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Reply to:

Abstract
Devine & Rutherford (2014), “Magma storage region processes of the Soufrière Hills Volcano, Montserrat”, published as Chapter 19 of the Geological Society of London Memoir, volume 39, criticised the data and methods of Humphreys et al. (2010), Magma hybridisation and diffusive exchange recorded in heterogeneous glasses from Soufrière Hills Volcano, Montserrat, Geophysical Research Letters, L00E06. We thoroughly review the melt inclusion dataset presented by Humphreys et al. (2010) and show the results to be robust. High volatile contents inferred ‘by difference’ from electron microprobe analysis are supported by direct volatile measurements in multiple independent studies. Plagioclase-hosted melt inclusions in many dome lavas have low H\textsubscript{2}O contents, which we attribute to diffusive loss through the host mineral. Our corrections for post-entrapment crystallisation did not introduce significant artefacts into the dataset that would affect our original conclusions. Rather, the anomalous minor element compositions found in a subset of our melt inclusions and matrix glasses is supported by other independent studies, and can be best explained as a result of mingling and hybridisation with mafic magmas that are compositionally variable through time.

Introduction
The recent study by Devine & Rutherford (2014; hereafter DR2014), “Magma storage region processes of the Soufrière Hills Volcano, Montserrat”, published as Chapter 19 of the Geological Society of London Memoir, volume 39, focused primarily on updating the record of magmatic temperatures recorded by Fe-Ti oxides in the andesite erupted from Soufrière Hills Volcano. However, a key result of the paper was that the compositions of some plagioclase-hosted melt inclusions indicate mixing of mafic magma components into the host andesite and trapping of the mixed melts within phenocrysts. This interpretation is consistent with our earlier work on the chemistry of melt inclusions (Humphreys et al. 2010) and of microlite crystal populations (Humphreys et al. 2009; 2013). Humphreys et al. (2010) proposed this interpretation by showing that a subset of Soufrière Hills melt inclusions and matrix glasses has anomalous concentrations of K$_2$O and/or TiO$_2$, as have mafic inclusion matrix glasses (see figure 2, Humphreys et al. 2010). However, in their Appendix, DR2014 suggest that enrichment in K$_2$O can only result from decompression crystallisation and not from magma mingling. They also assert that our melt inclusion dataset is fundamentally flawed and subject to faulty corrections for post-entrapment correction; and that our melt inclusions were actually matrix glasses. Finally, DR2014 doubt “whether or not one can look at melt-inclusion analyses and distinguish the effects of decompression crystallization… from the effects of mingling… with components derived from the injected mafic magma”.

Here we address the criticisms of the DR2014 Appendix, drawing on additional published data to support our arguments and explanations. We were unable to reproduce some of the key figures in DR2014, so we present revised, corrected versions. We demonstrate clearly the compositional effects of decompression crystallization and discuss carefully how this can be distinguished from mingling. Finally, we consider thoroughly, though ultimately reject, the suggestion that enrichment of melt inclusions and matrix glasses in K$_2$O can only be derived from slow crystallisation, and give our supporting arguments.

Volatile contents of melt inclusions and matrix glasses

Devine & Rutherford (2014) firstly called into question our estimates of melt volatile contents for melt inclusions and matrix glasses, as published in Humphreys et al. (2010), and suggested that some of our melt inclusions might actually be matrix
glasses. The volatile contents of the earliest erupted magmas at Soufrière Hills Volcano (from 1996) were reported to be up to ~4.6 wt% H₂O based on ‘volatiles by difference’ (VBD) from 100% analytical total by electron microprobe (Devine et al. 1998). This method has an uncertainty of approximately 0.6 wt% H₂O (Devine et al. 1995; Humphreys et al. 2006a). This result was supported by FTIR analyses of six quartz-hosted melt inclusions (Barclay et al. 1998), with H₂O contents of 3.52 – 5.05 wt% and <60 ppm CO₂. We note that this paper (on which both Devine and Rutherford were co-authors) also reports the volatile contents of two plagioclase-hosted melt inclusions, as 4.7 wt% and 0 wt% H₂O. In comparison, our VBD estimates of melt H₂O contents in pristine, plagioclase-hosted melt inclusions are similar but range from 0 up to 8.2 wt% (Humphreys et al. 2010). Obtaining anhydrous glass EPMA data with analytical totals in the region of 100% is consistent with the propagated uncertainty of the electron probe analyses and the VBD method (Devine et al. 1995; Humphreys et al. 2006a); for our dataset this fully propagated uncertainty was typically 1.2 wt%. We ran a suite of hydrous glass secondary standards (Humphreys et al. 2006a) at the same time as the unknowns and these give an average absolute deviation of VBD relative to known H₂O of ~1 wt%. We agree that our highest VBD values are probably an overestimate, but broadly, the high inferred H₂O concentrations are supported by direct analysis of H₂O in a subset of the same pumice-hosted inclusions by secondary ion mass spectrometry (SIMS), which gave 1.14 – 6.24 wt% H₂O (Humphreys et al. 2009a; figure 1). Likewise, FTIR measurements by Mann et al. (2013) gave 1.2 to 6.7 wt% H₂O in quartz- and plagioclase-hosted melt inclusions from pumices erupted in Vulcanian explosions during 1997 and 2003-2004. These values are consistent with our earlier measurements. We therefore do not consider our analyses to be a “divergence” from the DR2014 dataset. There need be no expectation that the maximum volatile contents of multiple aliquots of magma erupted over 15 years should be identical; melt inclusion datasets from volcanoes globally commonly show variable H₂O contents (e.g. Zimmer et al. 2010; Plank et al. 2013). However, the fact that the highest H₂O contents were found in samples erupted in January 2007 by Humphreys et al. (2010), and also in samples erupted in August 1997 by Mann et al. (2013) suggests that maximum volatile contents have probably not changed substantially over the course of the eruption. Suites of melt inclusions typically show a wide range of H₂O
contents, so the very limited data presented in the original work (Devine et al. 1998) are entirely consistent with the more detailed, recent studies.

Our volatile data for pumice-hosted melt inclusions equate to entrapment pressures of at least 220 MPa, or more if substantial CO$_2$ is present, for which there is good evidence from both gas emissions and melt inclusion data (Edmonds et al. 2014). Thus the melt inclusions define a range of pressures that is consistent with a vertically protracted magma reservoir of the type inferred from seismic tomography and from ground deformation (Elsworth et al. 2008; Voight et al. 2010; Elsworth et al. 2014). We interpreted the range of H$_2$O contents to be the result of differential entrapment pressures of the melt inclusion suite during decompression crystallisation, as previously proposed elsewhere including Shiveluch Volcano, Kamchatka (Humphreys et al. 2006b; 2008), Mount St Helens (Blundy & Cashman 2005), Stromboli, Italy (Metrich et al. 2001) and Jorullo, Mexico (Johnson et al. 2008) amongst others. This is consistent with the low volatile contents found for matrix glass (generally less than 0.5 wt% H$_2$O; see below) and with the overall geochemical variations of the melt inclusion and matrix glass data. These data show increasing concentrations of incompatible elements (e.g. K, Ti, Mg) and decreasing concentrations of elements that are compatible in plagioclase (e.g. Ca, Na) with increasing SiO$_2$. We accept that in part this may be related to our application of corrections for post-entrapment crystallisation, although this does not affect the key elements of interest (e.g. K, Ti, Fe; see below for further discussion). However, the combined dataset for matrix glasses (which of course are uncorrected) and melt inclusions is consistent with progressive fractionation of plagioclase together with minor pyroxenes + oxides during decompression (see figure 1 of Humphreys et al. 2010). This interpretation fits with observations of substantial groundmass crystallisation of these same phases. The compositions are also generally consistent with the results of phase equilibria experiments of Couch et al. (2003) which resulted in crystallisation of plagioclase and pyroxene (see later). We note that Mann et al. (2013) interpreted the variable H$_2$O contents of melt inclusions to reflect hydrogen loss from the melt inclusions during magma stalling in the conduit rather than variable entrapment pressures. However, the co-variation of H$_2$O and Cl measured by SIMS (e.g. Humphreys et al. 2009a) in some pumice-hosted melt inclusions would argue against this possibility, as Cl presumably
cannot be lost easily by diffusion through the host phenocryst but is known to degas
together with $\text{H}_2\text{O}$ during decompression (e.g. Villemant & Boudon 1999).

We do not dispute that plagioclase-hosted melt inclusions in many Montserrat dome
samples (including those dome samples studied by Humphreys et al. 2010) may have
substantially or completely lost their water; they are ‘leaked’ melt inclusions and not
matrix glasses. This is the reason that we excluded these samples from our study of
$\text{H}_2\text{O}$ and Cl degassing (Humphreys et al. 2009a). By comparison with the well
documented similar effect in both natural and experimental olivines (Hauri 2002;
Portnyagin et al. 2008; Gaetani et al. 2012) we infer that water loss may result from
diffusion of H through the host phenocryst, as demonstrated experimentally by
Johnson & Rossman (2013). The extent of diffusive loss likely depends on clast size
(Lloyd et al. 2013) as well as the permeability of the dome, and probably occurs
during prolonged storage at low pressures (perhaps in the lava dome itself) and at, or
close to, magmatic temperatures. We would anticipate that H loss through the host
phenocryst may explain anomalous trends in $\text{CO}_2$-$\text{H}_2\text{O}$ space that are otherwise
attributed to re-equilibration with highly $\text{CO}_2$-rich vapours (e.g. Collins et al. 2009 for
olivine-hosted inclusions). If this is the case, we would also expect these inclusions to
have anomalously oxidising compositions (e.g. Gaetani et al. 2012; Humphreys et al.
in press).

**Corrections for post-entrapment crystallisation**

In the supplementary dataset to our previous study (Humphreys et al. 2010) we
provided petrographic information for each melt inclusion in our original dataset
(including a description of shape, colour, presence or absence of bubble, textural
association in the host phenocryst, area, equivalent radius and volume, area and
volume of any post-entrapment crystallisation if present, as well as the raw, PEC-
adjusted and normalised anhydrous compositions). We now also present
representative backscattered SEM images of typical inclusions in figure 2. We also
estimated the amount of post-entrapment crystallisation (PEC) in each melt inclusion,
and attempted to correct the melt inclusion compositions for the observed PEC. In our
explanation of the correction procedure we used the term ‘pristine’ to refer to melt
inclusions that were free from post-entrapment crystallisation and thus did not need
correction. For the other inclusions, our post-entrapment crystallisation procedure
followed that of Saito et al. (2005), as described in the supplementary information to Humphreys et al. (2010). The method was indeed developed for basaltic melt inclusions but the principle is not changed by the composition of the melt or host phenocryst. For plagioclase-hosted melt inclusions in particular, it is not always clear exactly what host composition is truly in equilibrium with the inclusion as the textures may be complex; precisely defining the volume and surface area of the inclusions may also be very difficult (Humphreys et al. 2008). For this reason, we used a constant plagioclase composition of An$_{40}$ and made simplifying assumptions about the melt inclusion shape. This may have introduced some uncertainty or error into the corrected dataset, mainly for the key components of the host plagioclase (SiO$_2$, Al$_2$O$_3$, CaO and Na$_2$O; see discussion above), although we also provided the uncorrected compositions for reference. For completeness we show the effect of these corrections in the haplogranite ternary Ab-Or-Q (figure 3). However, we argue that for the minor elements of particular interest to this discussion, i.e. FeO, TiO$_2$ and K$_2$O, the PEC correction makes no significant difference.

DR2014 commented on the presence of normative Wo in some corrected melt inclusion compositions. All our compositions plotted were suitable for plotting in the haplogranite projection following Blundy & Cashman (2001), which requires glasses with < 20 wt% normative anorthite. All our published glasses have ≤16 wt% normative anorthite but typical values are <<10 wt%. Normative wollastonite is calculated for a minority of compositions but this amounts to an average of 0.85 wt% ± 0.54 normative Wo. This amount of normative Wo is equivalent to a very minor shift in the position of a few glasses within the haplogranite ternary, away from the Qz’’ apex (a shift smaller than the size of the symbols in figure 2). If anything, this may show that our PEC-corrected compositions have slightly overestimated CaO concentrations, but this does not affect the nature of our interpretations, which are largely based on minor element compositional variations. We would also add that many of DR2014’s own data show just as high dispersion in the haplogranite projection as ours (e.g. DR2014 figure 19A.6 and 19A.7) and that our data are not unique in having a small amount of normative wollastonite (see also Edmonds et al. 2002; Mann et al. 2013).

The anomalous groundmass composition of Soufrière Hills magma
Figure 19A.11 of DR2014 shows the anomalous composition of the bulk groundmass from Soufrière Hills magma, as recognised by several authors previously. Although DR2014 state that this may be due to analytical artefacts caused by inaccurate raster analysis of inhomogeneous groundmass, there are alternative explanations. One is that the groundmass is contaminated by disaggregation of mafic enclaves and transfer of small crystals into the andesite groundmass, including clinopyroxene and Ca-rich plagioclase (Humphreys et al. 2009b; 2013). This is consistent with the interpretation that the anomalous glass compositions may be related to hybridisation; see below (Humphreys et al. 2010; Devine & Rutherford 2014). It has also been suggested that the bulk groundmass composition could be affected by incorporation of substantial additional SiO$_2$ through precipitation of cristobalite in vesicles (Horwell et al. 2012).

**K$_2$O enrichment during slow decompression crystallisation?**

The substantive criticism of DR2014 about our earlier work is that the variably high K$_2$O contents seen in a subset of our matrix glasses and melt inclusions may be related to slow crystallisation of the andesite during ascent and decompression, rather than to hybridisation with mafic enclave components as we suggested (Humphreys et al. 2010). Occasional high-K glasses can also be observed in several previous studies of SHV eruptive products and include matrix glasses and melt inclusions in both plagioclase and hornblende (e.g. Edmonds et al. 2002; Harford et al. 2003; and in particular Buckley et al. 2006). Anomalous high-K melt inclusions have also been observed at Colima Volcano, Mexico, where they have been ascribed to recording heterogeneity in the melt caused by the breakdown of amphibole nodules (Reubi & Blundy 2008). In our earlier work we considered this process, alongside the breakdown of amphibole during heating or decompression (Buckley et al. 2006), but concluded that this could not fully explain either the compositions or the textural associations of the glasses. We also considered the possibility of entrapment of anomalous boundary layer melts into melt inclusions (Baker 1991) but rejected this on the basis that matrix glasses were also affected.

The suggestion of DR2014 that K$_2$O variability in matrix glasses and melt inclusions could be due to variations in magma ascent rate (and thus extent of decompression crystallisation) is interesting. This was based on experiments conducted by Hammer & Rutherford (2002), which produced high-K, low-Na glasses through decompression
crystallization of plagioclase. In their experiments, isothermal decompression of Pinatubo dacite resulted in groundmass crystallisation of plagioclase + quartz, with other phases in minor abundance. K$_2$O is essentially incompatible in these phases, so K$_2$O contents of the experimental matrix glasses increase with increasing groundmass crystallinity, accompanied by decreasing CaO, which is compatible in plagioclase. In those experiments, increasing the dwell time at the final pressure actually resulted in a greater spread of K$_2$O contents, rather than a uniform increase. However, the same trend of increasing K$_2$O and decreasing CaO is also observed, more clearly, in the experimental studies of Martel & Schmidt (2003), Couch et al. (2003) and Brugger & Hammer (2010). The experiments of Martel & Schmidt (2003) are particularly appropriate because their starting material was chosen to have the composition of the most evolved (i.e. highest SiO$_2$ on an anhydrous basis) plagioclase-hosted melt inclusions from Devine et al. (1998), equivalent to melts in equilibrium with phenocryst rims during decompression crystallisation (Martel & Schmidt 2003). In all sets of experiments, the very high-K compositions are achieved only at very low final pressures (e.g. 15 MPa, Martel & Schmidt 2003; <25 MPa, Hammer & Rutherford 2002; <20 MPa, Brugger & Hammer 2010), presumably as the melt approaches saturation in K-feldspar. The strong decrease in CaO that accompanies K$_2$O enrichment has important implications for plagioclase-melt thermometry (Humphreys et al. 2014).

DR2014 rightly point out that we do not find both high-K and low-K matrix glasses in the same sample. However, we do find both high-K and low-K melt inclusions in the same sample, and in the same crystal. Although our most evolved matrix glass compositions are similar to the very low pressure, high-K experimental glasses (figures 4, 5), it seems clear that slow, low-pressure decompression crystallisation cannot explain the enrichment of those compositions in TiO$_2$ (figure 4). Specifically, the experiments show that the very high K$_2$O contents can only be reached after extensive crystallisation at low pressure, resulting in extremely low CaO and MgO or FeO concentrations; this is not consistent with the spread to higher CaO and FeO at high K$_2$O contents. We therefore conclude that the most likely explanation for the anomalous, high-K melt compositions is still that of hybridisation with mafic-derived melt components.
Compositions of mafic enclave glasses

The compositional overlap of high-K melt inclusions and matrix glasses with mafic enclave glasses led us to propose that the anomalous, high-K glasses were derived from disaggregation of mafic material and hybridisation with the andesite (Humphreys et al. 2010). This would be consistent with similar observations made on the compositions of microlite crystal populations (Humphreys et al. 2009b; 2013). In our original study, the key observation was that both K$_2$O and TiO$_2$ anomalies are observed in melt inclusions and matrix glasses, and that they were apparently decoupled (see figure 2 of Humphreys et al. 2010). This decoupling led us to propose our model of diffusive fractionation, based on the facts that a) K and Ti are enriched in mafic enclave glasses relative to residual liquids in the andesite, generating a chemical gradient, and b) the diffusivity of K in melt is significantly faster than that of Ti (e.g. Bindeman & Davis 1999, Richter et al. 2003) raising the possibility for fractionation. In the main body of their paper, DR2014 supported our interpretation that the variance of glass chemistry was caused by disaggregation and mixing with mafic-derived components, though they based this on FeO contents in addition to TiO$_2$. Their issue with our interpretation seems to arise from the fact that their mafic enclave glasses were not enriched in K$_2$O, whereas ours were (figures 4-6). We have discussed the possible alternative for K-enrichment, that of low pressure crystallisation, above. If this were the controlling influence on mafic glass composition, comparison with experimental studies would imply that the mafic enclaves studied by DR2014 were quenched at very high pressure (~200 MPa) whereas those studied by Humphreys et al. (2010) quenched over a range of pressures, but below ~35 MPa; this seems implausible.

An alternative explanation is offered by compositions reported by more recent work (Mann 2010; Plail et al. 2014; Plail, 2014). Plail et al. (2014) identified two texturally and geochemically distinct types of mafic enclaves in Phase 5 of the eruption (rocks erupted during 2009-2010), as well as a third type that was a hybrid of the first two. Type A enclaves are glassy and vesicular, with more mafic compositions, and the framework-forming phase is high-Al amphibole. Type B enclaves are more evolved (less mafic, with higher SiO$_2$) in composition, with higher crystallinity and lower vesicularity; the framework-forming phase is plagioclase and high-Al amphibole is rare to absent (Plail et al. 2014). Type A enclaves are thought to form by rapid
thermal equilibration and vesiculation of the enclave magma during injection into the andesite. Type B enclaves were inferred to have resulted from significant hybridisation of enclave magma with the andesite, associated with slower cooling (Plail et al. 2014). The two types of mafic enclaves have very different residual matrix glass compositions, even for enclaves erupted in the same andesite magma (Plail et al. 2014). Type A enclaves are characterised by variably high FeO, TiO$_2$ and MgO. Type B enclaves have compositions that are more similar to the host andesite matrix glass (figure 4). Importantly, the rapidly quenched, primitive Type A enclaves show strong enrichment in K$_2$O, whereas the more slowly crystallised Type B are only slightly enriched (figure 4). Figures 4-6 show clearly the disparity between Type A and Type B mafic enclave glasses.

Devine & Rutherford (2014) suggested that the difference between their mafic enclave analyses and ours (see DR2014 figure 19A.10) is because the mafic enclaves in our study had undergone slower crystallisation in the conduit, resulting in a spread towards K$_2$O enrichment. In fact, the comparison with additional data shows that the DR2014 mafic glass compositions are unusual in having particularly high TiO$_2$ and FeO concentrations, yet no K$_2$O enrichment (figures 5,6). The contrast in Type A and Type B enclave glasses also suggests that variations in decompression rate are unlikely to be the cause for the observed compositional differences, as the most K$_2$O-enriched (Type A) enclave glasses are derived from enclaves that quenched rapidly (Plail et al. 2014), not those that crystallised slowly. Based on minor element compositional variations (figures 4 and 5), we therefore suggest that it is more likely that the Soufrière Hills magma system is regularly fluxed by mafic melts of variable composition. This should be investigated further using trace elements and isotopic compositions, to investigate possible heterogeneity in the magma source regions and thus melt generation processes.

Conclusions

We have thoroughly reviewed the melt inclusion dataset presented by Humphreys et al. (2010) and showed the results to be robust. High volatile contents inferred ‘by difference’ from electron microprobe analysis are supported by direct measurements in multiple independent studies. Plagioclase-hosted melt inclusions in many dome lavas have low H$_2$O contents, which we attribute to diffusive loss through the host
mineral. The anomalous K$_2$O-enrichment found in a subset of our melt inclusions and matrix glasses is supported by several other independent studies, and can be best explained as a result of mingling and hybridisation with mafic magma, as originally proposed. Our corrections for post-entrapment crystallisation did not introduce significant artefacts into the dataset that would contradict these conclusions. Additional mafic glass data from the recent literature shows that instead, intruding mafic magmas at Soufrière Hills were probably variable in composition; we therefore recommend further investigation using trace elements and isotopes.

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Figure captions

Figure 1. Comparison of volatile contents estimated by secondary ion mass spectrometry (SIMS) and ‘by difference’ from electron microprobe totals, for melt inclusions from Humphreys et al. (2009a).

Figure 2. Back-scattered electron SEM images for typical melt inclusions. White arrows mark inclusions analysed.


Figure 5. Variation of (a) CaO vs K$_2$O and (b) FeO vs K$_2$O concentration in melt inclusions and matrix glasses from Soufrière Hills Volcano. Symbols as in figure 4. Grey fields are as in figure 4; black arrow and numbers denote the effect of decreasing pressure in the Martel & Schmidt experiments, with approximate
equilibration pressures in MPa. Extremely low pressure crystallisation is required to produce residual matrix glasses with strongly enriched K$_2$O contents.

Figure 6. Haplogranite ternary showing variation in mafic enclave glass compositions.
Symbols as in figure 4.
Figure 1
Figure 3

- H et al. (2010) melt inclusions
- H et al. (2010) mg
- Couch et al. (2003) experimental glasses
- DR2014 melt inclusions and mg
- Mann et al. (2013) melt inclusions
Figure 4
Figure 5
Figure 6