

Appendix

Error Analysis

In order to appropriately determine the error incurred by our mass balance calculations, each source of error must be considered individually and compounded through each calculation. The sources of error in these mass-balance calculations are 1) measurement of the density of CO₂ in the bubble, 2) measurement of the concentration of CO₂ in the glass, 3) measurement of the dimensions of the MI and bubble, 4) assumed density of the glass, and 5) the assumed orientation of the MI as viewed within the host phenocryst.

To quantify the error associated with the Raman technique for measuring the density of CO₂ in the bubble, we conducted repeated measurements on a synthetic fluid inclusion bubble (Sterner & Bodnar, 1984) with a known density of CO₂ in the vapor phase (supplementary materials). We made one measurement during each analytical session when analyzing MI so that our estimation of error would include both errors associated with calibrating the spectrometer and errors associated with peak fitting. Over 21 measurements, the relative standard deviation of these measurements was 19% (1 σ). We used this value for the error for all of our measured bubble densities.

The error associated with the SIMS analysis of the MI glass ranges from 1 ppm to 46 ppm (<1% to 9%), and was determined based on calibration curves generated using glass standards at DTM and the standard deviation during 5 repeated collections during five repeated analyses.

Because the bubble and MI dimensions were measured in visible light, 0.5 μm is generally used as a minimum value for the precision of these measurements. The MI and bubble dimensions were clearly resolvable to within 1 μm under the 100x objective, so we use this value

(0.5 μm) for the estimation of MI diameters. With this amount of error, the relative uncertainty for the volume calculation for a sphere increases exponentially with decreasing volume: from about 5% error for a 30 μm bubble to 30% error for a 5 μm bubble.

We assume that the bulk density of the glass is 2.75 g/cm^3 . In order to determine the error that could be associated with this assumption, the relationship between glass bulk density and percent of total CO_2 in the bubble is shown for various concentrations of CO_2 in the glass in Figure S1. In general, our calculations become more sensitive to the bulk glass density as the contribution of the bubble to total CO_2 decreases and as the concentration of CO_2 in the glass increases. However, for the likely range in glass bulk density for mafic to intermediate melts (2.5 – 3.0 g/cm^3) and for the CO_2 concentrations that we have observed by SIMS analysis, the relative error associated with glass bulk density is unlikely to exceed about 10%.

When observing the MI in transmitted light, they appear to be oblate, and we can measure the long and short axis, but it is difficult to determine the depth of the MI, even by moving the stage. When calculating the volume of the MI, we assume that the depth of the MI is equal to the shorter diameter. To determine the range in error associated with the orientation of the MI, we assume that the third dimension of the MI (the depth when viewed through the microscope) could potentially be as long as the longest axis. If this were the case, the relative error would increase with increasing aspect ratio (long axis divided by short axis). For most of our inclusions, the source of error would not exceed about 10% to 50%

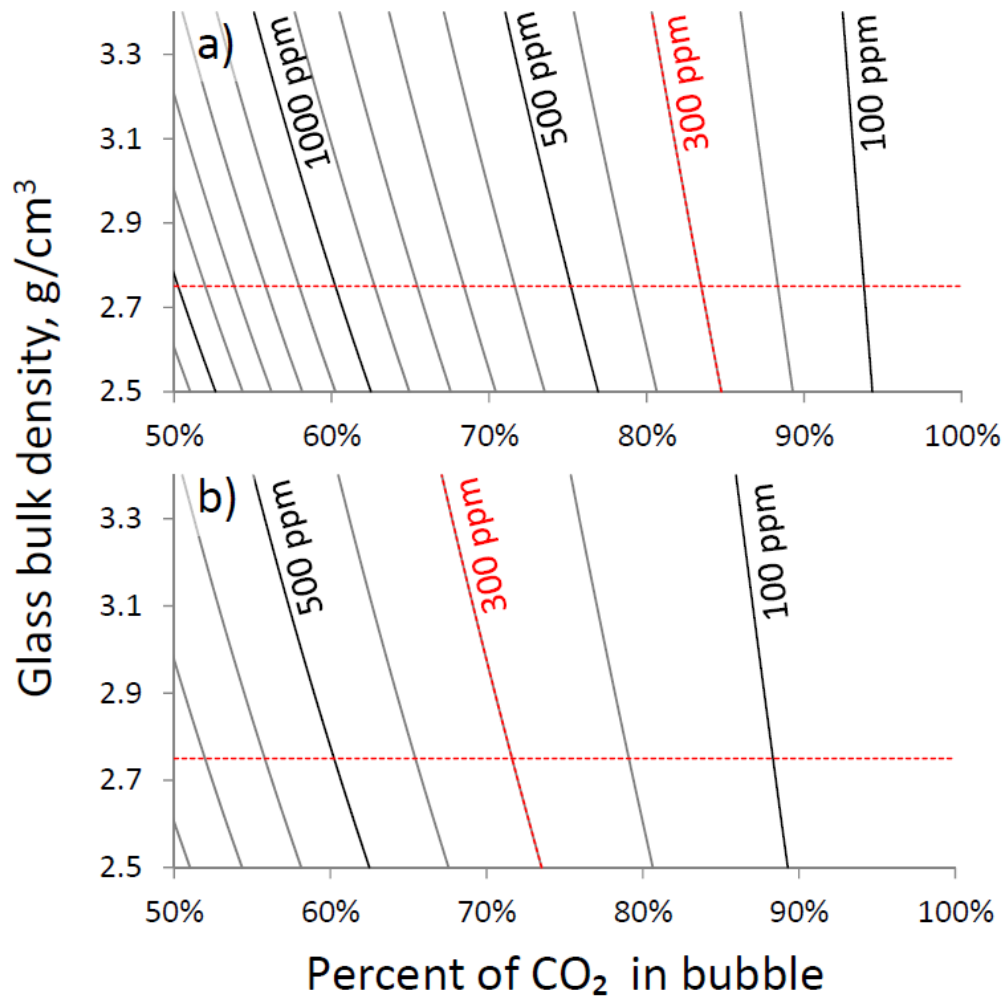
We compounded the errors of our calculations individually by reconstructing the CO_2 content of the bubble in two cases: a case where the bubble contributes the minimum amount of CO_2 , and a case where the bubble contributes the maximum amount of CO_2 . We then calculated the range in reconstructed CO_2 concentrations from these endmember values. In the case for a

minimum bubble concentration, we used the smallest bubble diameter (measured diameter – 0.5 μm error), the largest possible MI (measured diameters + 0.5 μm error) where the depth of the inclusion was assumed equal to the longest measured diameter, the greatest possible bulk density (3.0 g/cm^3), the lowest possible bubble density (calculated density – 19% error), and the greatest possible concentration of CO_2 in the bubble (measured value + error). For the case for maximum concentration due to the bubble, we did the opposite: we assumed a larger, denser bubble, a smaller, less dense, MI glass with a lower CO_2 concentration in the glass.

References

Sterner, S. M., Bodnar, R. J. (1984) Synthetic fluid inclusions in natural quartz I. Compositional types synthesized and applications to experimental geochemistry. *Geochimica et Cosmochimica Acta*, 48, 2659–2668.

62 **Figure**



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64 **Figure S1** Calculated isopleths of glass CO₂ concentration as a function of MI glass bulk density
 65 and bubble contribution (percentage of total CO₂ contained in the bubble) with CO₂ densities in
 66 the bubble of a) 0.1 g/cm³ and b) 0.05 g/cm³. These calculations were done assuming a typical
 67 bubble size (4 volume percent) and vapor density based on our measurements (Tables 2-4). The
 68 bulk density of the MI glass does not affect the reconstructed CO₂ contents significantly, as
 69 shown by sub-vertical isobars.

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