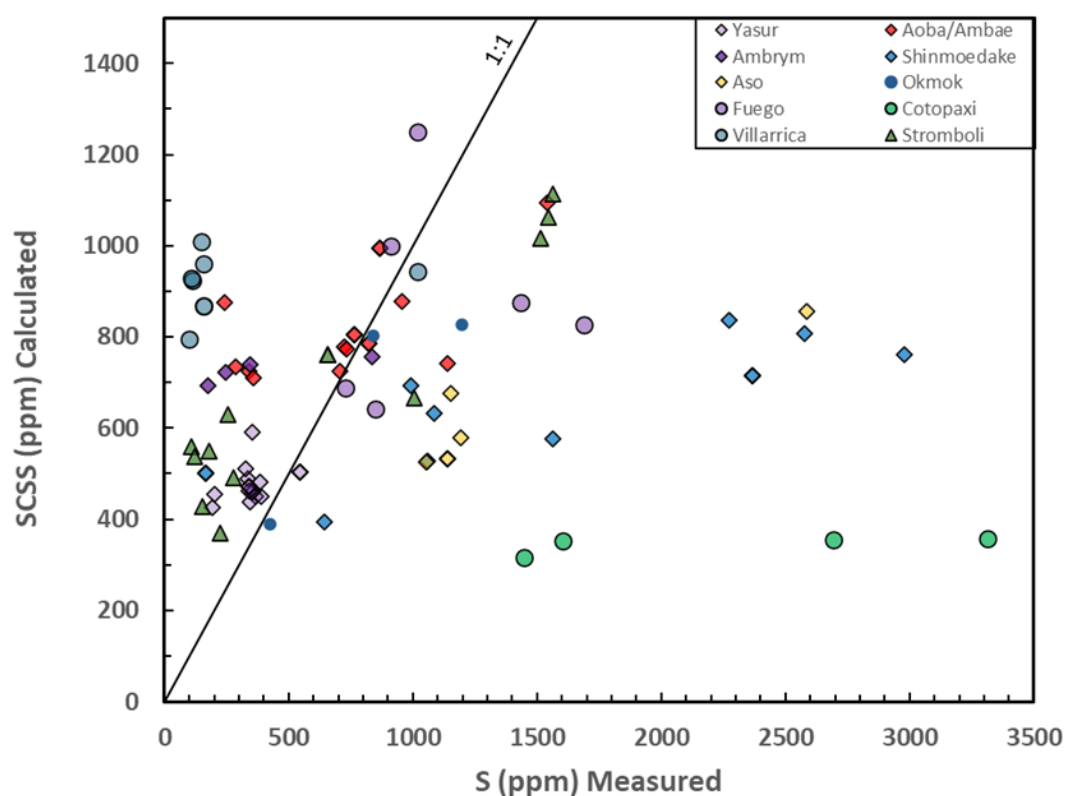


Figure S-6 Comparison between theoretical Sulfur content at sulfide saturation (calculated using the model of Fortin *et al.*, 2015) and measured sulfur content in melt inclusions. The absence of equivalence shows that the sulfur content of melt inclusions is not a reflection of equilibrium with sulfides.

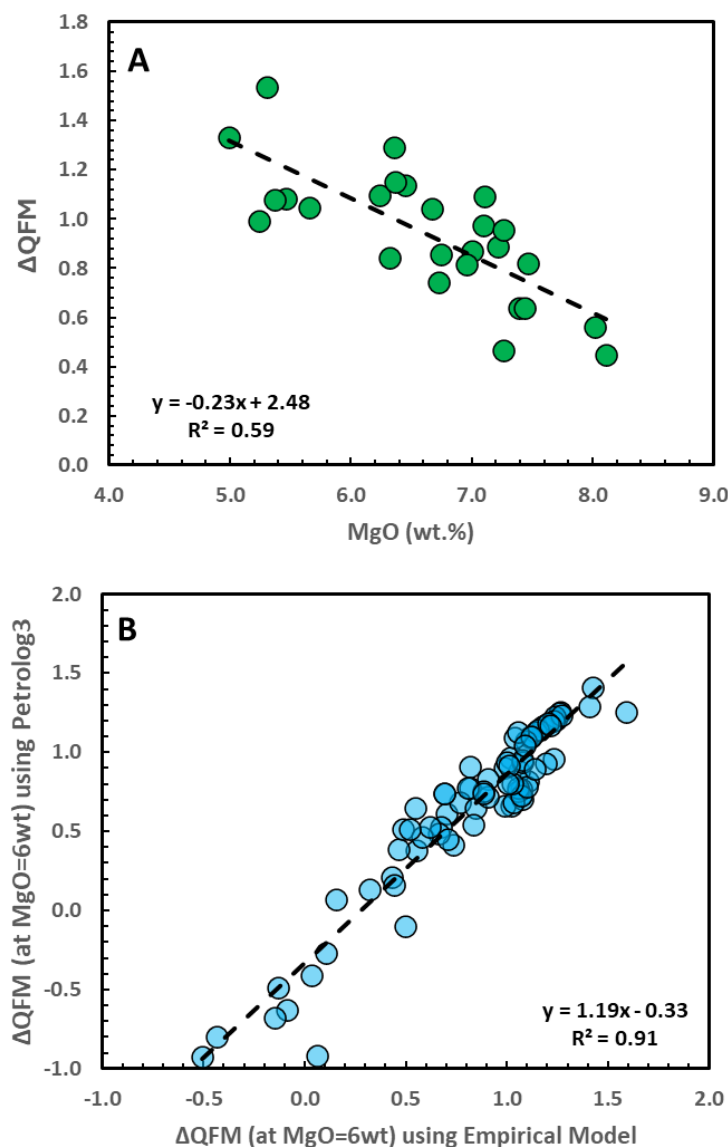


Figure S-7 A. Empirical relationship between MgO and oxidation state in melt inclusions and pillow glasses from the Mariannas over a narrow range in S content (1350 to 1650 ppm; original data from Kelley and Cottrell, 2012; Brounce *et al.*, 2014, 2016, as reported in Cottrell *et al.*, 2021). B. Comparison between oxidation state of melt inclusions (this study's dataset) after reversing fractional crystallization to a common value of MgO=6wt.% using either the empirical model approach or using Petrolog3.

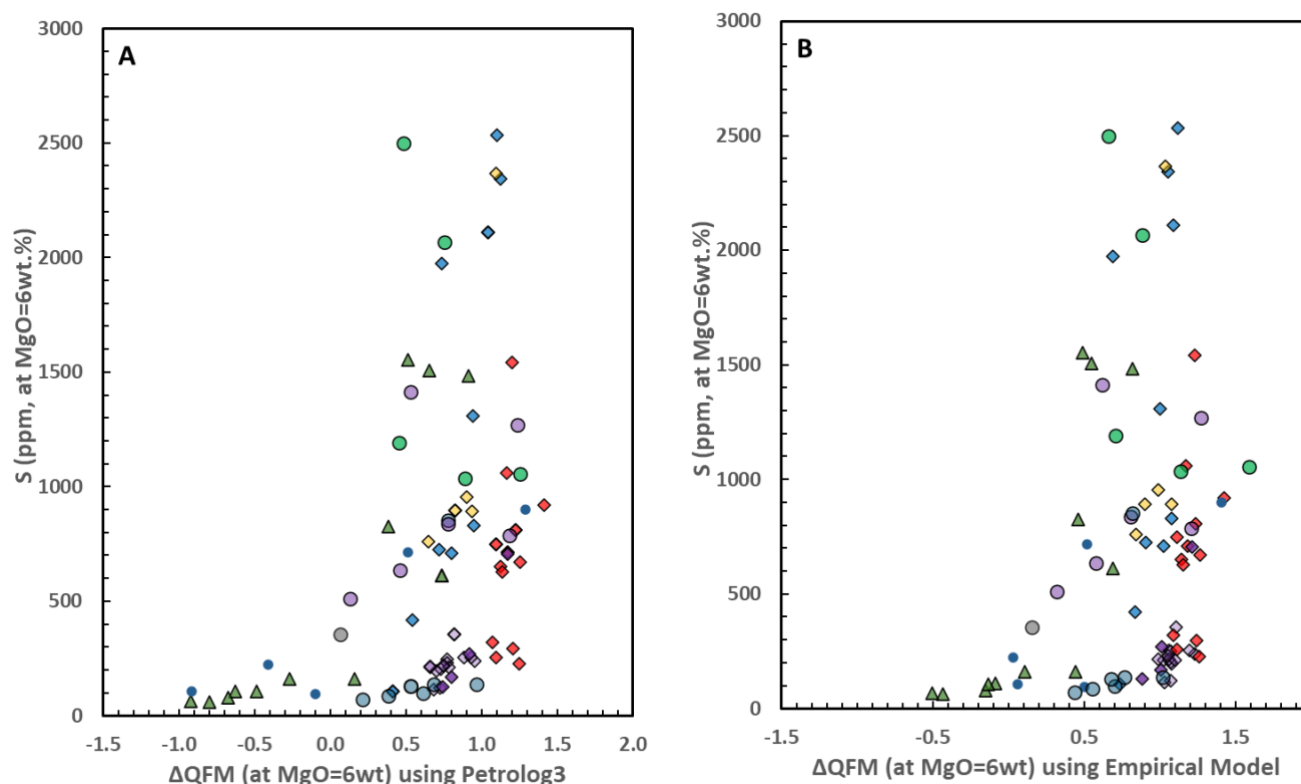


Figure S-8 Side by side comparison between the oxidation state of melt inclusions compared to their S content after reversing fractional crystallization to a common value of MgO=6wt.% using either Petrolog3 (A) or the empirical model approach (B). All symbols are the same as in Fig. 3 in the main text.

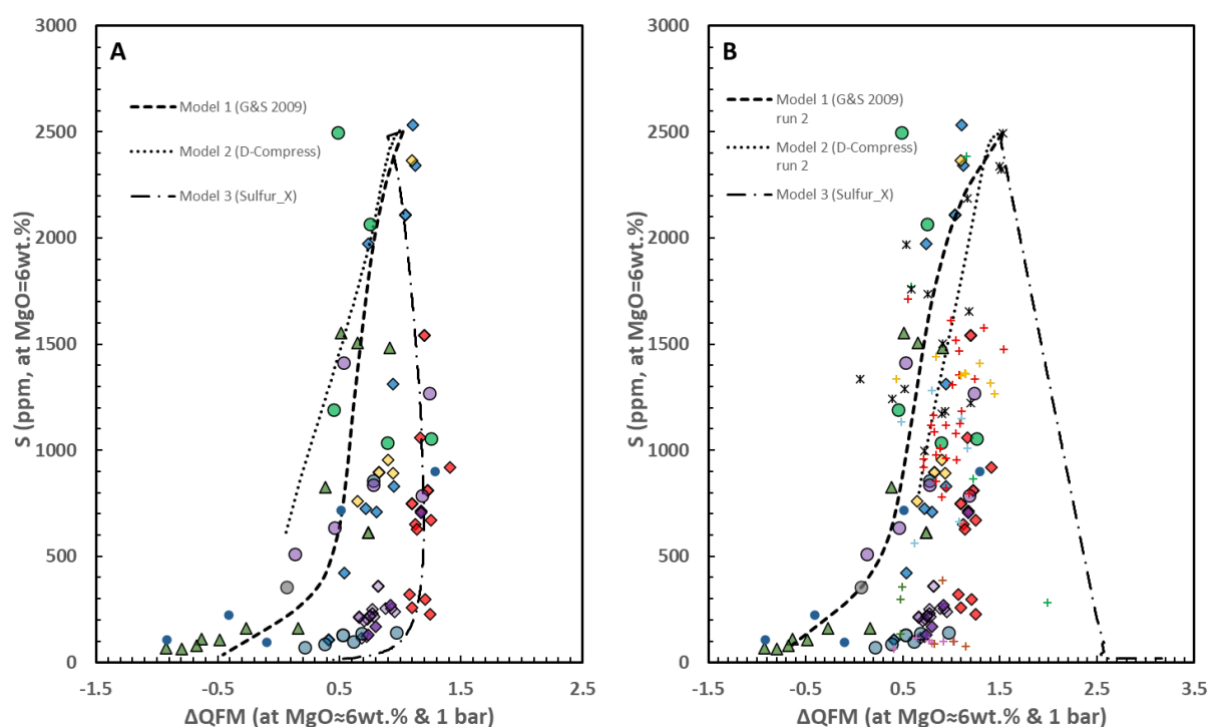


Figure S-9 **A.** Comparison between sulfur content and oxidation state of melt inclusions after reversing fractional crystallisation to a common value of MgO=6wt.% and the result of three forward modelling (closed-system, equilibrium) calculations of the effect of degassing for an average arc composition starting with 2500 ppm S; 2000 ppm CO₂; 5 wt.% H₂O and QFM+1. **B.** Same as A with the addition of melt inclusions (and pillow glasses) data from the literature, all with MgO content between 5 and 7 wt.%, and comparing with the result of three forward modelling (closed-system, equilibrium) calculations of the effect of degassing using the average composition of melt inclusions from Lassen cones, starting with 2500 ppm S; 3 wt.% H₂O at 560 MPa and QFM+1.5. All symbols are the same as in Fig. 3 in the main text.

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