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1	Origin of basalts by hybridisation in andesite-dominated arcs
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25 ABSTRACT

26 Mafic magmas are common in subduction zone settings, yet their high density restricts their ascent to the surface. Once stalled in the crust, these magmas may differentiate, 27 28 assimilate crust and other melts and mushes to produce hybridised intermediate magmas. The Soufriere Hills Volcano on Montserrat is a 'type locality' for these hybridisation processes 29 and yet, just 3 km south of the crater, voluminous basalts have erupted from the South 30 Soufriere Hills volcano within the same time period as the Soufriere Hills Volcano was 31 erupting hybrid andesites (131 - 128 ka). Basaltic South Soufriere Hills magmas have 48 - 53 32 wt% SiO₂ and 4 - 6 wt% MgO. They were hot (970 - 1160 °C), volatile-rich (melt inclusions 33 contain up to 6.2 wt% H_2O) and were stored at 8 – 13 km prior to eruption (based on olivine 34 and pyroxene-hosted melt inclusion volatile geochemistry). Melt inclusions do not preserve 35 basaltic liquids: they are andesitic to rhyolitic in composition, related to one another by a line 36 37 of descent controlled by simple closed-system fractionation. Whole rock compositions, however, are best described by a hybridisation model involving "back"-mixing of andesitic to 38 39 rhyolitic melts with mafic crystal phases such as magnetite, olivine, orthopyroxene and 40 clinopyroxene. Phenocryst zoning illustrates repeated mixing events between evolved melts and mafic phenocrysts, which, when coupled with the heterogeneity of crystal compositions, 41 strongly suggests that although the bulk composition is basalt (containing Fo₈₀ olivine), they 42 were assembled from disparate ingredients, likely derived from mafic crystal mushes and 43 more evolved melt lenses of variable composition. The mixing events occur days to weeks 44 prior to eruption. We propose that the South Soufriere Hills basaltic magmas, with their 45 higher bulk density over andesites from neighbouring volcanoes, ultimately may have been 46 eruptible owing to both the transtensional tectonics imposed by offshore grabens (related to 47 the oblique subduction of the Lesser Antilles) and to surface unloading caused by large scale 48 edifice collapse. Our observations support the idea that compositional changes in arcs might 49

reflect not only changes in source compositions, but also effects caused by patterns in crustalstrain and tectonics.

52 **INTRODUCTION**

Intermediate magmas are generated by intensive crustal magmatic processing 53 involving crystallisation, assimilation and mixing (Anderson, 1976; Eichelberger, 1978; 54 Rudnick, 1995; Eichelberger et al. 2006; Reubi and Blundy, 2009; Kent et al., 2010; 55 Melekhova et al., 2013). Mafic magmas are implicated in these processes through recharging 56 of magma bodies by mingling at the interface and by large-scale overturn in magma 57 reservoirs (Pallister et al., 1992; Bateman, 1995). These processes are well-illustrated by 58 volcanoes in the Lesser Antilles arc where andesitic lavas containing mafic enclaves are 59 commonly erupted. Andesites may erupt preferentially due to their relatively low density 60 compared to the denser mafic lavas that are "trapped" at depth by a density filter mechanism 61 62 (Plank and Langmuir, 1988). Rheological and lithological barriers may also inhibit the propagation of a basaltic melt (Eichelberger, 1978; Dufek and Begantz, 2005; Karlstrom et 63 64 al., 2009; Kent et al., 2010). Indeed, intermediate to rhyolitic magma reservoirs can obstruct 65 the passage of mafic magma, explaining why basaltic eruptions often only reach the surface on the periphery of silicic volcanoes (Hildreth, 1981). An interesting variant on this process is 66 illustrated on Montserrat, where basalts were erupted from the South Soufrière Hills (SSH) 67 volcano over the same broad time interval as crystal-rich andesites (with rhyolitic melts) were 68 being erupted from the Soufrière Hills Volcano (SHV) located less than 3 km away. This 69 raises the question as to what mechanisms allow eruption of felsic and mafic volcanic rocks 70 in such close proximity. 71

More detailed study of the SSH is also of interest because, while there is strong evidence that andesites are generated largely by mixing of repeated injections of mafic magma into high level silicic magma chambers (Anderson, 1976; Eichelberger, 1978; 75 Eichelberger et al. 2006; Reubi and Blundy, 2009; Kent et al., 2010), the petrogenesis and 76 history of the mafic magmas is not well understood and may itself be complex. The density filter trap (Plank and Langmuir, 1988) means that mafic enclaves from SHV are the only 77 78 evidence of deeper, mafic magmas that are available for petrologic analysis and in many cases these mafic inclusions have experienced varying degrees of intrusion, quenching and 79 degassing that obscures their earlier characteristics. Thus, a study of closely spaced (in 80 distance and time) andesitic and basaltic volcanism at SHV and SSH has the potential to 81 reveal more detail regarding the nature of basaltic magmas resident in the mid- to upper-82 83 crust, and can provide insights into the relative importance of magma mixing and fractionation in controlling the composition of all arc volcanic rocks, and how this relates to 84 processes of magma storage, hybridisation, eruption triggering and growth of the arc crust. 85

86 In this paper we present new whole rock and melt inclusion analyses of basaltic to 87 andesitic lavas erupted from the SHV. We compare their geochemical characteristics to the andesites erupted from SHV and examine the geochemistry of individual phenocrysts phases 88 89 to characterise compositional gradients related to normal crystal growth during cooling and also due to mixing. We assess whether their compositions could have been generated by 90 simple processes of fractional crystallisation alone or whether mixing between disparate 91 liquid and mush components is necessary. Mineral melt thermometry has been used (from 92 two-pyroxenes and plagioclase-glass pairs) and barometry (using H₂O-CO₂ systematics of the 93 94 melt inclusions) to estimate pre-eruptive storage conditions. We use the relaxed compositional steps across olivine crystals to infer pre-eruptive mixing timescales between 95 felsic liquids and mafic crystals. Using all of the available petrological and geochemical data 96 97 we develop a model for the generation of hybrid basalts on Montserrat and how they are assembled and speculate as to the possible reasons for extraction and eruption of higher 98 99 density hybrid magmas relating to tectonics and unloading.

100

101 Geological background

The Lesser Antilles, like many arcs, comprises predominantly andesitic volcanic 102 103 islands with relatively few basaltic centres. For example, in the northern and central islands (Saba to St. Lucia) <10% of the erupted volcanic rocks are basaltic. Where basaltic rocks are 104 present, they generally occur as small-volume centres adjacent to much larger andesitic 105 volcanoes (Westercamp and Mervoyer, 1976; Rea and Baker, 1980; Macdonald et al. 2000). 106 This is exemplified on Montserrat (Fig. 1), where andesite lavas are predominant (Rea, 107 108 1974), with a single isolated basaltic centre (SSH) in the southernmost part of the island. Apart from the SSH, basalt occurrences are restricted to mafic inclusions within andesites. 109 There is abundant petrological evidence (particularly from the currently active SHV) to show 110 that the erupted andesites are hybrids formed over long timescales (10^3 to 10^4 years) by 111 multiple recharges of deeply-sourced mafic magmas into large reservoirs of crystal-rich 112 andesite magmas prior to ascent to the surface (Murphy et al., 2000; Humphreys et al., 2009; 113 Plail et al., 2014). 114

The SSH basalts are, however, sufficiently geochemically distinct from the SHV 115 basaltic enclaves suggesting that they reflect different magma sources and processes, such as 116 increased relative contribution from slab fluids over subducted sediments (Zellmer et al., 117 2003; Cassidy et al., 2012; 2014), and thus provide information regarding magmas forming 118 119 within the arc that are not generally observed, at least in an identifiable form, at the surface. Indeed, the SSH volcanic rocks represent some of the most mafic lavas in the northern Lesser 120 Antilles arc (47 wt% SiO₂; 6 wt% MgO), with the exception of the high-Mg basalts in 121 Martinique (Westercamp and Mervoyer, 1976). These geochemical differences are not 122 simply related to temporal evolution of the volcanism on Montserrat, because Ar-Ar dating 123 and stratigraphic relationships clearly indicate that the SSH and the SHV were both active in 124

the interval 130±5 ka (with SHV-type rocks forming the basal unit to the main SSH
lithologies), and the predominant andesitic volcanic rocks of the island were emplaced before
and after eruption of the SSH (Harford et al., 2002; Cassidy et al., 2012).

The island of Montserrat is located in the northern part of the Lesser Antilles; a 750 128 km long chain of volcanic islands formed as a result of the slow (2 cm yr⁻¹) subduction of the 129 North American plate beneath the Caribbean plate (Fig. 1) (Wadge, 1984; DeMets et al., 130 2000). The oblique nature of this subduction means that the northern part of the arc is 131 influenced by transtensional forces that have led to intra-plate deformation (Feuillet, 2000; 132 133 Feuillet et al., 2010). Montserrat lies on crust ~30 km thick that sits on an asthenospheric mantle wedge that extends to ~130 km in depth (Wadge and Shepherd, 1984). The island 134 comprises four volcanic centres: Silver Hills (2600-1200 ka), Centre Hills (950-550 ka), SHV 135 136 (282 ka to present) and SSH (131-128 ka) (Harford et al., 2002). All these volcanic centres (except for the mafic-dominated SSH) are andesitic in composition, but their erupted 137 products all contain abundant inclusions of mafic magma (Rea, 1974; Murphy et al., 2000; 138 Zellmer et al., 2003; Barclay et al., 2010; Plail et al., 2014). 139

The SHV centre has been studied in most detail and is comprised of phenocrysts of 140 orthopyroxene, plagioclase and amphibole in a rhyolitic glass, with clear evidence for magma 141 mixing and mingling (Murphy et al., 2000; Humphreys et al., 2009; Humphreys et al., 2013). 142 Under-plating of the crystal-rich andesite by wet mafic magma causes instabilities to form at 143 144 the interface, forming enclaves (Plail et al., 2014; Edmonds et al., 2014), interspersed with sporadic magma overturn events that thoroughly mix the magmas (Woods and Cowan, 2009), 145 distributing widely dispersed mafic components (Fe-rich plagioclase microlites, K-rich glass; 146 Humphreys et al., 2010) into the andesite body. The petrography and geochemistry of the 147 mafic enclaves of the SHV is also best explained by a mixing process between a mafic end 148 member (which varies in composition with time owing to lower crustal cryptic amphibole 149

fractionation) and variable amounts of rhyolitic melt hosting up to 20 vol% phenocrysts ofplagioclase, amphibole and magnetite, although not in bulk rock proportions.

While there have been a large number of studies on the andesites of Montserrat, 152 petrological work on the SSH basalts is more limited. Murphy et al. (2000) report that the 153 mineral assemblage consists of plagioclase, olivine, clinopyroxene, and titanomagnetite. The 154 SSH exposures comprise a range of rock suites from lava flows, to scoria, to reworked 155 volcaniclastic material (Cassidy et al., 2014), with some more mafic enclaves and some lava 156 flows containing cumulate xenoliths of orthopyroxene and plagioclase, similar to those 157 158 described by Kiddle et al. (2010). The SSH exposures can be divided into two units on the basis of their distinct trace element and isotopic compositions: SSH Suite A has lower Sr/La 159 and Sm/Zr ratios, but higher Zr/Er ratios and more radiogenic Pb isotope compositions than 160 161 Suite B (Cassidy et al., 2014)

162

163 METHODS

164 *Samples*

Samples of SSH rocks were collected along the south coast of Montserrat (Fig. 1; 165 Table 1). Splits were crushed using an agate Mortar and powdered for whole rock analysis 166 and thin sections were also cut for electron microprobe (EMPA) and scanning electron 167 microscope (SEM) analysis. Fractions of samples were crushed coarsely and crystals of 168 169 enstatite, augite and olivine were picked from the 125-250 µm grain size fraction. The crystals were ground and polished to expose melt inclusions and mounted in indium for 170 secondary ion mass spectrometry (SIMS) analysis. All the inclusions analysed were natural 171 172 quenched, 40-200 µm in size and were not necked or breached by cracks.

173

174 Whole rock analysis

Major elements were analysed by X-ray Fluorescence (XRF) analysis of glass beads
prepared by fusion of a mixture of 0.5 g subsamples and lithium tetraborate in a ratio of 1:10.
Analyses were undertaken using a Philips Magix Pro WD-XRF at the National Oceanography
Centre (NOC), Southampton, UK. Error and external accuracy was generally <2%.

179

180 Microanalysis (EMPA, SEM and SIMS)

Concentrations of H₂O and CO₂ in glass were obtained by SIMS on a Cameca IMF 4f 181 ion microprobe at the NERC microanalytical facility at the University of Edinburgh, using a 182 15kV primary beam of O⁻ ions (Hauri et al., 2002; Blundy and Cashman, 2008). Positive 183 secondary ions were accelerated to 4500 eV, with an offset of -75eV (for ¹H and trace 184 elements) and -50eV (for 12 C) (\pm 20eV) to reduce transfer of molecular ions. A 50 µm raster 185 was performed for three minutes prior to the start of each analysis, and a primary beam 186 current of 5-6 nA used with a non-rastered, oval-shaped beam covering a 15-20 µm area on 187 188 single spots within the boundaries of the melt inclusions. Peak positions were verified before each analysis. The following elements were analysed by counting for 3 s in each of a 10 189 cycle run: ¹H, ²⁵Mg, ³⁰Si. These counts were then normalised to ³⁰Si and converted to 190 191 concentrations using a calibration curve populated by glass standards. The relative ion yield for H correlates with SiO₂ content, such that plotting ${}^{1}H/{}^{30}Si$ versus H₂O yields a single 192 working curve for glasses of variable SiO₂ content. CO₂ concentrations, however, require a 193 correction for SiO₂ content. 194

195 Carbon was measured independently of ¹H, using the same beam conditions, but with 196 a 50 μ m image field to improve transmission at moderate mass resolution, which was 197 sufficient to resolve ²⁴Mg²⁺ at the ¹²C peak position for background olivine measurements 198 and inclusion analyses. ¹²C was analysed for 3 s in each of 20 cycle runs in which ²⁴Mg²⁺, 199 ²⁸Si²⁺ and ³⁰Si were also measured. During data processing, the first 5 cycles of the ¹H

analyses and the first 10 cycles of the ¹²C data were discarded to avoid the effects of surface 200 contamination on the samples which may have survived the cleaning process. Instrumental 201 backgrounds were minimized by allowing samples held in epoxy to outgas in a separate 202 203 vacuum for at least ten hours prior to use in the SIMS instrument. The full list of glass standards used is shown in suppl. Table 1. The accuracy and precision were monitored 204 throughout the sessions by repeat analysis of the standards as unknowns: for H₂O analyses 205 these were <9% and <6% respectively; and for CO₂ <11% and <8% respectively. The 206 average CO₂ and H₂O backgrounds over seven sessions were 56 ppm and 0.03 wt% 207 respectively. There is lack of variation between Al2O3 and MgO in melt inclusions 208 compositions, suggests that they do not follow the vectors anticipated for post-entrapment 209 210 crystallisation of the host mineral.

211 The major element and volatile (S, Cl and F) compositions of the glasses, inclusions and phenocrysts were determined using the Cameca SX100 electron microprobe at the 212 University of Cambridge. Quantitative determinations of elements were made using the 213 wavelength dispersive system with TAP, PET and LIF crystals. A range of metal, oxide and 214 silicate (e.g. jadeite, wollastonite) standards was used for calibration of the spectrometers. 215 All analyses used an accelerating voltage of 15kV. For olivine, pyroxene and plagioclase a 216 spot size of 4 µm and a 100 nA beam current was used. For glasses, a 10 µm spot was used 217 with a beam current of 60 nA for Cl, F, S, P, Cr and Ni, and 4 nA for all other elements, with 218 219 counting times of 50-200 s per analysis. During glass measurements, Na peaks were counted first to avoid significant migration during the run. In addition to calibration of each X-ray 220 line, a series of secondary reference standards (olivines, pyroxenes, feldspars and glasses) 221 were measured daily to check accuracy, precision and totals. Standards used were periclase 222 for Mg, jadeite for Na, fused Si for Si, rutile for Ti, fayalite for Fe, K-feldspar for K 223 corundum for Al, apatite for P, and pure metals for Cr and Mn. Repeat analyses of standards 224

were used to estimate the precision of An, Mg# and Fo measurements. Forsterite content of the St. John's Island Olivine standard was determined with a precision of $2\sigma=0.46$ mol % (n=33). Precision of Mg# of clinopyroxene was similar to the precision of forsterite content in olivine. Anorthite content in the Anorthite55 standard was determined with a precision of $2\sigma=1.01$ mol % (n=46).

Accuracy was generally better than 5% for most elements, based on repeat analyses of EMPA secondary standard 2390-5 and by comparison with reference concentrations for the standard, with the exception of TiO₂, K₂O, P₂O₅ and Cl, which were better than 20-35 %. Detection limits for S, Cl and F were 40, 38 and 170 ppm, respectively, and precision was typically < 5% for all oxides, with the exception of MnO, P₂O₅ and F, which was better than 20%.

Backscattered SEM images were taken at the NOC, using a LEO 1450VP (variable pressure) SEM. Carbon-coated samples were imaged at 15 kV, a working distance of 10 mm and a nominal probe current of 50–500 pA, using both secondary electron (SE) and backscattered electron (BSE) detectors.

240

241 **RESULTS**

The whole rock samples are black to grey in colour, and poorly to moderately vesicular (6-38%, average 20%). The SSH samples have bulk rock compositions ranging from basalt to andesite (47-58% SiO₂) (Table 1; Fig. 2). Also shown in Figure 2 are the compositions of andesites and mafic enclaves erupted from the SHV during 1995-2010, together with previously published data from SSH (Murphy et al., 1998, 2000; Horwell et al., 2001; Zellmer et al., 2003; Humphreys et al., 2009, 2010; Cassidy et al., 2012). Relative to the SSH, the SHV volcanic rocks are more silicic, ranging from basaltic andesite to dacite (53-68% SiO₂), but the SHV contain mafic enclaves that range from basaltic to basaltic
andesite (49-55% SiO₂).

The SSH lavas are highly crystalline, with 31-53 vol.% phenocrysts and microphenocrysts (>100 \Box m) and 47-69% microlites. Plagioclase is the most abundant crystal phase (up to 61 vol.% of the crystal assemblage), followed by orthopyroxene (15 vol.%), olivine (11 vol.%) and clinopyroxene (10 vol.%), with titanomagnetite and rare amphibole in the basaltic andesite samples (SSH5B) comprising the remaining 3 vol.% (Fig. 3). The microlite crystal size fraction comprises a similar assemblage, however with less olivine present.

258 *Olivine petrography*

259 On average, olivines form the largest crystals (mean size 390 \Box m; range ±100 \Box m) 260 and are often euhedral to subhedral. They are commonly fractured and slightly altered (slightly reddened along cracks, visible in plane polarised light). The forsterite contents 261 (molar Fo% = Mg/(Mg+Fe) x 100) range from 56 to 80 mol. % (Figs. 4 and 5), with two 262 263 main peaks in olivine core compositions (Fo72-80 in Group 1; Fo56-68 in Group 2) and two peaks in olivine rim compositions that are slightly less forsteritic than the cores. Most of the 264 olivines are normally zoned or unzoned, but some exhibit reverse zoning (Fig. 6; Fig. 7), 265 suggesting multiple magma bodies which have experienced mixing. The reverse-zoned 266 olivines have core compositions of Fo71-80, compared to Fo56-80 in the normally-zoned 267 268 olivines (Figs. 5 and 6c). There is a negative correlation between olivine forsterite contents and CaO and MnO concentrations, with generally higher Fo% and lower Ca and Mn contents 269 in the cores (Fig. 5). These correlations are significant at >95% confidence, with P-values 270 <0.05. This correlation is especially strong between MnO and Fo% with a R^2 value of 0.9, 271 but this correlation is less apparent with CaO and Fo% (R^2 of 0.36). The normally-zoned 272 crystals show a trend of increasing Fo% from rim to core, mirrored by decreasing CaO and 273

274 MnO profiles (Figs. 6a and 6b). Figure 6d illustrates an olivine with reverse zoning towards 275 the outer edge of the crystal, with a thin ($<20 \ \Box m$) band of normal zoning at the rim and no 276 visible overgrowth. The core of this crystal has a constant forsterite composition of Fo₇₂, 277 except for the outer 50 $\Box m$. The increase in forsterite content in the reverse zone is 278 positively correlated with CaO, but negatively correlated with MnO content.

279 *Plagioclase petrography*

Plagioclase crystals range in size from microlites (<15 \[]m) to phenocrysts (>500 280 \Box m), with the latter commonly showing both normal and oscillatory zoning, as well as sieve 281 282 textures (Fig. 3). Anorthite contents (An mol.% = $Ca/(Ca+Na) \times 100$) range from 49-97% The feldspars are commonly normally-zoned, but with rare reverse-zoned (Fig. 4). 283 phenocrysts also present (Fig. 7), suggesting a complex set of magmatic processes have 284 285 occurred. The plagioclase crystals can be separated into two main groups based on their 286 anorthite compositions. The cores and reverse-zoned rims are anorthitic (An₇₉₋₉₇), while the rims of the normally-zoned plagioclase are more albitic (An_{52-70}) and are generally richer in 287 MgO, FeO and TiO₂ than the more anorthitic cores and rims (Fig.8). Complex dissolution 288 and resorption is also seen in some crystals (Fig. 8b). 289

290 *Pyroxene petrography*

The average size of the orthopyroxene crystals is $142 \ \Box m \pm 100 \ \Box m$. They are commonly zoned and often occur as overgrowths on olivine (e.g. Fig. 6c). Magnesium number (Mg# = Mg/(Mg+Fe) x 100) ranges from 60-74 (Figs. 4 and 9), and all are enstatite in composition. Enstatite TiO₂ and Al₂O₃ contents generally decrease with decreasing Mg# (Fig. 9), but do not correlate significantly with Al/Ti ratios. The enstatite shows common reverse zoning and some normal zoning, but rare unzoned crystals are also present (Fig. 7).

297 Clinopyroxenes have an average crystal size of $176 \square m \pm 100 \square m$, with Mg# 298 ranging from 58-80. The majority of the clinopyroxenes are augite, but some cores are diopside . The augites are commonly zoned, but rare unzoned crystals also exist. Some of the clinopyroxene occurs as pigeonite overgrowths on the olivines (Figs. 6 and 10). Plots of Mg# versus minor elements (Fig. 9) show that the clinopyroxenes contain higher concentrations of TiO₂, Al₂O₃ and Al/Ti ratios than the enstatites. A traverse of a normallyzoned crystal shows complex saw tooth zoning (Fig. 10b) that is particularly oscillatory in the last 70 \square m toward the rim, which occurs along with a sharp increase in Al₂O₃ and TiO₂ and a decrease in both Mg# and Al/Ti ratios.

306 *Melt inclusion geochemistry*

307 The melt inclusions are pristine, up to 90 \Box m in diameter, with no vapour bubbles and no daughter crystal phases. They span a range in compositions from andesitic to rhyolitic, 308 309 with 58.2-72.6 wt.% SiO₂, 0.45-2.6 wt.% K₂O and 0.01-2.8 wt.% MgO (Fig. 2; Table 2). 310 Their H₂O contents range from 1.50-6.19 wt.%, with CO₂ contents of 20-313 ppm (Fig. 11). CO₂ and S concentrations decrease with increasing melt SiO₂ contents, ranging from 395 311 ppm S and 313 ppm CO₂ at 58.2 wt.% SiO₂, to 18 ppm S and 20 ppm CO₂ at 72.6 wt.% SiO₂. 312 Cl shows a positive relationship with SiO₂, ranging from 2500 ppm at 58.2 wt.% SiO₂ to 313 3610 ppm at 72.6 wt.% SiO₂. 314

315

316 **DISCUSSION**

The range of compositions and textures in mineral, whole-rock and melt inclusion chemistry suggests that the SSH mafic magma petrogenesis was just as complex as that observed for the SHV andesitic volcanic system on Montserrat and involved the assembly of multiple components. Here we discuss the origin of these components by considering the pressure-temperature conditions of magma storage, fractional crystallization and magma mixing that are reflected in the crystal and melt phases in the SSH erupted products, as well as the conditions required for the eruption of these products at the surface. 325 Temperature estimates of the magma reservoir conditions are derived from the twopyroxene thermometry and plagioclase-whole rock equilibria after applying the equilibrium 326 test (where $K_D = 1.09 \pm 0.14$ for pyroxene and 0.1 ± 0.11 for plagioclase) (Table 3; Putirka, 327 2008). The calculated temperature range of 970-1170 °C is hotter than the estimates of the 328 temperature for the neighbouring SHV magma reservoir, which is thought to reside at 840 \pm 329 40 °C based on experimental studies and pyroxene thermometry, heated by mafic magmas 330 331 with temperatures of 900 \pm 100 °C (Devine et al., 1998; Barclay et al., 1998; Murphy et al. 2000; Devine et al. 2003; Humphreys et al., 2009) (Table 3). The SSH temperatures reported 332 here were calculated on different samples and give a wide temperature range, which supports 333 our argument that the erupted magma comprises components assembled from multiple 334 magma bodies with differing storage conditions. 335

336 The melt inclusion data were used to estimate equilibration pressures using Volatilecalc (Newman and Lowenstern, 2002; Table 4). Most of the calculated pressures 337 (using a temperature of 1000 °C) range from 194-267 MPa, which equates to depths of 8.4-338 11.6 km (using an upper crustal density of 2300 kg/m³; Hautmann et al., 2013), with one 339 sample yielding a pressure of 25 MPa and a depth of 1.2 km. By comparison the magma 340 stored beneath SHV is thought to reside in a dual reservoir system, one at 5-6 km depth, and 341 the other at 10-12 km depth (Devine et al., 1998; Murphy et al., 1998; Barclay et al., 1998; 342 Elsworth et al., 2008; Paulatto et al., 2010). With the exception of the low H₂O measurement 343 344 (1.5 wt.%), which likely represents a melt inclusion that either equilibrated at shallow depth (1.2 km) or has lost H⁺ by diffusive equilibration (Gaetani et al., 2012), the H₂O contents in 345 the SSH melt inclusions lie at the upper range of H₂O contents (1.0-6.3 wt.%) measured in 346 SHV melt inclusions (Humphreys et al., 2009; Mann et al., 2013; Edmonds et al., 2014). 347 Thus, the high anorthite contents in the cores of the SSH plagioclase crystals (up to An₉₇) are 348

most likely due to the high dissolved H_2O contents (water contents exert a first order control on anorthite content and can elevate the anorthite contents to >An₉₀; Figure 4 in Lange et al., 2009).

352 *Melt inclusion chemistry*

353 With one exception, H₂O contents are approximately constant over the entire range of K₂O, SiO₂ and MgO concentrations (Fig. 11). At depths of 8-12 km, the exsolved vapour is 354 likely to be CO₂-rich (Blundy et al., 2010), and the invariant water contents may thus reflect 355 that the source of magmas hosting the phenocrysts erupted at SSH had similar primary H₂O 356 357 contents (Tables 2 and 4). Cl concentrations are positively correlated with those of SiO₂, consistent with Cl behaving incompatibly with little or no degassing. Both CO₂ and S 358 contents decrease with increasing SiO₂, indicating that these volatiles were progressively 359 partitioned into a vapour phase as melts evolved. This is consistent with experimental data 360 that suggests that oxidised arc rhyolites are associated with high vapour-melt partition 361 362 coefficients for sulphur (Clemente et al., 2004; Zajacz et al., 2012). Similar melt inclusion trends have been observed in melt inclusion suites from Grenada which range from basalt to 363 rhyolite and thought to be related by fractional crystallisation (Devine, 1995), as well as 364 365 other examples from Kermadec arc (Haase et al., 2006; 2011; Barker et al., 2013), South Sandwich islands (Pearce et al., 1995), Mt Shasta (Grove et al., 2003) and from experimental 366 studies (Sisson et al., 2005). 367

Figure 2 illustrates a comparison of melt inclusion and whole rock data from SSH and SHV with models of fractional crystallisation at pressures of 100-200 MPa under moderately oxidizing conditions using the AlphaMelts/RhyoliteMELTS model (Ghiorso and Sack 1995; Gualda et al., 2012). Two different scenarios are considered, the first models fractional crystallisation from a mafic bulk rock starting composition, and the second starts the model

from the most mafic melt inclusion composition. In the first, the starting composition is 373 defined by the most mafic of the SSH whole rocks (~47% SiO₂). The input parameters 374 include a fixed pressure (100 or 200 MPa), a starting temperature of 1200°C (as defined by 375 376 the two pyroxene thermometer above, and close to the calculated liquidus temperature from RhyoliteMELTS) and an oxygen fugacity, fo2, buffered at QFM+2 or NNO (Devine et al., 377 1998; Murphy et al., 2000). The melt was then cooled at 50 °C intervals to simulate isobaric 378 fractional crystallization involving olivine, plagioclase, magnetite, augite, enstatite and 379 amphibole (Table 2, Fig. 2). 380

Regardless of the pressure or f_{02} , simple isobaric fractional crystallization predicts non-linear liquid lines of descent that fail to reproduce the simple linear trends defined by the majority of the whole rock data. Hence, the range in whole rock data from both SSH and SHV are best described by a hybridization model in which the rocks are mixtures between andesitic to rhyolitic melts and mafic crystal phases, as observed in many other arc volcanic settings (Davidson et al., 2005; Reubi and Blundy, 2009; Kent et al, 2010; Cashman and Blundy, 2013; Humphreys et al., 2013; Cooper and Kent, 2014).

In contrast, a fractional crystallisation history can explain most of the melt inclusions 388 from SSH, and a significant proportion of those from SHV. These melt inclusions do not lie 389 on the linear trend defined by the whole rock data. For the melt inclusions, the best fit to the 390 391 AlphaMelts/RhyoliteMELTS model (Ghiorso and Sack 1995; Gualda et al., 2012) is provided 392 by a scenario in which the starting composition is defined by the most mafic of the SSH melt inclusions (58.7% SiO₂). The input and cooling parameters are the same as for the first 393 modelling scenario above and, again, the effects of pressure and f_{O2} do not yield major 394 395 variation in the liquid line of descent (Fig. 2).

To summarise, the melts are related to one another by fractionation crystallisation and likely evolve in closed systems in storage lenses in the crust. The bulk basaltic lavas are ³⁹⁸ "assembled" by mixing liquids along this line of descent with mafic crystal mushes ³⁹⁹ containing mixtures of plagioclase, olivine and clinopyroxene. The whole rocks therefore ⁴⁰⁰ represent hybrids or mixtures between melts and mush components. In detail, it can be ⁴⁰¹ observed that most of the melt inclusion liquids are in equilibrium with their host crystals ⁴⁰² (Table 2), which means that at the time of melt entrapment, the crystal and its carrier liquid ⁴⁰³ were in equilibrium. The crystals are strongly zoned however, and the melts are therefore not ⁴⁰⁴ necessarily in equilibrium with other parts of the crystal, or with other crystals in the magma.

The melt inclusions were trapped over a pressure range corresponding to depths of 405 406 between 8 and 12 km (Table 4). We speculate that the more mafic liquids are sourced from the deeper parts of the magma reservoir system. In contrast to SHV, the crystal assemblage at 407 408 SSH is markedly more mafic, likely derived from deeper in the crust. For the basalts of the 409 SSH, the depths recorded from volatile solubilities in melt inclusions suggest that melt 410 entrapment occurs at the deeper end of the range estimated for the SHV system Edmonds et al., (2014), thus preserving a greater range of melt inclusion compositions (from andesite to 411 412 rhyolite), further suggesting that in general melts become more evolved upward through the crust. This is supported by a broad negative correlation in the melt inclusion data, between 413 SiO_2 and equilibration pressure ($R^2 = 0.45$), indicating that the least evolved compositions 414 were generally formed at deeper depths. 415

It is important to note that the record of pressures recorded by the melt inclusions is itself subject to bias. The depths of melt entrapment are probably governed not only by the physical dimensions of the reservoir but also and perhaps more importantly by the conditions under which melt inclusions form, which requires both high degrees of undercooling and a period of isothermal crystal growth (Kohut and Nielsen, 2004; Kent et al., 2008). Mafic phenocrysts may have not experienced sufficient undercooling, until mixing, by which time the compositions had been modified by time isothermal crystallisation occurs and meltinclusions become trapped (Koleszar et al., 2012).

Mixing is well documented in other arc systems. A notable example of the mixing 424 425 process described above is associated with the Mount St Helens dacite, where temperature fluctuations of 20-40 °C were a consequence of incremental, or pulsed assembly of crustal 426 magma bodies wherein each pulse interacts with ancestral, stored magmas, accounting for 427 much of the plagioclase zoning and textural complexity seen in the erupted magmas 428 (Cashman and Blundy, 2013). These authors suggest that magma storage systems under most 429 430 arc volcanoes are dominated by similar processes, where crystal mushes are fed by hotter, slightly more mafic magma, coupled with episodes of magma ascent from one storage region 431 to another. The presence of common enclaves of cumulate material, such as gabbro and 432 433 pyroxenite, in the SSH lavas (Cassidy et al., 2014) is also consistent with the remobilisation 434 of plutonic material. The way in which the model we propose differs from this fundamental mixing scenario is that we propose "back-mixing" to generate mafic bulk compositions by 435 436 mixing more evolved melts with mafic mushes, illustrating the importance of not only mushes, but also regions of andesitic to rhyolitic liquids in magma reservoirs for generating 437 bulk compositions. 438

439 *Textural evidence for mixing*

The olivine, plagioclase and pyroxene phenocryst compositional profiles all record normal and reverse zoning, suggesting a combination of growth zoning and magma mixing (Figs. 6, 7, 8 and 11). Major element mineral chemistry is modified during growth in response to cooling, melt compositional changes and magma reservoir conditions; including pressure, temperature, volatile content and f_{02} (Housh & Luhr 1991; Nelson & Montana, 1992; Sisson and Grove, 1993; Couch et al., 2003a, 2003b; Streck, 2008; Cashman and Blundy 2013). Minor element concentrations are particularly useful for discriminating between magma mixing and growth zoning, as they are almost entirely a function of melt
composition and are largely unaffected by changes in magma storage conditions (Ruprecht
and Worner, 2007; Aigner-Torres et al., 2007).

450 Zoning profiles in plagioclase crystals shows that anorthite contents are negatively correlated with Fe, Mg and Ti (Fig. 8), with magma crystallisation and differentiation 451 yielding less An-rich compositions, and increases in magma temperature or water content 452 raising An contents. Although Fe partitioning in plagioclase strongly depends on crystal 453 composition, and melt temperature and f_{02} (Longhi et al., 1976; Sugawara, 2001; Aigner-454 455 Torres et al., 2007), melt composition has the greatest effect on Fe plagioclase content (Ginibre et al., 2002). By comparison, experimental data show a clear negative correlation 456 between Ti and An% that is largely independent of temperature, and Mg partitioning depends 457 458 weakly on An content (Bindeman et al., 1998) and temperature (Longhi et al., 1976; Aigner-Torres et al 2007). Therefore, changes in An content, temperature, f_{O2} alone cannot fully 459 replicate the observed increases in Fe, Mg and Ti observed at the rim of the crystals (Fig. 8). 460 461 Rather, these observations suggest that the increases in these elements must be due, at least in part, to disequilibrium crystallisation prior to eruption as a result of mixing with melts 462 enriched in Fe, Mg and Ti. This interpretation is supported by the kernel density plots of 463 anorthite content (Fig. 4), where two populations of cores are evident, as well as a large range 464 of anorthite values at the rims. The population of cores with An₇₆₋₉₅ likely represents deeper, 465 466 more stable plagioclase crystallisation, but the cores with lower anorthite contents (An₅₀₋₆₅) may represent plagioclase crystals that evolved in a shallower (lower PH₂O), more evolved, 467 magma body. Zoning profiles (Figure 8a) show cores with high anorthite contents (An₈₆) and 468 469 increasingly albitic rims (down to An₅₆) with a corresponding increase in Fe, Mg and Ti contents.. This zoning profile is consistent with a plagioclase from a wet mafic mush being 470 471 mixed into a more evolved melt at lower pressures.

472 A history of mixing is supported by the presence of two distinct groups in olivine core compositions (Figs. 4 and 5): Group 1, Fo₇₂₋₈₀ and Group 2, Fo₅₆₋₆₈. These groups suggest 473 mixing between two distinct magma batches, or with the entrainment of more forsteritic 474 475 olivines from a crystal mush into a more evolved crystal-rich magma. The olivine crystals (both Group 1 and Group 2) exhibit both normal (most common) and reverse zoning at the 476 rim of the crystal (Figs. 5c, 6, 7 and 12a). Many of the Group 1 olivines exhibit normal 477 zoning at the rims, consistent with magma from a primitive mush entrained into a more 478 evolved storage system. This hypothesis is illustrated by the zoning profile in Figure 6b, 479 480 which contains shows a Group 1 olivine with a lower forsterite, but higher Ca and Mn rim. Simple fractional crystallisation would reduce the CaO content along with Fo content, but 481 while Ca and Mn partitioning are not directly affected by melt f_{O2} and temperature (Dunn, 482 483 1987; Libourel, 1999), Ca concentration of olivines is strongly dependent on the alkali 484 composition of the melt (Jurewicz and Watson, 1988; Libourel, 1999). Mixing of the Group 1 olivines into an evolved melt with a higher alkali content may therefore explain the 485 486 observed increased Ca content with decreasing Fo. The reverse zoning observed in some of the Group 2 olivines is consistent with olivine from the partially crystalline andesite being 487 exposed to more mafic compositions and hotter temperatures of the intruding magma. 488

Pyroxene Mg# can change in response to changes in melt composition or f_{O2} (Streck 489 et al., 2002). Thus, the saw-tooth major element zoning in Figure 10 is likely related to a 490 491 combination of open system fractionation and recharge (Ginibre et al., 2002; Ruprecht and Worner, 2007), while the relatively large increases in Mg# approaching the rims (the outer 40) 492 μ m) of a fraction (~5%) of the pyroxenes are consistent with a change in the composition 493 494 and/or temperature of the intruding mafic magma (Fig. 10). Indeed, similar orthopyroxenes have been erupted at SHV eruption since May 1996, with well-developed reverse zoned rims 495 496 (10–25 µm) (Murphy et al., 2000).

498 The mixing of a phenocryst into a melt of a different composition would lead to a sharp step in the mineral composition crystallising at the rim, assuming that conditions for 499 crystal growth are maintained and that the mixing event results in an instantaneous, rather 500 501 than gradual, change in the composition of the host melt. This sharp step then relaxes over time, via diffusion, as the interior of the crystal begins to equilibrate with its new host melt 502 composition. The resulting diffusion profiles may be used to estimate the timescales between 503 504 magma mixing and eruption, by assuming a particular temperature (Costa and Chakraborty, 2004 Morgan et al., 2004; Costa and Dugan, 2005; Costa et al., 2008). This diffusion 505 chronometric approach has been applied to reverse zoning profiles in our SSH samples (we 506 cannot apply it to normal zoning profiles, because it is difficult to distinguish mixing-driven 507 disequilibrium from fractionation-dependent growth zoning in this case). We use the DIPRA 508 509 model (Girona and Costa, 2013) for both forsterite and Mn zoning, at 1000°C. The shapes of the compositional profiles in the reverse zones at the rims of two olivines (Fig. 5b) are 510 consistent with relaxation of an initial compositional step over 10 to 60 days (Supplementary 511 figures 1 and 2). This timescale is similar to that estimated from compositional profiles in 512 Fe-Ti oxides induced by heating in SHV lavas, where andesite remobilisation by mafic 513 intrusions occurred days to weeks prior to eruptions (Devine et al., 2003). A timescale of 514 days to weeks between mixing and eruption is comparable to the short pre-eruptive mixing 515 timescales calculated at Ceboruco, Quizapu, Nea Kamini and Mount Unzen volcanoes (days 516 517 to months; Nakamura, 1995; Chertkoff and Gardner, 2004; Ruprecht and Cooper, 2012; Martin et al., 2008). Other mixed systems at Trident, Taupo and Volcan San Pedro, give 518 longer timescales (months to decades; Coombs et al. 2000; Costa and Chakraborty, 2004; 519 Millet et al., 2014). Our results imply a relatively short period between the assembly of the 520 SSH magmas and their ascent and eruption at the surface. 521

522 Formation of basaltic magmas at the SSH

Basalts are often thought to represent relatively unmodified primary melts from the 523 mantle. However observations in this study from whole rock trends, melt inclusions, 524 fractional crystallisation modelling and phenocryst zoning attest to a hybridisation model 525 526 similar to that previously inferred for the formation of andesites at many intermediate systems. Magma mixing commonly occurs between mafic and felsic melts to form andesitic 527 compositions, following the recharge filtering model of Kent et al. (2010). However, the 528 529 basaltic whole rock compositions of SSH are generated mixing components from multiple magma bodies, comprising andesitic to rhyolitic melt compositions and mafic mineral phases. 530 The SSH preserves a wide range of melt inclusion compositions unlike the SHV which 531 comprises only limited range of evolved rhyolitic melt inclusions. This is likely a 532 consequence of the deeper mixing of multiple different magma bodies and the lack of a 533 534 further shallow crystallisation stage, which would otherwise increase the likelihood of preserving silicic melt inclusions through the incorporation of crystals derived from a shallow 535 crystal mush. 536

537 Tectonic control for the eruption of basalts

While many of the observations relating to magma mixing as a control over whole 538 539 rock and melt inclusion compositions have been well-documented in arc volcanic rocks (Reubi and Blundy, 2009), they do not explain the closely-spaced and near coeval eruption of 540 basaltic and andesite lavas at SSH and SHV ~130 ka. In this context, it is noteworthy that the 541 volatile contents of melt inclusions and geophysical investigations of SHV support the 542 existence of two upper crustal magma chambers; one at 10-12 km that feed into a shallower 543 544 chamber at 5-6 km depth that serves as the source of the erupted material (Devine et al., 1998; Barclay et al., 1998; Elsworth et al., 2008; Humphreys et al., 2009; Paulatto et al., 545 2010; Mann et al., 2013; Edmonds et al., 2014). We hypothesise that the eruption of more 546

547 mafic rocks at SSH was because these lavas were assembled directly from a magma chamber 548 of similar depth (8-12 km) to the deeper of the two chambers below SHV, but without 549 passing through the shallower chamber. But what allows the SSH basalts to bypass this 550 shallow density filter?

In general, eruption of basaltic compositions in dominantly andesitic settings requires 551 a favourable stress field (Hildreth, 1981). Indeed, density is not the only factor which limits 552 the ascent of mafic magmas; structural controls imposed by lithology and rheological 553 boundaries within the crust can also act to slow and sometimes stall magma ascent 554 (Eichelberger, 1978; Dufek and Bergantz, 2005; Karlstrom et al 2009; Kent et al., 2010). 555 Faulting systems may promote the ascent of denser magmas, particularly within an 556 extensional and therefore decompressional regime. Volcanoes are also commonly found 557 558 along major strike-slip faults, such as the great Sumatran fault zone, the Sulawesi fault and the Liquiñe-Ofqui fault zone (LOFZ) in Chile (Bellier and Sébrier, 1994; Lécuyer et al., 559 1997; Cembrano and Lara, 2009). In these areas, local extensional features are associated 560 with individual volcanoes, and it is suggested that a causal relationship exists between 561 extension and volcanism or intrusion (Moore, 1979; Aydin and Nur, 1982; Hutton and 562 Reavey, 1992; Tibaldi, 1992; Milia and Torrente, 2003; Spinks et al. 2005; Brogi et al. 2010; 563 Davis et al. 2010). In addition, there is evidence that tectonics can strongly control the 564 composition of magmas. For instance, at the Taupo volcanic zone basaltic volcanism occurs 565 566 at the intersection between major faults and caldera boundaries (Cole et al., 1990; Millet et al. 2014) whereas, more intermediate magmatism occurs in areas which have experienced less 567 crustal extension (Allan et al. 2013 ; Millet et al. 2014), following the recharge filtering 568 569 process. Transtensional faults in the neighbouring island to Montserrat, Guadeloupe, which lies along the same en echelon fault system, are thought to control the location of volcanism 570 and may be the cause for the frequent sector collapses on the island (Mathieu et al., 2011). 571

Transtensional tectonics in this region may not only control the source of these magmas 572 (Cassidy et al., 2012), but may also lead to localised faulting that thus provides a pathway for 573 these higher density mafic magmas, that would otherwise be trapped within the crust (Fig. 574 12). Over time, however, the crust in these areas may impose lithostatic control as the 575 eruption of the basalts thickens the crust. As a result, later magmas would be required to 576 undergo differentiation by crystal segregation to become buoyant enough to erupt at the 577 surface (Plank and Langmuir, 1988; Devine, 1995), thus increasing the likelihood of 578 generating more evolved andesites. This is supported by numerical modelling from Pinel and 579 580 Jaupart (2000), which predicts that as the edifice grows the ascension of lower density magma is favoured, thus promoting stalling in the crust and magma differentiation. Hence, 581 the eruption of basaltic lavas may be characteristic of the early products of new eruption 582 583 centres where extensional tectonics are operative in arc settings. This may be the case for many volcanic regions which comprise early phases of basaltic activity before evolving into 584 mature andesitic systems, including northern Japan (Katsui et al., 1978; 1979); central south 585 586 Chile (Lopez-Escobar et al., 1977), New Zealand (Price et al., 2005), the Aleutians and Alaska (Marsh, 1980; Myers and Marsh, 1981). The role of transtensional tectonics is 587 strengthened by the observation that both Redonda and Kahouanne, two adjacent islands to 588 Montserrat which lie on the same transtensional fault systems (Fig. 1), also produce mafic 589 These seamounts represent the emergence of new volcanism in the Lesser 590 volcanism. 591 Antilles, and again suggest that early arc volcanism in this region may be controlled by tectonics, until further growth of the edifice inhibits the ascent of high density mafic magmas, 592 producing the commonly observed andesitic volcanoes. Although fault structures thus 593 594 provide a possible mechanism for promoting the ascent of the SSH magmas, this alone does not explain the timing of SSH basaltic magmatism. Basaltic eruptions have not been 595 identified at other periods in Montserrat's history. The conditions favourable to basaltic 596

597 eruptions at SSH thus appear to have been transient, and are unique in the currently identified history of Montserrat. The SSH doesn't clearly correspond to an initial phase of volcanism, in 598 the sense of the birth of a new volcanic centre, since the event is bracketed by andesite 599 600 eruptions at the adjacent SHV, and there have been no subsequent eruptions (since 130 ka) at SSH. We know of no reasons why fault activity at the time of SSH volcanism would have 601 been enhanced relative to other periods in Montserrat's history. Thus, although fault 602 structures may have promoted ascent of dense mafic magmas at this location, this alone does 603 not provide a satisfactory explanation for the timing of the SSH episode of basaltic 604 605 volcanism. Other processes affecting crustal stress conditions, such as collapse of the volcanic edifice, may help explain the precise timing of SSH volcanism. 606

607

608 CONCLUSIONS

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There is now abundant evidence that arc andesites are generated by hybridisation processes, involving the mixing of felsic melts and abundant crystal phases, for instance at the SHV on Montserrat, Mt St Helens and at Mount Hood (USA). Arc basalts, on the other hand, are commonly attributed to simple closed-system fractionation. Our study of the SSH, shows that olivine-bearing basalt petrogenesis can be just as complex as the generation of andesites at the SHV, implying that basalts in arcs may have a less simple history than is commonly assumed on account of the hybridisation processes explored in this study.

This study also shows how two volcanoes active at similar times and located very close to each other can erupt different bulk compositions. Basalts erupted from the SSH in Montserrat were stored under different magmatic conditions to the andesites of the SHV, yet underwent similar magmatic processes of mixing, recharge and cumulate entrainment prior to eruption. The range of magmatic temperature estimates (970 - 1160°C), reservoir depth 622 estimates (8-12 km), coupled with crystal and whole rock compositions, strongly indicates the presence of multiple magma bodies, which interact and feed basaltic eruptions. Melt 623 inclusion data, phenocryst chemistry and fractional crystallisation modelling suggests that 624 625 mixing and crystal entrainment were involved in the petrogenesis of the SSH mafic magmas. The SSH magmatic system seems to match the deeper mafic-proposed SHV magma 626 reservoir, but geophysical and petrological studies suggest that this deeper SHV system is 627 much larger in volume than the shallow SHV reservoir. This is in contrast with the SSH, the 628 results here show evidence for small, discrete pockets of crystal mushes with melt batches, 629 630 which might appear in geophysical surveys as one large reservoir. We suggest that ascent of mafic magmas can be promoted by tectonics, which may ascend along faults or under 631 specific stress conditions (i.e. post collapse). 632

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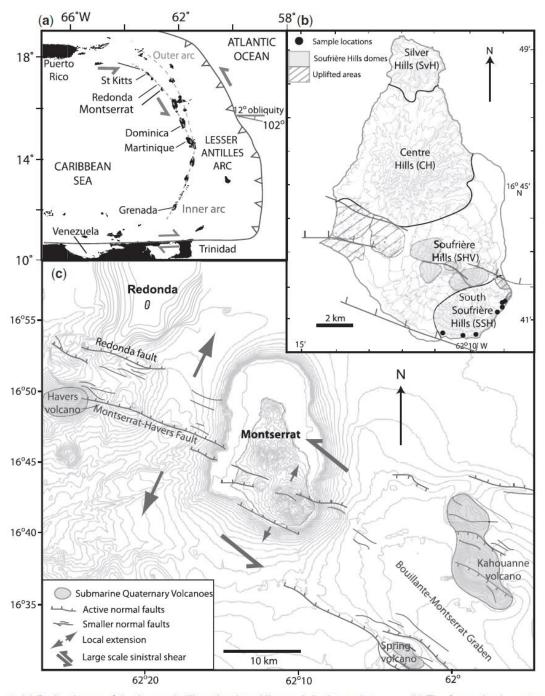


Fig. 1. (a) Regional map of the Lesser Antilles, showing oblique subduction and stresses. (b) The four volcanic centres of Montserrat, with locations of the sampled rocks from the SSH for this study indicated by the filled circles. (c) Submarine and subaerial faults and transtensional stresses in the region.

Table 1: XRF bulk-rock data along with the standard Japanese Andesite 2 (JA-2), for localities and stratigraphic units sampled at the SSH

Latitude (N): Longitude (W):	16∙67672 62∙1693	16·67622 62·1683	16∙69131 62∙1487	16∙69226 62∙1479	16∙69005 62∙1488	16∙69005 62∙1488	16.6769 62.179	JA-2	2 RSD (%)
Stratigraphic unit:*	SSH B	SSHA	SSH A	SSH B	SSH A	SSH A	SSH A		
Sample name:	SSH3	SSH4	SSH5B	SSH10	SSH7G	SSH7B	SSH1F		
Major elements (wt %)									
SiO ₂	49.22	50.87	58.77	48.06	47.84	50·19	52.69	56-35	0.2
TiO ₂	1.00	0.87	0.59	0.97	0.90	0.77	0.87	0.65	1.7
Al ₂ O ₃	18.67	19.20	17.95	18.89	19.30	19.67	17.63	15.64	2.1
Fe ₂ O ₃	10.90	9.58	7.58	10.65	10.31	9.02	8.76	6.12	0.4
MnO	0.18	0.18	0.19	0.19	0.18	0.18	0.18	0.11	1.2
MgO	5.93	4.83	3.19	5.33	5.33	4.58	4.17	7.51	1.6
CaO	10.93	10.65	7.27	10.96	11.52	10.39	9.04	6.36	1.3
Na ₂ O	2.45	2.73	3.52	2.53	2.41	2.69	3.21	3.18	3.0
K₂Õ	0.63	0.69	0.60	0.59	0.53	0.34	0.77	1.85	3.3
P_2O_5	0.12	0.13	0.15	0.10	0.09	0.11	0.21	0.15	3.7
Total	100.0	99.7	99.8	98.3	98-4	97.9	97.5	99.2	

*According to Cassidy et al. (2012, 2014).

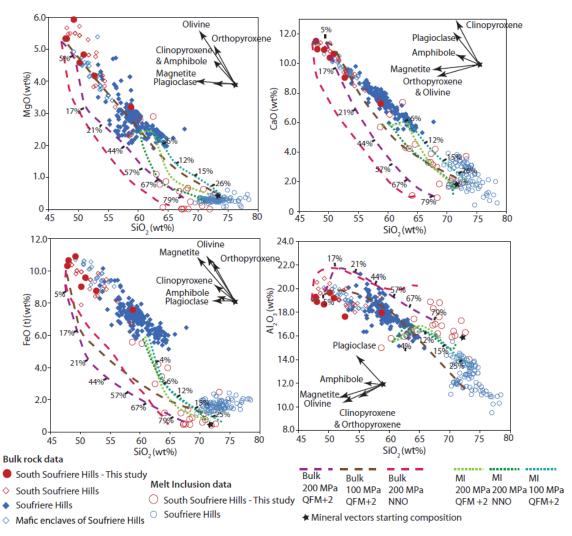


Fig. 2. Whole-rock variation diagrams from the Soufrière Hills, the mafic enclaves within the Soufrière Hills (SHV), the South Soufrière Hills (SSH), and the SSH samples used in this study. Data sources include Murphy *et al.* (1998, 2000), Horwell *et al.* (2001), Zellmer *et al.* (2003) and Cassidy *et al.* (2012). Dashed lines indicate fractional crystallization modelling under variable pressure and f₀₂ conditions.

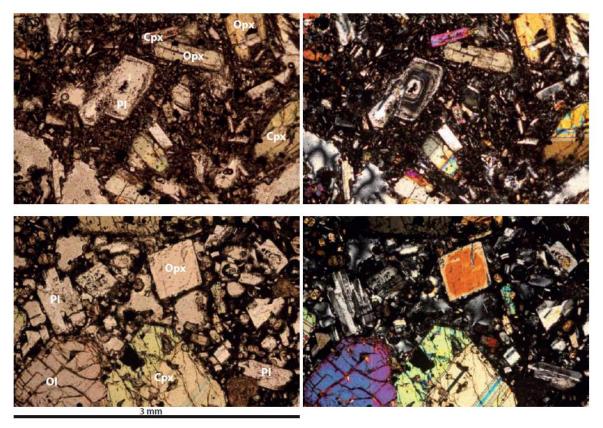


Fig. 3. Photomicrographs of two representative basalts, in plane-polarized light (left) and cross-polars (right). Each slide is 3 mm across. Some mineral phases are labelled: Cpx, clinopyroxene; Opx, orthopyroxene; Ol, olivine; Pl, plagioclase. Samples shown are 6_SSH1F (top) and 9_SSH4. The high crystallinity, large phenocrysts and features such as oscillatory zoning (e.g. plagioclase in top right photograph), normal zoning, twinning and sieve textures should be noted. Olivine is commonly large and fractured in appearance. Some pleochroism is present in clinopyroxenes.

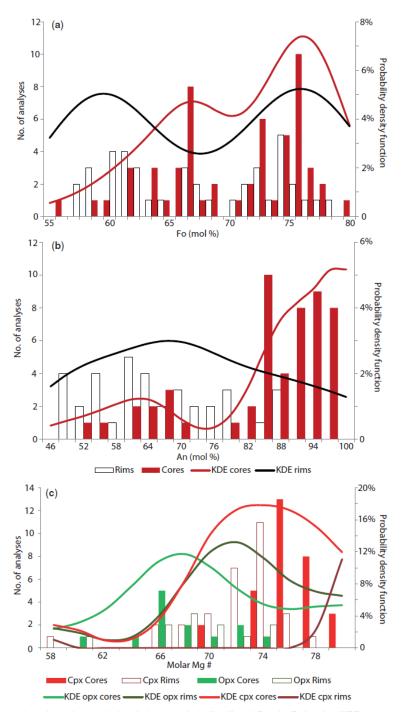


Fig. 4. Histograms to show the main crystal chemical ranges, including Kernel Density Estimation (KDE) curves, which correspond to the probability density function axis. (a) Distribution of forsterite content in olivines. (b) Anorthite distribution of cores and rims in plagioclase crystals. (c) Mg-number distribution of cores and rims in ortho- and clinopyroxene. The key for (a) and (b) is provided at the bottom of (b).

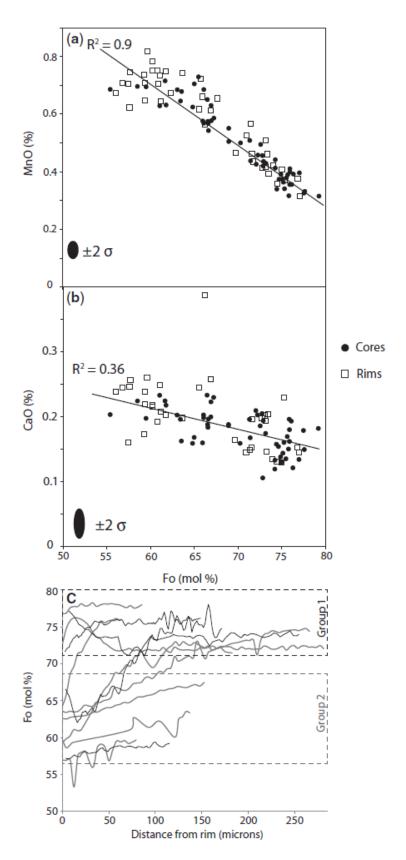
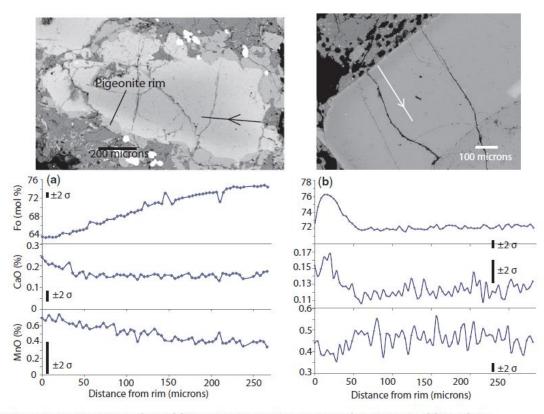


Fig. 5. (a, b) Point data plots of the chemical ranges in forsterite content against minor elements for olivines. (c) Zoning profiles for olivines. Black lines are traverses calculated by calibrated backscattered SEM images and grey lines are traverses measured directly with the electron microprobe.



1124 Fig. 6. (a) Normally zoned olivine profile (9_SSH4_ol01); (b) complex zoned rim of an olivine (15_SSH7B_Ol02).

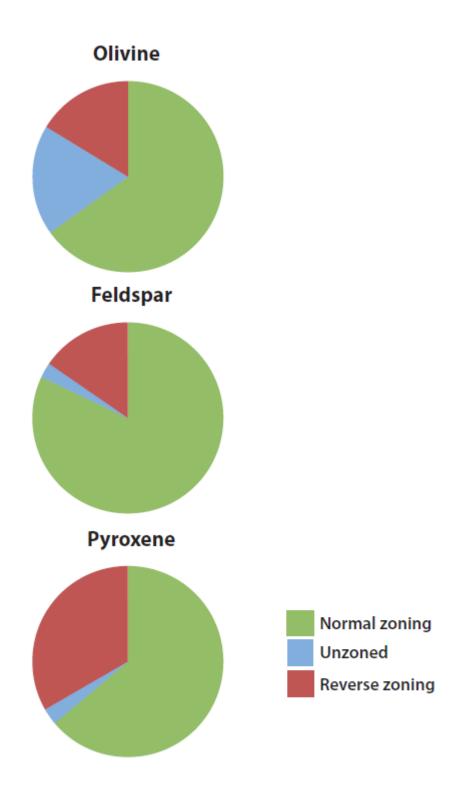


Fig. 7. Pie charts showing the proportion of crystals exhibiting normal, reverse or no zoning for olivine, plagioclase feldspars, and clino- and orthopyroxenes.

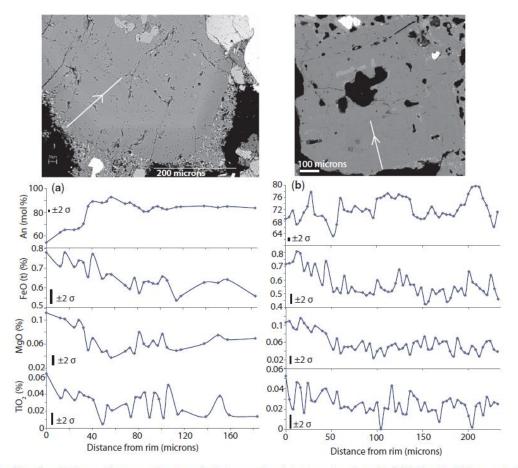


Fig. 8. Profiles of multi-element traverses showing plagioclase zoning: (a) s normal zoning of 9_SSH4Plag02; (b) demonstrates complex oscillatory zoning (15_SSH7BPlag02). 1126

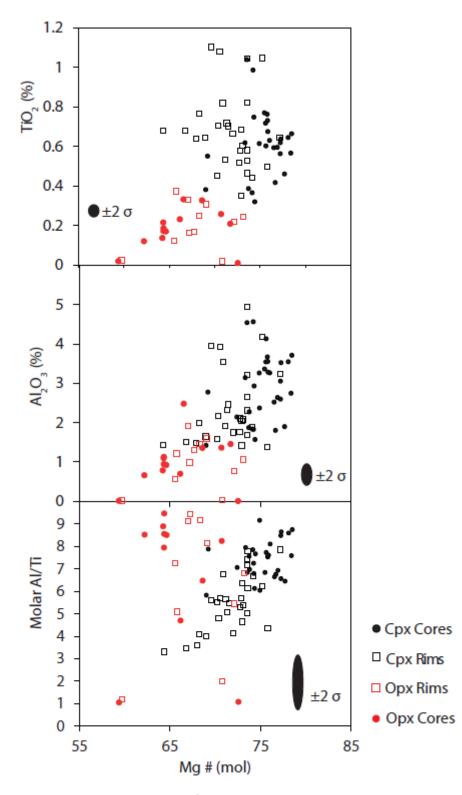


Fig. 9. Point data plots of the chemical ranges in Mg-number against minor elements for clino- and orthopyroxene.

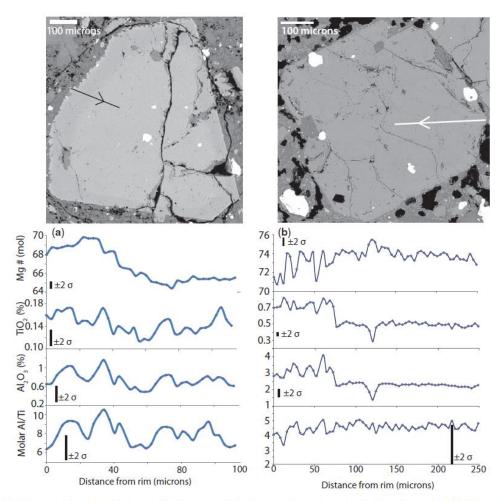
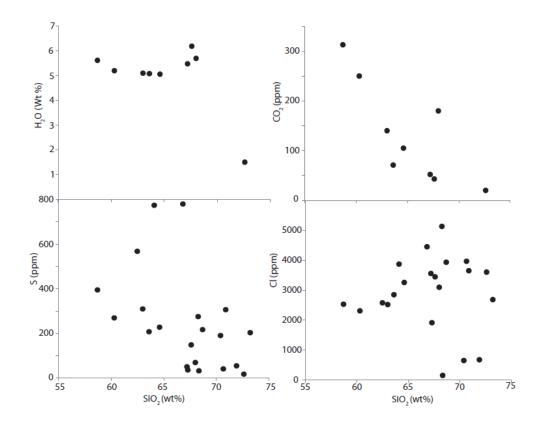


Fig. 10. Profiles of multi-element traverses showing pyroxene zoning: (a) reverse zoning of an orthopyroxene (12_SSH5Bopx09); (b) normal zoning of clinopyroxene (15_SSH7Bpyx02).

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1129 Fig. 11. Melt inclusion plots, SiO₂ versus volatile contents.

Table 2: Major and volatile element compositions for melt inclusions analysed by electron microprobe and secondary ion mass spectrometry (SIMS) analysis

Sample	Phase	SiO_2	MgO	AI_2O_3	Na ₂ O	K ₂ O	CaO	${\rm TiO}_2$	Cr_2O_3	$\rm FeO_{tot}$	Total	H_2O	CO_2	S	CI	Mg# MI	Mg# host	K _D
SSH3	Рух	72.60	0.44	16-30	4.91	2.61	1.97	0.22	b.d.	0.50	100.20	1.50	20	18	3610	67	79	0.54
SSH4	Pyx	64.60	0.87	16.40	4.08	1.05	4.53	0.86	0.009	3.98	97.00	5.06	105	228	3260	33	62	0.31
SSH4	Pyx	63.60	0.78	16.50	4.31	0.97	4.24	0.59	0.008	3.44	95.00	5.08	71	208	2850	34	63	0.31
SSH10	O	58·70	2.36	15.00	2.13	2.22	5.70	0.68	0.003	5.57	92.90	5.62	313	395	2530	49	77	0.29
SSH5B	Рух	67.60	0.02	17.00	4.51	1.58	3.14	0.48	b.d.	0.48	95-40	6·19	43	149	3450	8.7	66	0.05
SSH5B	Pyx	67·20	0.01	16.90	4-49	1.60	3.31	0.70	b.d.	0.46	95·10	5.48	52	51	3560	4.8	65	0.03
SSH10	O	60.30	2.80	15.80	1.94	0.32	6-56	0.81	b.d.	5.49	94.00	5.20	250	270	2310	54	80	0.29
SSH5B	Pyx	68·00	0.60	16.20	4.20	1.60	4.20	0.41	0.006	1.40	96.60	5.70	180	70	3100	50	76	0.31
SSH4	Pyx	63·00	1.10	16.00	3.30	0.90	4.30	0.62	0.008	4.20	93.40	5.10	140	310	2520	38	65	0.32
SSH10	Pyx	68·33	0.01	18.82	6-12	7.55	0.92	0.04	b.d.	0.47	102.28			33	141	4.8	72	0.02
SSH4	Pyx	71.91	0.01	17.29	8.11	0.67	1.87	0.06	b.d.	0.41	100.42			55	667	3.6	73	0.01
SSH4	Рух	68·68	0.24	16.86	3.92	4.78	1.85	0.57	0.004	0.89	98.47			217	3940	32	74	0.17
SSH4	Pyx	70 .39	0.22	15.91	6.04	2.03	1.75	0·17	0.002	0.60	97.30			191	642	40	73	0.24
SSH4	Pyx	68·26	0.66	17.62	5.23	2.53	2.99	0.36	0.013	0.79	99·21			276	5142	60	73	0.56
SSH3	Pyx	64·10	0.06	17.43	3.70	7.92	1.07	0.59	b.d.	1.18	96.74			774	3877	8.5	71	0.04
SSH3	Pyx	70 .88	0.63	16.04	5.31	2.15	2.24	0.38	0.006	0.76	98.99			307	3657	60	77	0.45
SSH3	Рух	67·29	0.01	18.31	5.70	2.48	3.90	0.03	0.014	0.52	98.46			37	1910	3.0	75	0.01
SSH10	Pyx	70.65	0.11	16.41	4.35	3.09	2.06	0 -88	0.002	1.15	102.93			42	3973	15	70	0 ⋅08
SSH3	Pyx	66·81	0.57	18.88	5.83	2.05	3.31	0.57	b.d.	1.17	100.02			780	4460	46	74	0.31
SSH3	Pyx	62.49	2.90	15.95	3.48	3.17	7.38	0.70	b.d.	2.98	99.55			568	2577	63	73	0.65
SSH5B	Pyx	73·21	0.28	14.03	3.65	2.26	3.73	0.57	b.d.	2.48	101.03			204	2685	17	67	0.10

All major element oxide concentrations are given in units of wt %; CO₂, S and Cl are given in ppm. FeO_{tot} denotes the FeO oxide concentration assuming that all Fe in the sample exists as Fe²⁺. Mg# Ml is the magnesium-number of the melt inclusion [molar Mg/(Mg + Fe)]; Mg# host is the magnesium-number of the crystal host (olivine, pyroxene—augite or enstatite); K_D is given by $(X_{Fe}/X_{Mg})_{host}/(X_{Fe}/X_{Mg})_{melt}$ (in moles), which is equal to 0.3 ± 0.04. Values outside this range indicate disequilibrium between melt inclusion and host (Roeder & Emslie, 1970). b.d., below detection limit.

Sample	<i>T</i> (°C)	An% or Mg# value	Method
11_SSH10_cpx04	1043	70.7	Two pyx
11_SSH10_opx06	1024	70.4	Two pyx
6_SSH1F_cpx04	970	69.0	Two pyx
14_SSH7G_plag01	1166	89.7	Plag-melt
14_SSH7G_plag06	1166	94.0	Plag-melt

 Table 3: Temperatures and pressures estimated using results of Putirka (2008)

Table 4: Equilibration pressures for melt inclusions trapped in clinopyroxene and olivine phenocrysts using the saturation models of Dixon et al. (1997) in VOLATILECALC (Newman & Lowenstern, 2004)

H ₂ O (wt %)	CO ₂ (ppm)	<i>Τ</i> (°C)	H₂O _v (mol %)	CO ₂ (mol %)	P(MPa)	Depth (km)
1.50	20	1000	86.8	13-2	26.8	1.2
5.06	105	1000	90.7	9.3	199-3	8.7
5.08	71	1000	93·5	6.5	193-9	8.4
5.62	313	1000	79.4	20.6	266-6	11.6
6-19	43	1000	97·0	3.0	247·1	10.7
5.48	52	1000	95·7	4.3	211.4	9.2
5.20	250	1000	81·1	18-9	233.7	10.2
5.70	180	1000	87.2	12.8	246.6	10.7
5-10	140	1000	88·1	11·9	208.0	9.0

Depths are estimated using a lithostatic pressure gradient of 23MPa km⁻¹ (Hautmann *et al.*, 2013).

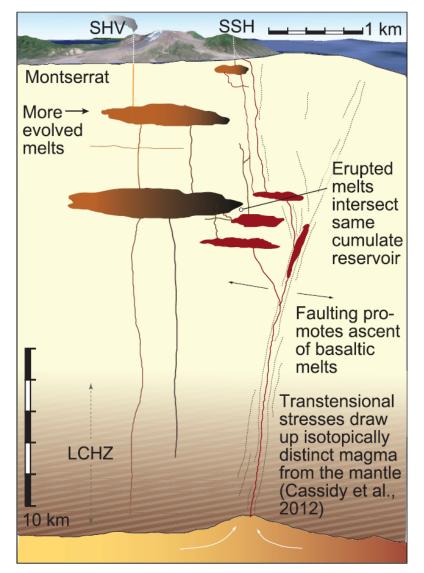


Fig. 12. Schematic figure showing how transtensional faulting can lead to the ascent of basaltic SSH magmas. The depths and processes involved in the generation of the SSH and SHV volcanism are shown. LCHZ, Lower Crustal Hot Zone of Annen *et al.* (2006).