- Mafic enclaves record syn-eruptive basalt intrusion and mixing 1 2 Melissa Plail^{1,2}, Marie Edmonds³*, Andrew Woods³, Jenni Barclay¹, Madeleine C.S. 3 Humphreys⁴, Richard A. Herd¹, Thomas Christopher^{5,6} 4 5 6 ¹School of Environmental Sciences, University of East Anglia, Norwich, NR4 7TJ, UK 7 ²School of Geosciences, Witwatersand University, Johannesburg, South Africa 8 ³Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge, CB2 3EQ, UK 9 ⁴Department of Earth Sciences, University of Durham, Durham 10 ⁵Montserrat Volcano Observatory, Montserrat 11 ⁶Seismic Research Centre, University of the West Indies St Augustine, Trinidad and Tobago W.I. 12 13 *corresponding author: marie.edmonds@esc.cam.ac.uk
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15 Abstract

Mafic enclaves hosted by andesite erupted at the Soufrière Hills Volcano between 1995 and 16 17 2010 yield insights into syn-eruptive mafic underplating of an andesite magma reservoir, 18 magma mixing and its role in sustaining eruptions that may be widely applicable in volcanic 19 arc settings. The mafic enclaves range in composition from basalt to andesite and are 20 generated from a hybrid thermal boundary layer at the interface between the two magmas, 21 where the basalt quenches against the cooler andesite, and the two magmas mix. We show, 22 using an analytical model, that the enclaves are generated when the hybrid layer, just a few 23 tens of centimetres thick, becomes buoyant and forms plumes which rise up into the andesite. 24 Mafic enclave geochemistry suggests that vapor-saturated basalt was underplated quasi-25 continuously throughout the first three eruptive phases of the eruption (the end member basalt 26 became more Mg and V-rich over time). The andesite erupted during the final phases of the 27 eruption contained more abundant and larger enclaves, and the enclaves were more 28 extensively hybridised with the andesite, suggesting that at some time during the final few 29 years of the eruption, the intrusion of mafic magma at depth ceased, allowing the hybrid layer to reach a greater thickness, generating larger mafic enclaves. The temporal trends in mafic 30 31 enclave composition and abundance suggests that basalt recharge and underplating sustained 32 the eruption by the transfer of heat and volatiles across the interface and when the recharge 33 ceased, the eruption waned. Our study has important implications for the petrological 34 monitoring of long-lived arc eruptions.

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36 Keywords: Mafic enclaves, andesite, arc, mixing, eruption triggering

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38 1. Introduction

39 Andesites are the most common volcanic rock type erupted at convergent margins and are 40 fundamental to the formation and evolution of continental crust (Rudnick, 1995). It is 41 increasingly clear that many intermediate magmas in arcs are hybrids, formed during long 42 residence periods in the crust (Cooper and Kent, 2014) and recharged frequently by mafic 43 magmas (Eichelberger et al., 2000; Reubi and Blundy, 2009). Many intermediate 44 composition magmas erupted in arcs preserve evidence of mingling and mixing with mafic 45 magma shortly prior to eruption in the form of disequilibrium (Nakagawa et al., 2002; Singer 46 et al., 1995; Tepley et al., 1999) or heating textures, diverse isotopic compositions (Davidson 47 and Tepley, 1997; White et al., 2017), the presence of cryptic mafic components (Humphreys 48 et al., 2009b) or macroscopic mafic enclaves (Bacon, 1986; Browne et al., 2006; Clynne, 49 1999; Martin et al., 2006). It remains unclear, however, whether mafic recharge of reservoirs 50 is in itself a trigger for eruption (Murphy et al., 2000; Tepley et al., 1999; Victoria et al., 51 2008), or an outcome of unrest and impending eruption (Christopher et al., 2015). Such 52 mingled magmas have been erupted at Mount Unzen, Japan (1991-1995) (Browne et al., 53 2006), Lassen Peak (Clynne, 1999; Tepley et al., 1999), Augustine volcano, Alaska (2006) 54 (De Angelis et al., 2013; Nakamura, 1995), Arenal, Costa Rica (Reagan et al., 1987) and 55 Soufrière Hills Volcano, Montserrat (Murphy et al., 2000).

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57 Mafic enclaves typically comprise a complex hybrid assemblage of groundmass crystals, 58 residual melt and host magma-derived components, such as "inherited" crystals and melt 59 derived from the host magma (Bacon, 1986; Clynne, 1999; Humphreys et al., 2013; 60 Humphreys et al., 2009b; Plail et al., 2014). Further, host magmas often contain crystals that 61 preserve evidence of their admixture and subsequent ejection from the mafic magma. Thus, 62 suites of cognate enclaves display a range of compositions and textures that reflect variable 63 extents of differentiation of the mafic magma, or the degree of mixing with the host magma 64 and/or changes in end-member magma composition (Bacon, 1986; Browne et al., 2006; 65 Clynne, 1999). While we are making progress in understanding the role of mafic underplating 66 in heating and mobilising magma bodies (Bachmann and Bergantz, 2006; Bergantz et al., 67 2015; Huber et al., 2011), we lack an overarching understanding of the controls on the nature 68 of mixing with overlying host magmas. It is not known, for example, whether the enclaves 69 reflect quasi-continuous, syn-eruptive mafic underplating, or whether episodes of mafic intrusion are decoupled from eruptions. Host and mafic enclave compositions may evolve
during eruptions; these trends might allow understanding of how magmas mingle and on
what timescales and what implications this may have for eruption triggering and sustenance.

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74 There is evidence from a wide range of volcanic systems of magma mixing and mingling 75 between a "host" evolved magma with intruding mafic magma, suggesting this is a common 76 process and integral to the petrogenesis of complex, hybrid andesites. As well as the 77 macroscopic evidence in the form of decimeter-scale mafic enclaves, there is abundant 78 textural and petrological evidence for mixing and mingling. In Soufrière Hills lavas, the 79 presence of plagioclase, pyroxene and oxide microlites, crystal clots, plagioclase phenocrysts 80 with high Fe rims and high K_2O glass derived from the disaggregation of mafic enclaves 81 (Humphreys et al., 2013; Humphreys et al., 2009b), shows that the bulk andesite contains up 82 to 6 % vol. of a 'cryptic' mafic component (Humphreys et al., 2013) as well as the 1-12 % 83 vol. in the form of mafic enclaves (Barclay et al., 2010; Komorowski et al., 2010; Mann et 84 al., 2013; Murphy et al., 2000; Plail et al., 2014). In Kanga Island in the Aleutian arc, 85 decreasing andesite bulk SiO₂ concentrations have been correlated with increasing mafic 86 cumulate crystal clots proportions (Brophy, 2009). The 1953-1974 eruption of Trident 87 volcano in Katmai National Park Alaska also shows a decrease in the host dacite bulk SiO₂ 88 over the course of the eruption as the storage region homogenises with mafic magma 89 (Coombs et al., 2000). The host magma erupted at Unzen, Japan during the 1991-5 eruption 90 is calculated to contain between 10 and 25 % mafic magma (Browne et al., 2006), which is a 91 result of multiple mixing events (Vogel et al., 2008). The mixing efficiency may change over 92 the course of an eruption caused by gradual changes in bulk viscosity and temperature of the 93 overlying magma, as demonstrated in the 1915 eruption products of Lassen Peak, USA 94 (Clynne, 1999; Klemetti and Clynne, 2014; Sparks and Marshall, 1986). The degree and style 95 of magma mixing can strongly influence eruptive style, as demonstrated by eruptions at 96 Quizapu, Chile (Ruprecht and Bachmann, 2010). The 1846-47 effusive eruption with 97 geochemical and petrological evidence for magma mixing resulted significant reheating of 98 host magma from 830 to 1000 °C; this mixing enhanced magma degassing, but reduced rapid 99 magma expansion and explosive behaviour. Changing mafic enclave compositions over the 100 lifetime of an eruption, where magma mingling is prevalent, have also been observed during 101 many eruptions, but the significance of this is unclear. At Unzen, Japan, the repeated 102 intrusion of two mafic magmas was inferred from differences in REE and trace elements 103 between two distinct mafic enclave types (Vogel et al., 2008). At Shirataka volcano Japan,

two mafic enclave types are identified on the basis of high and low K₂O over a 200 kyr
period (Hirotani and Ban, 2006).

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107 In this paper we present whole rock major and trace element data and glass major element 108 data for andesites and their mafic enclaves erupted during the 1995-2010 eruption of the 109 Soufrière Hills Volcano (Montserrat, West Indies) (Wadge et al., 2014). This well-studied eruption produced magma mixing and mingling features that are common to many 110 111 intermediate arc magmas globally. The data are used to infer the petrogenesis of the enclaves 112 and to develop an analytical model to describe the formation of the enclaves from a mixed 113 boundary layer at the interface between mafic magma and overlying andesite. The temporal 114 trends in composition are used to assess whether basalt was intruded in a discrete event at the 115 beginning of the eruption, or quasi-continuously throughout, and whether enclave 116 compositions might be useful for evaluating whether long-lived eruptions may be waning.

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118 *1.1 Geological setting*

119 Most of the volcanic centres on Montserrat (West Indies; Figure 1) are andesitic, with mafic 120 enclaves ubiquitous in the lava domes and block and ash deposits. South Soufrière Hills 121 Volcano, which abuts Soufrière Hills at the south of the island (Figure 1) has erupted 122 magmas of basaltic to basaltic andesite composition (Cassidy et al., 2015; Zellmer et al., 123 2003a). The recent eruptions of Soufrière Hills included five phases of andesitic dome-124 forming activity, separated by pauses in lava extrusion (Table 1) (Wadge et al., 2014), 125 allowing examination of temporal trends in magma composition. Samples of andesite and 126 mafic enclaves emplaced during phases III, IV and V were collected at locations shown in 127 Supplementary Table 1. Data for phases I and II are compiled from the literature (Mann et 128 al., 2013; Murphy et al., 2000; Zellmer et al., 2003a).

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130 Previous work has characterised the andesite and its mafic inclusions. The complex zoning 131 patterns and disparate age histories of plagioclase phenocrysts indicate that the andesite is a 132 hybrid that has undergone multiple cooling and reheating events in the shallow crust for 10^{3} -133 10⁴ years prior to the current eruption (Zellmer et al., 2003a; Zellmer et al., 2003b). Excess 134 sulfur emissions, mafic enclaves and phenocryst disequilibria textures (Barclay et al., 1998; 135 Edmonds et al., 2001; Humphreys et al., 2009b; Murphy et al., 2000; Murphy et al., 1998; 136 Sparks et al., 1998) are interpreted as evidence of an intrusion of a hotter, volatile-saturated 137 mafic magma at depth underplating the andesite (Christopher et al., 2010). Remobilisation 138 and reheating of the crystal-rich andesite has been proposed to have been initialised via 139 'convective self-mixing', 'gas sparging' or 'a defrosting front' (Bachmann and Bergantz, 2006; Burgisser and Bergantz, 2011; Couch et al., 2001). Diffusion profiles across Fe-Ti 140 141 oxides in the andesite constrain the timing of the interaction between mafic and andesitic magmas to be days to weeks before eruption (Devine et al., 2003). Seismic "crises" which 142 143 occurred prior to the onset of the 1995-2011 eruption and in 1933-7 and 1966-7 (Shepherd et 144 al., 1971) have been proposed to have been caused by magma intrusion at depth (Aspinall et 145 al., 1998).

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147 The volume fraction of mafic enclaves in the erupted and esite has increased from $\sim 1\%$ in phase I to ~8-12% in phase V (Barclay et al., 2010; Komorowski et al., 2010; Murphy et al., 148 149 2000; Plail et al., 2014), but the causes for this are enigmatic. Substantial quantities of mafic-150 derived material are also found disaggregated at the crystal-scale within the andesite (Humphreys et al., 2013). Mafic enclaves have become larger with time: 87% of 158 phase I 151 152 hybrid enclaves observed in the field were under 6 cm in longest dimension (Plail et al., 153 2014). In phase III, the maximum size for an enclave measured was 14 cm (Barclay et al, 154 2010). The largest phase V enclave measured to date is 26 cm.

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156 **2.** Samples and Methods

157 Samples of lava dome blocks containing mafic enclaves were acquired from block and ash 158 deposits erupted during phases III to V (details of sampling locations and deposit types are given in Supplementary Material). Splits of samples were crushed and powdered and 159 160 blocks were also made into 30 micron thick probe sections. Seventy-four powders from mafic enclave and andesite samples were analysed using X-ray fluorescence (XRF) (details of 161 162 methods and precision are given in **Supplementary Material**). Glasses in the groundmass of 163 mafic enclaves and andesite samples were analysed using a Cameca 5-spectrometer SX-100 164 electron probe at the University of Cambridge (details of operating conditions and standards 165 are given in Supplementary Material).

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167 **3. Results**

168 3.1. Host andesite geochemistry

The andesite lavas from Soufrière Hills Volcano have a narrow range of bulk compositions
(58–63 wt % SiO₂), and are highly porphyritic, with 45-55% macrocrysts (Barclay et al.,
1998; Devine et al., 1998; Humphreys et al., 2009b; Murphy et al., 2000) (figure 2). The

172 macrocryst assemblage is predominantly plagioclase, hornblende, orthopyroxene and Fe-Ti 173 oxides (ilmenite and magnetite). The groundmass assemblage is plagioclase, orthopyroxene, 174 clinopyroxene and Fe-Ti oxides, with variable amounts of rhyolitic glass (Humphreys et al., 175 2010; Humphreys et al., 2009b). Soufrière Hills Volcano andesite falls in within the calc-176 alkaline field (Miyashiro, 1974) (figure 2). Major element covariation plots display linear 177 trends (figure 2). Matrix glass in the andesite is of rhyolitic composition, with 73–80 wt % SiO₂ (figure 2) and variable K₂O, FeO and TiO₂ concentrations (Humphreys et al., 2010). 178 179 Trace element profiles are shown in figure 3 and show, in general, enrichments in the large 180 ion lithophile elements (LILE) Rb, K and Ba, and depletions in the high field strength (HFS) 181 elements Ti, Nb and Ta and a trough-like pattern in the MREE to HREE. Covariation plots of 182 trace elements display linear trends (figure 4): Dy and Yb increase with increasing La, 183 whereas V, Sr and Sc decrease with increasing La.

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185 3.2. Mafic enclave geochemistry

Mafic enclaves have a diktytaxitic groundmass framework of elongate, randomly-oriented crystals indicative of quench crystallisation (**figure 1**). The groundmass consists of plagioclase, clinopyroxene, amphibole, orthopyroxene and Fe-Ti oxides. Variable amounts of interstitial rhyolitic glass are found within the enclaves. 'Inherited' phenocrysts of plagioclase, amphibole and orthopyroxene derived from the andesite are present within the mafic enclaves with sieve textures and morphologies typical of those within the host andesite (Humphreys et al., 2009b; Plail et al., 2014).

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194 The mafic enclaves have a wide range of bulk rock compositions (48–57 wt % SiO₂) (figure 195 2) and they fall in the tholeiite field (Miyashiro, 1974) (figure 2). The suites of mafic enclave 196 compositions from all eruptive phases fall on linear arrays (that include the andesite 197 compositions) for the major elements (Figure 2). Trace element profiles show, similar to the 198 andesite, LILE enrichment and HFS element depletion (figure 3). Overall, the mafic enclave 199 trace element concentrations are more depleted relative to the host andesite in the LILE and 200 some light REE (La to Pr) (figure 3), but there is either enrichment in or overlap with the 201 andesite concentrations in middle REE (Nd, Sm, Eu) the heavy REEs (Dy to Lu) and Zr. 202 Covariation plots of trace elements with La are linear, where Dy and Yb increase whereas V, 203 Sr and Sc decrease with increasing La (Figure 4). Similar to the andesite, most of the trace 204 elements and in particular elements such as Sm, Y, Yb, V, Sc show displacements in 205 composition between eruptive phases (V and Yb against La are shown in Figure 4), with earlier eruptive phases richer in Dy, Yb and incompatibles and poorer in V, Sc. Correlation
 coefficient analysis (R, using the corrcoeff function; Supplementary Material) shows that
 trace elements across the eruptive phases correlate positively with one other apart from the
 Fe-Mg-Ca-Al-V-Ti group, which correlate negatively with all other elements.

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The glass in the mafic enclaves is rhyolitic (70–79 wt% SiO₂) (figure 2) with variable K₂O, FeO, TiO₂, SiO₂, CaO and MgO contents, with considerable overlap between mafic enclave and andesite glass compositions (Figure 2) (Humphreys et al., 2010). Some mafic enclave glasses have anomalously low Na₂O and overall, glass in the mafic enclaves has higher TiO₂ (and lower CaO contents) than the glass from the andesite (Supplementary Material).

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217 *3.3. Temporal variations in magma composition and petrography*

218 Although overall the major, trace and rare earth element patterns of mafic enclaves are similar throughout the eruption, there are changes in the composition of the enclaves and, 219 220 albeit to a lesser extent, the host andesite, through time (figure 5). Host andesite major 221 element compositions show minor displacements in FeO_{tot} and MgO at a fixed SiO₂, with 222 andesite erupted in phases I to II more Fe-rich and Mg-poor than phases III and V andesite 223 (Christopher et al., 2014). These changes are manifest most clearly as a change of 0.5 to 1.0 224 in FeO_{tot}/MgO in both andesite and mafic enclaves at a fixed SiO₂ (figure 2E) and in plots of 225 Mg/Fe and V/La with time (figure 5). The compositional gap in SiO_2 between the mafic 226 enclaves and the andesite (between 55.5 and 57.5 wt%) observed in phases I-III no longer 227 exists in phase V, with enclave and andesite compositions overlapping (figure 2; Plail et al, 228 2014). The SiO₂ content of the host andesites extends to a higher upper range in phases IV and V than in phase I (figure 5). The mean SiO₂ content of the mafic enclaves increases 229 230 towards the end of the eruption: phase I enclaves have on average 52.4 wt% SiO₂ (with a 231 standard deviation of 2 wt%), compared with a mean of 54.0 wt % SiO₂ for phase V enclaves 232 (with a standard deviation 1.6 wt%). The most evolved phase I enclave has 54.7 wt% SiO₂, 233 whereas the most evolved enclave erupted in phase V has 57.2 wt% SiO₂.

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Mafic enclave bulk MgO and FeO compositions (at a fixed SiO₂ content), and to a lesser extent TiO₂ and Al₂O₃, are displaced between phases I to II to III and IV in the mafic enclaves (Barclay *et al.*, 2010) (**figures 2, 5**). Mafic enclaves erupted in phases III and V have systematically higher concentrations of V and Sc and lower concentrations of Y relative

to those erupted during phases I and II (Figure 4, 5). High field strength elements such as Zr,

Hf, Th and U have relatively constant concentrations in the mafic enclaves through the eruptive phases. There is also no variation in the Ba/La and Th/La ratios of mafic enclaves (**Supplementary Material**). Primitive mantle-normalised REE profiles show steepening from phases I to III to V (**Figure 3**) as LREE/MREE ratios increase from phase I (La_N/Sm_N 1.0–1.8) to phase 5 ($La_N/Sm_N = 1.5-2.4$). In the more differentiated enclaves, the MREE (Sm, Eu, Gd, Tb, Dy) are enriched relative to the andesite in phases I to III, but form colinear arrays with the andesite in phase V (**Supplementary Material**).

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248 4. Discussion

249 In summary, the mafic enclaves within the andesite at Soufrière Hills Volcano are basaltic to 250 basaltic andesite in bulk composition, with trace element patterns typical of subduction-251 related magmas. We show in this section that mixing between a basalt end member and the 252 overlying andesite controls the mafic enclave compositions (Figures 2-5). Over time, the end 253 member basalt became slightly more Mg-rich (and Fe-poor) between phases I and III, with 254 associated enrichments in MREE and HREE, suggesting continual recharge of the 255 underplating basaltic layer (as this trend cannot be explained by in situ fractional 256 crystallisation of the basalt beneath the hybrid layer). The geochemical and petrological data are used to develop a model to describe how mafic enclaves form due to the formation of 257 258 buoyant plumes formed in a hybridised boundary layer at the interface between the two 259 magmas, its thickness controlled by thermal diffusion and magma density. We suggest, based 260 on the temporal changes in abundance and size of the enclaves through the eruption, and the 261 more silica-rich nature of the enclaves erupted in phase V, that basaltic recharge may have 262 ceased sometime after phase III, allowing thickening of the hybridised boundary layer between the magmas by thermal diffusion and generation of larger, more hybridised 263 264 enclaves.

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266 4.1. Hybridization between host andesite and intruding mafic magma

First we demonstrate that the range in mafic enclave compositions is not well explained by fractional crystallisation. We use RhyoliteMELTS (Ghiorso and Gualda, 2015) to construct fractional crystallisation trends for the most primitive mafic enclave composition (at water contents of 4 and 6 wt% (Barclay et al., 1998; Edmonds et al., 2014; Humphreys et al., 2009a), pressure of 150 MPa and fO_2 of QFM and QFM+2 (Devine et al., 1998)(shown on **Figure 2**), similar to the analysis by Cassidy et al., (2015). The geochemical trends in the mafic enclave arrays cannot be produced by fractional crystallisation of a mafic end member 274 melt (**figure 2**), unless a very high oxidation state is invoked, which is not consistent with 275 petrological constraints (Devine et al., 1998; Murphy et al., 2000). Furthermore, the 276 systematic straight-line trends instead suggest that mixing is the dominant control on the 277 major element geochemistry of the enclaves.

278 The presence in the mafic enclaves of phenocrysts inherited from the andesite (Murphy et al., 279 2000; Zellmer et al., 2003a; Barclay et al., 2010; Humphreys et al., 2009; Mann 2010; Plail 280 et al., 2014; Humphreys et al., 2013) is further evidence for physical mixing between the 281 mafic and andesitic magmas, indicating that the mafic enclaves have undergone hybridisation 282 with the andesite. Co-linear trends in major elements between the mafic enclaves and 283 andesite suggest that simple mixing can resolve the range of compositions across the eruptive 284 phases (Figures 2, 4), as well as the loss of the compositional gap between the andesite and 285 mafic enclaves in phase V (Figure 2). The range in types of mafic enclave erupted in phase V has been explained previously by differing degrees of interaction between a mafic end 286 287 member and the andesite bulk composition (Plail et al., 2014). "Type A" enclaves are basaltic with a narrow range of compositions (~49-52 wt % SiO₂), and have chilled margins, high-Al 288 289 amphiboles, a higher glass fraction, high vesicularity and low inherited phenocryst 290 abundance. These are interpreted to be closest in composition to the underplating mafic 291 magma end member composition. The more abundant "type B" enclaves have a broad range 292 of compositions (53–57 wt % SiO₂), and are identified by a lack of chilled margins, a lower 293 glass fraction, low vesicularity and high inherited phenocryst abundance ($\sim 25\%$), and rare to 294 absent high-Al amphiboles; these enclaves are interpreted to have been affected most by 295 mixing with the overlying andesite. As well as type A and B, there are also composite 296 enclaves in andesites erupted during phase V, which comprise a vesicular basaltic core, 297 surrounded by more evolved magma (Plail et al., 2014).

298 Major element mixing (see Supplementary Material for calculations) between the phase V 299 andesite (MVO1538-b) and the mafic magma type A end-member (MT27) show that a high 300 percentage (67%) of andesite magma is required to achieve the most evolved type B mafic 301 enclave composition (MT25). This amount of mixing fits the distributions of most trace elements (figure 4, mixing lines), with a Σr^2 of 0.2, with the exception of Sm, Tb, Dy, Ho 302 and Er (Supplementary Material). The 2:1 bulk rock mixing ratio is also qualitatively 303 304 consistent with the observed high percentage of inherited phenocrysts (15–25 %) in the type 305 B enclaves, and undoubtedly also a proportion of rhyolitic melt from the andesite must have 306 been mixed into the mafic magma during phenocryst incorporation.

307 Systematic variations that may be explained by mixing are also observed in the glass 308 compositions in the enclaves, in particular in K₂O, FeO and TiO₂ content (Humphreys et al., 309 2010; Devine et al. 2014; Humphreys et al. 2015). Phase V type A enclave glasses contain high K₂O, TiO₂ and FeO rhyolitic concentrations compared to the type B enclave glass 310 compositions which overlap with the host andesite matrix glasses (Humphreys et al., 2015). 311 312 Textural observations indicate that, upon intrusion into the andesite, quench-crystallisation of the mafic magma occurred in situ involving an assemblage of plagioclase, ±amphibole, 313 314 ±clinopyroxene and magnetite. Using a simple assemblage of plagioclase 80 % and 315 amphibole 20 %, to represent roughly the appropriate modal proportions of a type A enclave diktytaxitic framework assemblage, a rhyolitic melt of ~76 wt% SiO₂ can be generated after 316 317 ~58 % crystallisation from the phase V mafic end-member composition (from 318 RhyoliteMELTS). Varying the modal proportions of the framework assemblage and 319 substituting clinopyroxene for amphibole, this amount of crystallisation would generate a range of melt compositions (\sim 70–80 wt% SiO₂) that fits the observed range in the enclaves. 320 321 Although this model is a good fit for SiO_2 it does not resolve the full range of compositional 322 differences in K₂O, TiO₂ and FeO between the enclaves. Instead, the overlap between the 323 enclave and host andesite matrix glasses compositions (Figure 2) may be evidence for the 324 mixing of heterogeneous melts during enclave formation at the interface between the two 325 magmas (Laumonier et al., 2014). The andesite matrix glass has also been shown to be highly 326 heterogeneous with respect to K₂O, consistent with mingling of melts, with the variable 327 diffusion kinetics of K and Ti producing a spread in concentrations (Humphreys et al., 2010).

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329 4.2. Temporal changes in composition and abundance of mafic enclaves through the330 eruption

There are clear changes in enclave geochemistry through the eruption. Displacements in Fe, Mg and V, Yb (and other MREE and HREE) concentrations are observed between enclaves erupted during the different phases (**Figures 2, 4, 5**). There was also a small increase in the andesite temperature between phases I and V by 10 °C, inferred from Fe-Ti oxide geothermometry, (Devine & Rutherford, 2014), consistent with heating by basaltic underplating below.

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We have established in the previous section that the colinear trends in the mafic enclaves are the result of mixing. Displacements in elemental composition for a fixed SiO_2 or La composition may be explained by changes in the composition of the end member basalt with 341 time. In order to explain the enrichment in MgO and V in the end member mafic magma 342 between phases I and III (figure 5), we invoke intrusion of slightly more primitive magma 343 with time. This change in end member magma composition between phases I and III is good 344 evidence for sustained and continuous replenishment of the underplating basaltic layer during 345 this time (up to 2008; figure 5). The presence of larger and more abundant enclaves in phase 346 V (Barclay et al., 2010; Komorowski et al., 2010) suggests perhaps that a boundary layer 347 between the magmas changed in dimension, thickening and cooling, which would be 348 consistent with the supply of mafic magma having ceased some time after phase III. We will 349 explore this idea further in the next section, where we develop an analytical model.

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351 *4.3.* The evolution of the interface between underplating mafic magma and andesite

352 The interaction between a cooler evolved andesite above a hot basaltic melt intrusion 353 involves heat transfer from the basalt to andesite. Initially the heat transfer is governed by 354 thermal conduction in both layers, with crystals in the partially crystalline andesite melting, 355 while the cooling basalt gradually crystallises. There is an abundance of evidence in the 356 andesite for heating and partial melting: sieve textures in the plagioclase (Humphreys et al., 357 2009b; Murphy et al., 2000), resorbed phenocrysts of orthopyroxene (with reversely zoned overgrowths or overgrowths of clinopyroxene) (Humphreys et al., 2009b; Murphy et al., 358 359 2000), quartz either strongly resorbed or jacketed by clinopyroxene crystals (Humphreys et 360 al., 2013; Murphy et al., 2000) and hornblende surrounded by thermal reaction rims 361 (Humphreys et al., 2009a; Rutherford and Devine, 2003) are all present. Owing to the 362 different composition and solidus temperatures of the basalt and andesite, the andesite near 363 the interface with the basalt may become fully molten if the basalt is sufficiently hot. In the 364 diffusive boundary layers, the bulk density of the magmas gradually changes. In particular, 365 the bulk density of the andesite typically decreases as the relatively dense mineral phases 366 melt, while the bulk density of the basalt initially increases on cooling and crystallisation. 367 However, if the basalt becomes vapor-saturated then on continued cooling, the basalt will 368 exsolve volatiles, leading to a gradual decrease in the bulk density. Eventually, the basalt may 369 become less dense than the overlying andesite and small plumes of volatile-laden basalt may 370 rise into the andesite. Since the andesite in the thermal boundary layer is hotter than the 371 overlying layer, this layer will become much less viscous through both reduction in the 372 crystal content and also heating of the melt. As a result, the plume rise within the boundary 373 layer will occur more rapidly than the mixing into the overlying cooler layer of andesite, so 374 that the basalt and andesite intermingle only on the scale of the boundary layer.

375 Subsequently, as the thermal boundary layer becomes deeper, this buoyant intermingled layer 376 may rise up into the main body of andesite, forming hybrid mafic enclaves consisting of a mixture of basalt and the heated andesite from the boundary layer, just as is observed in the 377 378 geochemical data (figures 2-4).

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380 Although the process is a continuum, we present a series of calculations in which we model the temperature evolution of the boundary layer in the limit that thermal conduction governs 381 382 the heat exchange until the point at which plumes of buoyant basalt, bearing exsolved 383 volatiles, rise up into the heated boundary layer of andesite. We then estimate the thickness of 384 the bulk thermal boundary layer at the point that it can rise into the overlying main body of 385 andesite, which has much higher viscosity than the boundary layer. Although the calculations 386 are simplified they provide some insight into the possible process of intermingling within the 387 local boundary layer, which can occur more quickly than the rise of basaltic enclaves into the 388 main body of andesite, and which leads to the enclaves consisting of a mixture of remelted 389 andesite mixed with the basalt, as seen in the samples discussed here.

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We model the crystal fraction of the basalt and the andesite as following laws of the form 391 $X_s(T)$ and $X_b(T)$, so that the temperature above (z>0) and below (z<0) the interface (z=0) 392 393 is given by the thermal diffusion equation 394

$$\left[\rho C + \rho L \frac{dX_i}{dX_i}\right] \frac{\delta T}{\delta T} = k \frac{\delta^2 T}{\delta t}$$

$$395 \qquad \left[\rho C_p + \rho L \frac{dX_i}{dT}\right] \frac{\delta T}{\delta t} = k_i \frac{\delta^2 T}{\delta z^2} \tag{1}$$

396

where *i* denotes the basaltic (*b*) or and esitic (*s*) melt and k_i is the thermal conductivity of the 397 398 basalt or andesite. In modelling X_b and X_s we use the approximate linearised laws: 399

$$X_{b}(T) = 0.02(1200 - T)$$
400 and
$$X_{s}(T) = 0.035(972 - T)$$
(2)

401

402 obtained by linear approximation to calculations carried out using the program Melts (Ghiorso and Sack, 1995). Here $T_m = 972$ °C is the temperature at which the andesitic magma 403

404 is purely molten, with no crystal phase, while $T_b = 1200$ °C is the temperature at which the 405 basalt is purely molten. These linearised relations are useful since they lead to analytical 406 solutions for the thermal diffusion equation (1), as presented below.

407

In our calculations, we assume that the latent heat of crystallisation has value $L = 7.42 \times 10^5$ 408 J/kg, with $\rho = 2500 \text{ kg/m}^3$ and $C_p = 1000 \text{ J/K/kg}$. We assume the initial temperature of the 409 basalt is $T_b = 1200$ °C on injection into the magma chamber, while the andesite has initial 410 temperature $T_s = 835$ °C. Two pyroxene thermometry in mafic enclaves yields temperatures 411 of 1,074–1,196° C (n = 5) (Humphreys et al., 2009b). QUILF (Andersen et al., 1993) used in 412 single-pyroxene mode (Murphy et al., 2000) gave typical orthopyroxene phenocryst core 413 temperatures of 800–900° C (average 850° C, n = 77). Phenocryst rims are typically slightly 414 415 hotter (average 870° C, n = 61). Temperature estimates were also obtained using the 416 hornblende- plagioclase geothermometer of (Holland and Blundy, 1994). One phenocryst 417 pair gave a temperature of 844° C. The interface temperature, T_{i} between the basalt and the andesite is then given by the continuity of heat conduction across the interfaces. Using the 418 419 above values for the properties of the melt, we find that the interface temperature is predicted to have value $T_{I} \sim 1008 \text{ K} > T_{m}$ (see below), so that immediately above the interface the 420 421 andesite is purely molten. The thermal diffusivity of the melt layer of andesite has value: 422

423
$$\kappa_m = \frac{k}{\rho C_p}$$
(3)

424

In contrast, in the basalt and the cooler region of the andesite, in which there is a crystal
phase present, the effective thermal diffusivity accounts for the thermal buffering associated
with the latent heat, leading to the relation

428

429
$$\kappa_{i} = \frac{\left[\rho C_{p} + \rho L \frac{dX_{i}}{dT}\right]}{\rho C_{p}}$$
(4)

430

for the basalt (i = b) and the silicic (i = s) melt when partially crystalline. With these three regions of partially crystalline basalt in the region z < 0, the molten and site in the region 0 433 < z < a(t) and the partially molten and esite in the region z > a(t), then prior to the onset 434 of convection, we find the approximate diffusive temperature profile

435
$$T(z,T) = T_I + (T_b - T_I) erf\left[\frac{-z}{\sqrt{(\kappa_b t)}}\right]$$
 for $z < 0$ (5)

436
$$T(z,T) = T_{I} + (T_{m} - T_{I}) \frac{erf\left[\frac{z}{\sqrt{(\kappa_{m}t)}}\right]}{erf\left[\frac{a(t)}{\sqrt{(\kappa_{m}t)}}\right]} \text{ for } a(t) > z > 0$$
(6)

437

439

440 where the interface temperature T_{I} is given by continuity of heat flux across the 441 interface

442

443
$$T_{I} = \frac{T_{m} + T_{b}\beta erf(\lambda)}{1 + \beta erf(\lambda)}$$
 where $\beta = \left(\frac{\kappa_{m}}{\kappa_{b}}\right)^{1/2}$ and $\lambda = \frac{a(t)}{\kappa_{m}t^{1/2}}$ (8)

444

445 where the location of the pure melt front in the andesite, z = a(t), is given by the solution for 446 λ of the equation 447

448
$$\gamma (1 + \beta erf(\lambda)) exp(-\lambda^2 \gamma^2) (T_m - T_s) = exp(-\lambda^2) (1 - erf(\lambda \gamma)) (T_b - T_m)$$
 (9)

449

450 with

$$451 \qquad \gamma = \left(\kappa_m - \kappa_s\right)^{1/2} \tag{10}$$

452

In **figure 6A**, we show a calculation of the structure of the thermal boundary layers across the interface, calculated after 1 and 10 days. After about 1 day, the thermal boundary layer extends about 10-20 cm into the andesite and basalt, while after 10 days, the boundary layer has grown to a thickness of >60 cm.

457

458 In calculating the potential for convective overturn within this boundary layer, we note three 459 key effects. First, the andesite has a much reduced crystal fraction in the boundary layer leading to a reduction in the viscosity by a factor of 10-100, from values of order 10^5 to 10^6 460 Pa s, to 10^3 to 10^4 Pa s, while the viscosity of the basaltic layer increases from values of 10-461 100 Pa s to 10^3 to 10^4 Pa s in the cooled region of the boundary layer. Also as the basalt 462 463 crystallises, it will tend to become saturated in volatiles at a particular crystal content. As 464 volatiles are exsolved, the density of the basaltic melt gradually decreases. The decrease in density depends on the volatile content of the magma, but will typically be of order 10-150 465 kg/m³ (Phillips and Woods, 2002). The solubility of water in the melt can be approximated 466 by Henry's law, $n_A = sP^{\frac{1}{2}}$ and so with a crystal content X the mass of exsolved volatiles is 467 468 given by

469

470
$$n_e \sim n_0 - s P^{\frac{1}{2}} (1 - X)$$
 (11)

471

472 where n_0 is the total volatile content of the basalt. This leads to an effective bulk density of 473 the bubble-melt mixture given by

474

$$475 \qquad \rho = \left[\frac{n_e}{\rho_g} + \frac{1 - n_e}{\rho_m}\right] \tag{12}$$

476

where ρ_g and ρ_m are the gas and melt densities. With a total volatile mass fraction in the range $n_0 = 0.02 - 0.04$ and a crystal fraction in the range X = 0.2 - 0.4 the melt density is typically 10-200 kg/m³ lower than the andesite (**figure 6B**). These approximate calculations based on this simplified parameterised Henry's law with magma of total volatile content (mass fraction) 0.02 (green) and 0.04 (red) are consistent in magnitude with full numerical 482 calculations using Melts (Woods and Cowan, 2009). As this boundary layer of cooled basalt 483 becomes less dense than the overlying boundary layer of heated andesite, plumes of the 484 bubble-laden basalt will tend to rise into the boundary layer of andesite owing to a Rayleigh-485 Taylor type instability once the rate of growth of the instability exceeds the rate of growth of 486 the boundary layer (Thomas et al., 1993). For a boundary layer of thickness h(t), the growth 487 rate of the instability, σ , scales as

488

489
$$\sigma \approx \frac{0.2g\Delta\rho h}{\mu_{sb}} \left(\frac{\mu_{sb}}{\mu_{bb}}\right)^{\frac{1}{3}}$$
(13)

490

491 where μ_{sb} denotes the viscosity in the boundary layer of magma *i*, where i = s or *b*. This 492 exceeds the growth rate of the boundary layer, $(\frac{1}{h})\frac{dh}{dt}$, which is driven by thermal 493 conduction, $h \approx (\kappa_b t)^{\frac{1}{2}}$, when

494

$$495 \qquad \frac{0.5g\Delta\rho h^3}{\kappa_b \mu_{sb}} \left(\frac{\mu_{sb}}{\mu_{bb}}\right)^{1/3} > 1 \tag{14}$$

496

497 At this point, plumes of the cooled basalt will rise and mingle with the heated boundary layer 498 of partially molten andesite. In figure 6C, we present an estimate of the thickness of the 499 boundary layer at the point that this convective mingling is expected to become established 500 and dominate the heat conduction within the boundary layer. Using the values for the reduced viscosity of the heated andesite, and values of the buoyancy of the basalt ranging from 50-501 150 kg/m³ (figure 6C), we estimate that the size of unstable boundary layer and hence the 502 503 plumes of basalt which will mingle with the heated andesite are of order 3-6 cm, 504 corresponding to times of order 0.1-1 days. Subsequently, as the boundary layer continues to 505 deepen by heat conduction into the overlying andesite and the underlying hot basalt, the layer 506 of mixed basalt and heated andesite will rise up into the cooler overlying andesite, once the rate of ascent of buoyant plumes through this cooler andesite is comparable to the rate of 507 deepening of the boundary layer. Using comparable values for the buoyancy, but with the 508 original viscosity of the crystalline and esite, 10^6 to 10^7 Pas, we estimate that such plumes will 509 510 rise into the original andesite when the mixed layer has deepened to a scale of 10-25 cm, and

511 so will emerge from the boundary layer after the longer time of order 1-10 days. We therefore 512 envisage a two-stage process of convective intermingling within the boundary layer, followed 513 by the ascent of intermingled plumes from the boundary layer into the overlying andesite. We 514 also observe that if the upper layer of andesite is sufficiently crystalline, then the crystal-515 crystal contacts may lead to the development of a yield stress in the upper layer, and this will 516 tend to suppress the ascent of plumes of buoyant melt into the main upper body of andesite, 517 whereas in the heated boundary layer, the crystal content will fall to lower values, thereby 518 reducing or eliminating this yield stress in the boundary layer itself. This additional effect 519 will again tend to promote an initial phase of local mixing in the boundary layer, and 520 provided the yield stress in the upper layer is not too large, this could then be followed by the 521 ascent of the mixed plumes into the upper layer once the buoyancy force associated with the 522 mixed layer exceeds the yield stress associated with the main body of andesite. Estimates of 523 yield stress in a crystalline magma suggest that it depends on the crystal shape and crystal 524 mass fraction (Hoover et al., 2001; Saar et al., 2001), but to allow mafic inclusions of size 0.1 m and buoyancy 100 kg/m³ to rise through the melt, it should not exceed values of order 100 525 526 Pa, which is at the low end of the range. For larger values of yield stress, it may be that the 527 ascent of convective plumes into the upper layer would be restricted, further promoting the 528 mixing in the heated boundary layer. In that case, some limited intermingling of the hybrid 529 boundary layer into the overlying melt, and generation of mafic inclusions, might occur 530 during an eruptive phase, if the upper layer becomes mobilized by the larger scale stresses 531 driving the eruption. It is also relevant to note here that as bubbles are exsolved from the 532 basalt, the boundary layer in the basalt becomes less dense than the underlying basalt, and 533 hence stable to convective mixing into the main body of basalt (Cardoso and Woods, 1996).

534

535 Conclusions

536 Mafic enclaves erupted with crystal-rich andesite during the 1995-2011 eruption of Soufrière 537 Hills Volcano, Montserrat, display compositions and geochemical trends consistent with their 538 derivation at the interface between a basalt layer and an overlying andesite layer (figure 7). 539 The linear trends in major and trace element geochemistry of the mafic enclaves suggest that 540 they are formed by mixing between an end member tholeiitic basalt and the overlying 541 andesite, with the end member basalt changing in composition over time through the 542 eruption: becoming more primitive during phases I and III, then less primitive towards the 543 end of the eruption in phase V (figure 5). We propose that underplating vapor-saturated 544 mafic magma cools at the interface, and mixing with semi-molten andesite occurs in the

545 interface zone. The interface zone then becomes buoyant during cooling, mixing and 546 vesiculation and breaks off to form plumes, which rise into the overlying andesite. The mafic 547 enclaves become more primitive during phases I to III, which might be interpreted as being due to quasi-continuous intrusion of increasingly primitive basalt from depth; it is certainly 548 549 not consistent with the continuous evolution of a basalt layer emplaced at the beginning of 550 the eruption which then evolves by cooling and crystallisation. After phase III, mafic enclaves become more hybridised and larger, consistent with the cessation of intrusion of 551 552 mafic magma from depth, allowing the thermal diffusion front to migrate further into the 553 basalt, generating a thicker mixed boundary layer from which the enclaves form. It is worthy 554 of note that, from the analytical model we develop here, the timescales required to generate a 555 boundary layer of thickness similar to the dimension of mafic enclaves (decimetres) is on the 556 order of 1-10 days, which is similar to the timescales derived from diffusion profiles in 557 reverse-zoned phenocrysts and in Fe-Ti oxides both at Soufrière Hills (Devine et al., 1998) 558 and globally (Cooper and Kent, 2014). Quasi-continuous magma intrusion during 1995-2008 559 is consistent with continued high fluxes of SO₂ from Soufrière Hills (Edmonds et al., 2010) 560 and inflation of the volcano between eruptive phases (Elsworth et al., 2008).

561

562 Our analysis demonstrates that trends in mafic enclave geochemistry through an eruption 563 might yield insights into basalt underplating and its role in eruption triggering. The apparent 564 cessation of the eruption after phase V suggests that mafic magma intrusion may have played 565 a critical role in sustaining the eruption by heating and remobilising the overlying andesite 566 (Couch et al., 2001) and furthermore, this process and mechanism may be important at many 567 other arc volcanoes globally.

568

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- 762
- 763

764 Table and figures

765

Phase I	Phase II	Phase III	Phase IV	Phase V
Nov. 95 –	Nov. 99 –	Aug. 05 –	Jul. 08 –	Oct. 09 –
Mar. 98	Jul. 03	Apr. 07	Jan. 09	Feb. 10

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Table 1: Soufrière Hills Volcano (Montserrat) eruption phases (Wadge et al., 2014).

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Figure 1: A: Map of Montserrat showing the location of the Soufrière Hills Volcano. B:

photomicrograph of a mafic inclusion, showing a typical diktytaxitic texture made up of

blocky plagioclase and dark brown amphibole crystals. Light brown glass surrounds the crystals. C: photomicrograph of the interface between crystal-rich andesite (top) and mafic inclusion (bottom). Orthopyroxene and magnetite are visible in the mafic enclave. The andesite has a highly crystalline groundmass. D: an image of a typical andesitic block on outcrop scale with mafic enclaves from phase V block and ash deposits in the Belham Valley, Montserrat.

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778 Figure 2: Major element compositions of whole rock lavas (solid circles) and glasses 779 (crosses) from the Soufrière Hills eruption, phases I to V (see legend) (Barclay et al., 2010; 780 Mann et al., 2013; Murphy et al., 2000; Plail et al., 2014; Zellmer, 2003). Also shown are 781 lava compositions from South Soufrière Hills (Cassidy et al., 2015). A: MgO versus SiO₂, in 782 wt%; B: Al₂O₃ versus SiO₂, in wt%; C: CaO versus SiO₂, in wt%; D: K₂O versus SiO₂, in wt% and E: FeO_{tot}/MgO versus SiO₂, in wt%. Tholeiite (TH) and calc-alkaline (CA) fields 783 are marked. Fractional crystallisation trends from Rhyolite Melts (Ghiorso and Gualda, 2015) 784 785 are shown.

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Figure 3: Trace element plot to show lava compositions from phases 1 to V of the Soufrière
Hills Volcano eruption. Data from this work and from Zellmer (2003). Trace element
compositions normalised to primitive mantle *(Sun and McDonough, 1989)*.

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791 Figure 4: Trace element compositions of whole rock lavas from the Soufrière Hills eruption, 792 phases I to V (see legend) from this work and Zellmer [2003]. A: Dy versus La, in ppm; B: 793 Yb versus La, in ppm; C: V versus La, in ppm; D: Sc versus La, in ppm; E: Sr versus La, in 794 ppm. Vectors to show the effect of 20% fractionation of various crystal phases are shown (for 795 method and partition coefficients used see Supplementary Material). Mixing lines are 796 shown on the V vs La plot, showing the amount of mixing required to form the array of mafic 797 enclaves for mixing between an end member basalt and the most evolved andesite bulk 798 composition. To generate the most Si-rich mafic enclave erupted in phase V requires mixing 799 67% andesite and 33% basalt; fits and residuals for each element are given in 800 Supplementary Table 3.

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Figure 5: Box and whisker plots to show Mg/Fe and V/La over the five phases of the eruption. The box is bounded by the 75th and 25th quartile values, with a horizontal line through the box denoting the median. Whiskers show the minimum and maximum values. Mafic enclave compositions are shown in blue; andesite in red. Below is a timeline to show when eruptive phases occurred and interpretative labelling (see main text discussion for justification).

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Figure 6: A model to describe the interaction between basalt magma and overlying andesite. A: Thermal profile across the boundary between basalt and andesite after 1 and 10 days; B: how the buoyancy (density difference between the boundary layer and the overlying andesite) varies during cooling and crystallisation, for two water contents (2 and 4 wt%); C: the varation of the scale of the plume rising from the boundary layer with viscosity of the boundary layer magma, for various boundary layer buoyancy values. A more buoyant boundary layer is expected to form smaller mafic enclaves.

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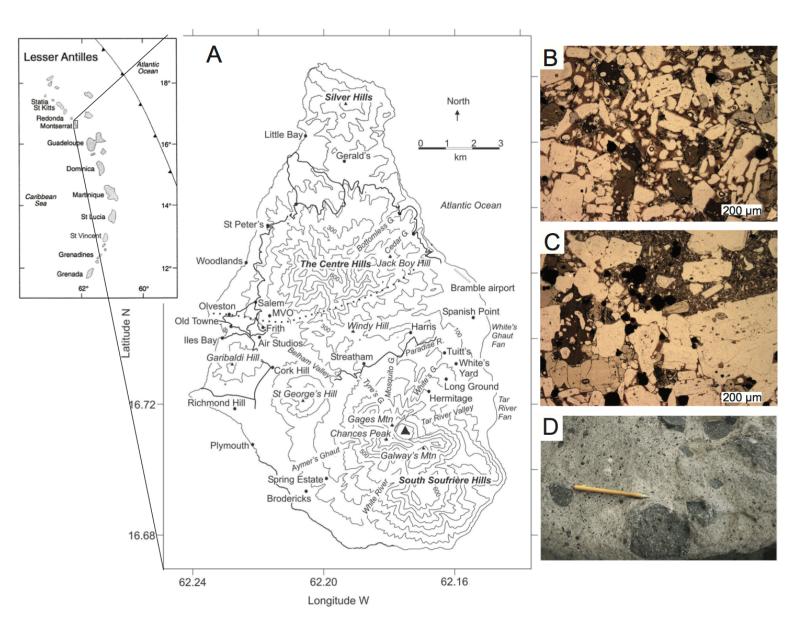
817 Figure 7: Schematic diagram to illustrate the controls on mafic enclave composition and how it may evolve with time. Left: vapor-saturated mafic magma (basalt) underplates a reservoir 818 819 of crystal-rich andesite containing a range of crystal phases (pl: plagioclase; opx: 820 orthopyroxene; am: amphibole). An initially thin hybrid boundary layer forms as the basalt 821 quench-cools and crystallises while the andesite partially melts, allowing them to mix in this 822 layer. Middle: When the buoyancy of the layer is sufficient buoyant plumes form at the 823 interface, which rise into the heated lower viscosity layer of andesite and form mafic 824 enclaves, which may be stirred into the heated andesite and which have a dimension similar 825 to the thickness of the boundary layer. Quenched-rim basaltic mafic inclusions are among the 826 first to break off from the interface. Composite enclaves may form from hybrid material 827 surrounded by more mafic material as the latter rises up in the wake of boundary layer break-828 up. Initially the proportion of mafic material is small (~1 vol% (Barclay et al., 2010)). Right: 829 if the intrusion of mafic magma is shut off and the basalt cools, the boundary layer will grow 830 by thermal diffusion, the mafic enclaves will become larger, and there will be a higher 831 volume proportion of mafic enclaves in the erupted lavas, as has been observed (Barclay et 832 al., 2010).

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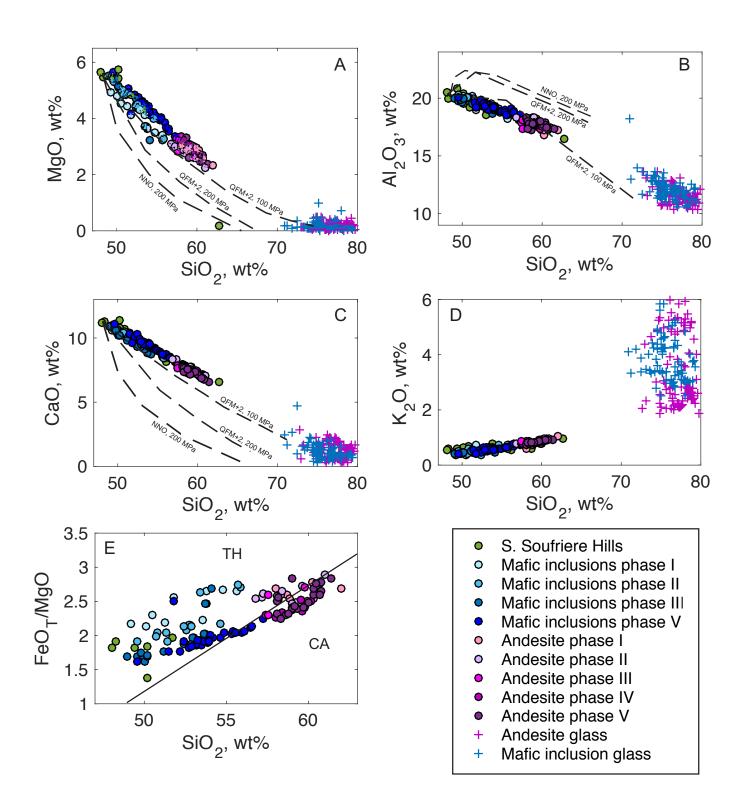
Phase I	Phase II	Phase III	Phase IV	Phase V
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Table 1: Soufrière Hills Volcano (Montserrat) eruption phases (Wadge et al., 2014).









Element/Primitive Mantle

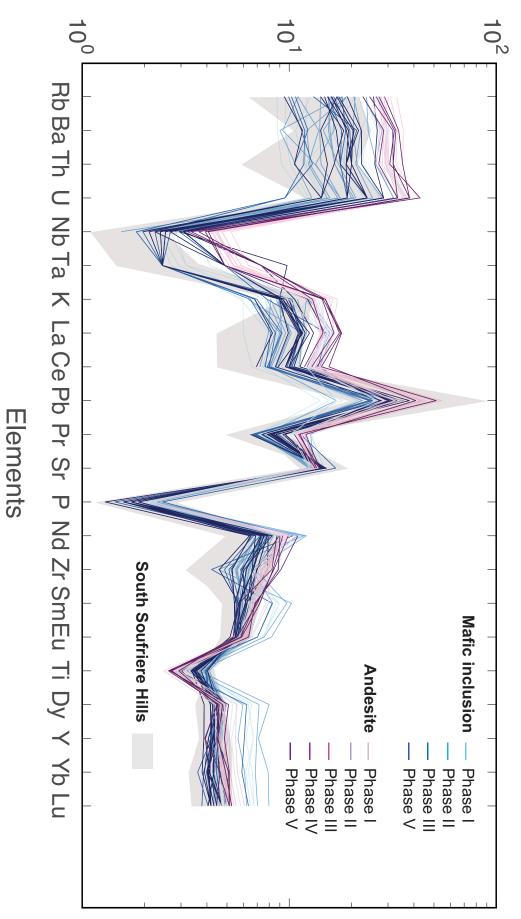


Figure 4

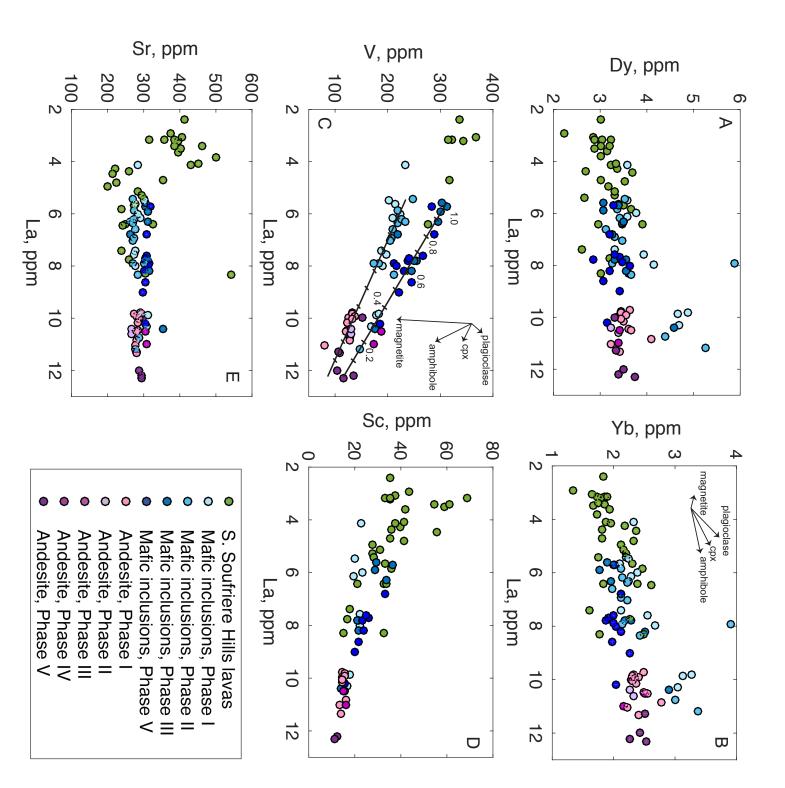


Figure 5

