Origin of basalts by hybridisation in andesite-dominated arcs

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Running title: Hybridised basaltic volcanism

Keywords: Crystal zoning; Magma mixing; Melt inclusions; Tectonics; Mineral chemistry, Olivine, Magma chamber
ABSTRACT

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, 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 and yet, just 3 km south of the crater, voluminous basalts have erupted from the South Soufriere Hills volcano within the same time period as the Soufriere Hills Volcano was erupting hybrid andesites (131 - 128 ka). Basaltic South Soufriere Hills magmas have 48 - 53 wt% SiO₂ and 4 - 6 wt% MgO. They were hot (970 - 1160 °C), volatile-rich (melt inclusions contain up to 6.2 wt% H₂O) and were stored at 8 – 13 km prior to eruption (based on olivine and pyroxene-hosted melt inclusion volatile geochemistry). Melt inclusions do not preserve basaltic liquids: they are andesitic to rhyolitic in composition, related to one another by a line 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 rhyolitic melts with mafic crystal phases such as magnetite, olivine, orthopyroxene and clinopyroxene. Phenocryst zoning illustrates repeated mixing events between evolved melts and mafic phenocrysts, which, when coupled with the heterogeneity of crystal compositions, strongly suggests that although the bulk composition is basalt (containing Fo₈₀ olivine), they were assembled from disparate ingredients, likely derived from mafic crystal mushes and more evolved melt lenses of variable composition. The mixing events occur days to weeks prior to eruption. We propose that the South Soufriere Hills basaltic magmas, with their higher bulk density over andesites from neighbouring volcanoes, ultimately may have been eruptible owing to both the transtensional tectonics imposed by offshore grabens (related to the oblique subduction of the Lesser Antilles) and to surface unloading caused by large scale edifice collapse. Our observations support the idea that compositional changes in arcs might
reflect not only changes in source compositions, but also effects caused by patterns in crustal strain and tectonics.

INTRODUCTION

Intermediate magmas are generated by intensive crustal magmatic processing involving crystallisation, assimilation and mixing (Anderson, 1976; Eichelberger, 1978; Rudnick, 1995; Eichelberger et al. 2006; Reubi and Blundy, 2009; Kent et al., 2010; Melekhova et al., 2013). Mafic magmas are implicated in these processes through recharging of magma bodies by mingling at the interface and by large-scale overturn in magma reservoirs (Pallister et al., 1992; Bateman, 1995). These processes are well-illustrated by volcanoes in the Lesser Antilles arc where andesitic lavas containing mafic enclaves are commonly erupted. Andesites may erupt preferentially due to their relatively low density compared to the denser mafic lavas that are “trapped” at depth by a density filter mechanism (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 al., 2009; Kent et al., 2010). Indeed, intermediate to rhyolitic magma reservoirs can obstruct 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 illustrated on Montserrat, where basalts were erupted from the South Soufrière Hills (SSH) volcano over the same broad time interval as crystal-rich andesites (with rhyolitic melts) were being erupted from the Soufrière Hills Volcano (SHV) located less than 3 km away. This raises the question as to what mechanisms allow eruption of felsic and mafic volcanic rocks in such close proximity.

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;
Eichelberger et al. 2006; Reubi and Blundy, 2009; Kent et al., 2010), the petrogenesis and 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 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 degassing that obscures their earlier characteristics. Thus, a study of closely spaced (in distance and time) andesitic and basaltic volcanism at SHV and SSH has the potential to reveal more detail regarding the nature of basaltic magmas resident in the mid- to upper-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 processes of magma storage, hybridisation, eruption triggering and growth of the arc crust.

In this paper we present new whole rock and melt inclusion analyses of basaltic to 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 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 simple processes of fractional crystallisation alone or whether mixing between disparate liquid and mush components is necessary. Mineral melt thermometry has been used (from two-pyroxenes and plagioclase-glass pairs) and barometry (using H₂O-CO₂ systematics of the melt inclusions) to estimate pre-eruptive storage conditions. We use the relaxed compositional steps across olivine crystals to infer pre-eruptive mixing timescales between felsic liquids and mafic crystals. Using all of the available petrological and geochemical data 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 density hybrid magmas relating to tectonics and unloading.
Geological background

The Lesser Antilles, like many arcs, comprises predominantly andesitic volcanic 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 present, they generally occur as small-volume centres adjacent to much larger andesitic volcanoes (Westercamp and Mervoyer, 1976; Rea and Baker, 1980; Macdonald et al. 2000).

This is exemplified on Montserrat (Fig. 1), where andesite lavas are predominant (Rea, 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. There is abundant petrological evidence (particularly from the currently active SHV) to show that the erupted andesites are hybrids formed over long timescales ($10^3$ to $10^4$ years) by multiple recharges of deeply-sourced mafic magmas into large reservoirs of crystal-rich andesite magmas prior to ascent to the surface (Murphy et al., 2000; Humphreys et al., 2009; Plail et al., 2014).

The SSH basalts are, however, sufficiently geochemically distinct from the SHV basaltic enclaves suggesting that they reflect different magma sources and processes, such as increased relative contribution from slab fluids over subducted sediments (Zellmer et al., 2003; Cassidy et al., 2012; 2014), and thus provide information regarding magmas forming 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 Antilles arc (47 wt% SiO$_2$; 6 wt% MgO), with the exception of the high-Mg basalts in Martinique (Westercamp and Mervoyer, 1976). These geochemical differences are not simply related to temporal evolution of the volcanism on Montserrat, because Ar-Ar dating and stratigraphic relationships clearly indicate that the SSH and the SHV were both active in
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 km long chain of volcanic islands formed as a result of the slow (2 cm yr\(^{-1}\)) subduction of the North American plate beneath the Caribbean plate (Fig. 1) (Wadge, 1984; DeMets et al., 2000). The oblique nature of this subduction means that the northern part of the arc is influenced by transtensional forces that have led to intra-plate deformation (Feuillet, 2000; 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 comprises four volcanic centres: Silver Hills (2600-1200 ka), Centre Hills (950-550 ka), SHV (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 products all contain abundant inclusions of mafic magma (Rea, 1974; Murphy et al., 2000; Zellmer et al., 2003; Barclay et al., 2010; Plail et al., 2014).

The SHV centre has been studied in most detail and is comprised of phenocrysts of orthopyroxene, plagioclase and amphibole in a rhyolitic glass, with clear evidence for magma mixing and mingling (Murphy et al., 2000; Humphreys et al., 2009; Humphreys et al., 2013). Under-plating of the crystal-rich andesite by wet mafic magma causes instabilities to form at 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), distributing widely dispersed mafic components (Fe-rich plagioclase microlites, K-rich glass; Humphreys et al., 2010) into the andesite body. The petrography and geochemistry of the mafic enclaves of the SHV is also best explained by a mixing process between a mafic end member (which varies in composition with time owing to lower crustal cryptic amphibole
fractionation) and variable amounts of rhyolitic melt hosting up to 20 vol% phenocrysts of plagioclase, amphibole and magnetite, although not in bulk rock proportions.

While there have been a large number of studies on the andesites of Montserrat, petrological work on the SSH basalts is more limited. Murphy et al. (2000) report that the mineral assemblage consists of plagioclase, olivine, clinopyroxene, and titanomagnetite. The SSH exposures comprise a range of rock suites from lava flows, to scoria, to reworked volcaniclastic material (Cassidy et al., 2014), with some more mafic enclaves and some lava flows containing cumulate xenoliths of orthopyroxene and plagioclase, similar to those 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 and Sm/Zr ratios, but higher Zr/Er ratios and more radiogenic Pb isotope compositions than Suite B (Cassidy et al., 2014)

METHODS

Samples

Samples of SSH rocks were collected along the south coast of Montserrat (Fig. 1; Table 1). Splits were crushed using an agate Mortar and powdered for whole rock analysis and thin sections were also cut for electron microprobe (EMPA) and scanning electron microscope (SEM) analysis. Fractions of samples were crushed coarsely and crystals of 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 secondary ion mass spectrometry (SIMS) analysis. All the inclusions analysed were natural quenched, 40-200 μm in size and were not necked or breached by cracks.

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%.

**Microanalysis (EMPA, SEM and SIMS)**

Concentrations of H$_2$O and CO$_2$ in glass were obtained by SIMS on a Cameca IMF 4f ion microprobe at the NERC microanalytical facility at the University of Edinburgh, using a 15kV primary beam of O$^-$ ions (Hauri et al., 2002; Blundy and Cashman, 2008). Positive secondary ions were accelerated to 4500 eV, with an offset of -75eV (for $^1$H and trace elements) and -50eV (for $^{12}$C) (± 20eV) to reduce transfer of molecular ions. A 50 µm raster was performed for three minutes prior to the start of each analysis, and a primary beam current of 5-6 nA used with a non-rastered, oval-shaped beam covering a 15-20 µm area on 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 cycle run: $^1$H, $^{25}$Mg, $^{30}$Si. These counts were then normalised to $^{30}$Si and converted to concentrations using a calibration curve populated by glass standards. The relative ion yield for H correlates with SiO$_2$ content, such that plotting $^1$H/$^{30}$Si versus H$_2$O yields a single working curve for glasses of variable SiO$_2$ content. CO$_2$ concentrations, however, require a correction for SiO$_2$ content.

Carbon was measured independently of $^1$H, using the same beam conditions, but with a 50 µm image field to improve transmission at moderate mass resolution, which was sufficient to resolve $^{24}$Mg$^{2+}$ at the $^{12}$C peak position for background olivine measurements and inclusion analyses. $^{12}$C was analysed for 3 s in each of 20 cycle runs in which $^{24}$Mg$^{2+}$, $^{28}$Si$^{2+}$ and $^{30}$Si were also measured. During data processing, the first 5 cycles of the $^1$H
analyses and the first 10 cycles of the $^{12}$C data were discarded to avoid the effects of surface contamination on the samples which may have survived the cleaning process. Instrumental backgrounds were minimized by allowing samples held in epoxy to outgas in a separate 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 throughout the sessions by repeat analysis of the standards as unknowns: for H$_2$O analyses these were <9% and <6% respectively; and for CO$_2$ <11% and <8% respectively. The average CO$_2$ and H$_2$O backgrounds over seven sessions were 56 ppm and 0.03 wt% respectively. There is lack of variation between Al$_2$O$_3$ and MgO in melt inclusions compositions, suggests that they do not follow the vectors anticipated for post-entrapment crystallisation of the host mineral.

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 University of Cambridge. Quantitative determinations of elements were made using the wavelength dispersive system with TAP, PET and LIF crystals. A range of metal, oxide and silicate (e.g. jadeite, wollastonite) standards was used for calibration of the spectrometers. All analyses used an accelerating voltage of 15kV. For olivine, pyroxene and plagioclase a spot size of 4 µm and a 100 nA beam current was used. For glasses, a 10 µm spot was used with a beam current of 60 nA for Cl, F, S, P, Cr and Ni, and 4 nA for all other elements, with 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 line, a series of secondary reference standards (olivines, pyroxenes, feldspars and glasses) were measured daily to check accuracy, precision and totals. Standards used were periclase for Mg, jadeite for Na, fused Si for Si, rutile for Ti, fayalite for Fe, K-feldspar for K corundum for Al, apatite for P, and pure metals for Cr and Mn. Repeat analyses of standards
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 TiO$_2$, K$_2$O, P$_2$O$_5$ 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, P$_2$O$_5$ 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.

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% SiO$_2$) (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% SiO$_2$), but the SHV contain mafic enclaves that range from basaltic to basaltic andesite (49-55% SiO$_2$).

The SSH lavas are highly crystalline, with 31-53 vol.% phenocrysts and microphenocrysts (>100 μm) 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.

**Olivine petrography**

On average, olivines form the largest crystals (mean size 390 μm; range ±100 μm) 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 (molar Fo% = Mg/(Mg+Fe) x 100) range from 56 to 80 mol. % (Figs. 4 and 5), with two main peaks in olivine core compositions (Fo$_{72-80}$ in Group 1; Fo$_{56-68}$ in Group 2) and two peaks in olivine rim compositions that are slightly less forsteritic than the cores. Most of the olivines are normally zoned or unzoned, but some exhibit reverse zoning (Fig. 6; Fig. 7), suggesting multiple magma bodies which have experienced mixing. The reverse-zoned olivines have core compositions of Fo$_{71-80}$, compared to Fo$_{56-80}$ in the normally-zoned 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 in the cores (Fig. 5). These correlations are significant at >95% confidence, with P-values <0.05. This correlation is especially strong between MnO and Fo% with a R$^2$ value of 0.9, but this correlation is less apparent with CaO and Fo% (R$^2$ of 0.36). The normally-zoned crystals show a trend of increasing Fo% from rim to core, mirrored by decreasing CaO and
MnO profiles (Figs. 6a and 6b). Figure 6d illustrates an olivine with reverse zoning towards the outer edge of the crystal, with a thin (<20 μm) band of normal zoning at the rim and no visible overgrowth. The core of this crystal has a constant forsterite composition of Fo72, except for the outer 50 μm. The increase in forsterite content in the reverse zone is positively correlated with CaO, but negatively correlated with MnO content.

Plagioclase petrography

Plagioclase crystals range in size from microlites (<15 μm) to phenocrysts (>500 μm), with the latter commonly showing both normal and oscillatory zoning, as well as sieve textures (Fig. 3). Anorthite contents (An mol.% = Ca/(Ca+Na) x 100) range from 49-97% (Fig. 4). The feldspars are commonly normally-zoned, but with rare reverse-zoned phenocrysts also present (Fig. 7), suggesting a complex set of magmatic processes have occurred. The plagioclase crystals can be separated into two main groups based on their anorthite compositions. The cores and reverse-zoned rims are anorthitic (An79-97), while the rims of the normally-zoned plagioclase are more albitic (An52-70) and are generally richer in MgO, FeO and TiO₂ than the more anorthitic cores and rims (Fig. 8). Complex dissolution and resorption is also seen in some crystals (Fig. 8b).

Pyroxene petrography

The average size of the orthopyroxene crystals is 142 ± 100 μm. 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).

Clinopyroxenes have an average crystal size of 176 ± 100 μm, with Mg# 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 TiO$_2$, Al$_2$O$_3$ and Al/Ti ratios than the enstatites. A traverse of a normally-zoned crystal shows complex saw tooth zoning (Fig. 10b) that is particularly oscillatory in the last 70 µm toward the rim, which occurs along with a sharp increase in Al$_2$O$_3$ and TiO$_2$ and a decrease in both Mg# and Al/Ti ratios.

Melt inclusion geochemistry

The melt inclusions are pristine, up to 90 µm in diameter, with no vapour bubbles and no daughter crystal phases. They span a range in compositions from andesitic to rhyolitic, with 58.2-72.6 wt.% SiO$_2$, 0.45-2.6 wt.% K$_2$O and 0.01-2.8 wt.% MgO (Fig. 2; Table 2). Their H$_2$O contents range from 1.50-6.19 wt.%, with CO$_2$ contents of 20-313 ppm (Fig. 11). CO$_2$ and S concentrations decrease with increasing melt SiO$_2$ contents, ranging from 395 ppm S and 313 ppm CO$_2$ at 58.2 wt.% SiO$_2$, to 18 ppm S and 20 ppm CO$_2$ at 72.6 wt.% SiO$_2$. Cl shows a positive relationship with SiO$_2$, ranging from 2500 ppm at 58.2 wt.% SiO$_2$ to 3610 ppm at 72.6 wt.% SiO$_2$.

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.
Pre-eruptive temperature, pressure and volatile content

Temperature estimates of the magma reservoir conditions are derived from the two-pyroxene thermometry and plagioclase-whole rock equilibria after applying the equilibrium test (where $K_D = 1.09 \pm 0.14$ for pyroxene and $0.1 \pm 0.11$ for plagioclase) (Table 3; Putirka, 2008). The calculated temperature range of 970-1170 °C is hotter than the estimates of the temperature for the neighboring SHV magma reservoir, which is thought to reside at 840 ± 40 °C based on experimental studies and pyroxene thermometry, heated by mafic magmas with temperatures of 900 ± 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 here were calculated on different samples and give a wide temperature range, which supports our argument that the erupted magma comprises components assembled from multiple magma bodies with differing storage conditions.

The melt inclusion data were used to estimate equilibration pressures using Volatilecalc (Newman and Lowenstern, 2002; Table 4). Most of the calculated pressures (using a temperature of 1000 °C) range from 194-267 MPa, which equates to depths of 8.4-11.6 km (using an upper crustal density of 2300 kg/m$^3$; Hautmann et al., 2013), with one sample yielding a pressure of 25 MPa and a depth of 1.2 km. By comparison the magma stored beneath SHV is thought to reside in a dual reservoir system, one at 5-6 km depth, and the other at 10-12 km depth (Devine et al., 1998; Murphy et al., 1998; Barclay et al., 1998; Elsworth et al., 2008; Paulatto et al., 2010). With the exception of the low H$_2$O measurement (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 H$_2$O contents in the SSH melt inclusions lie at the upper range of H$_2$O contents (1.0-6.3 wt.%) measured in SHV melt inclusions (Humphreys et al., 2009; Mann et al., 2013; Edmonds et al., 2014). Thus, the high anorthite contents in the cores of the SSH plagioclase crystals (up to An$_{97}$) are
most likely due to the high dissolved H$_2$O contents (water contents exert a first order control on anorthite content and can elevate the anorthite contents to >An$_{90}$; Figure 4 in Lange et al., 2009).

_Melt inclusion chemistry_

With one exception, H$_2$O contents are approximately constant over the entire range of K$_2$O, SiO$_2$ and MgO concentrations (Fig. 11). At depths of 8-12 km, the exsolved vapour is likely to be CO$_2$-rich (Blundy et al., 2010), and the invariant water contents may thus reflect that the source of magmas hosting the phenocrysts erupted at SSH had similar primary H$_2$O contents (Tables 2 and 4). Cl concentrations are positively correlated with those of SiO$_2$, consistent with Cl behaving incompatibly with little or no degassing. Both CO$_2$ and S contents decrease with increasing SiO$_2$, indicating that these volatiles were progressively partitioned into a vapour phase as melts evolved. This is consistent with experimental data that suggests that oxidised arc rhyolites are associated with high vapour-melt partition 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 rhyolite and thought to be related by fractional crystallisation (Devine, 1995), as well as 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 studies (Sisson et al., 2005).

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 (Ghiorsø 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
defined by the most mafic of the SSH whole rocks (~47% SiO$_2$). The input parameters
include a fixed pressure (100 or 200 MPa), a starting temperature of 1200°C (as defined by
the two pyroxene thermometer above, and close to the calculated liquidus temperature from
RhyoliteMELTS) and an oxygen fugacity, $f_{O2}$, buffered at QFM+2 or NNO (Devine et al.,
1998; Murphy et al., 2000). The melt was then cooled at 50 °C intervals to simulate isobaric
fractional crystallization involving olivine, plagioclase, magnetite, augite, enstatite and
amphibole (Table 2, Fig. 2).

Regardless of the pressure or $f_{O2}$, 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
from SSH, and a significant proportion of those from SHV. These melt inclusions do not lie
on the linear trend defined by the whole rock data. For the melt inclusions, the best fit to the
AlphaMelts/RhyoliteMELTS model (Ghiorso and Sack 1995; Gualda et al., 2012) is provided
by a scenario in which the starting composition is defined by the most mafic of the SSH melt
inclusions (58.7% SiO$_2$). The input and cooling parameters are the same as for the first
modelling scenario above and, again, the effects of pressure and $f_{O2}$ do not yield major
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
“assembled” by mixing liquids along this line of descent with mafic crystal mushes containing mixtures of plagioclase, olivine and clinopyroxene. The whole rocks therefore represent hybrids or mixtures between melts and mush components. In detail, it can be observed that most of the melt inclusion liquids are in equilibrium with their host crystals (Table 2), which means that at the time of melt entrapment, the crystal and its carrier liquid were in equilibrium. The crystals are strongly zoned however, and the melts are therefore not 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 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 SSH is markedly more mafic, likely derived from deeper in the crust. For the basalts of the SSH, the depths recorded from volatile solubilities in melt inclusions suggest that melt 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 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 SiO$_2$ and equilibration pressure ($R^2= 0.45$), indicating that the least evolved compositions were generally formed at deeper depths.

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 melt inclusions become trapped (Koleszar et al., 2012).

Mixing is well documented in other arc systems. A notable example of the mixing 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 magma bodies wherein each pulse interacts with ancestral, stored magmas, accounting for much of the plagioclase zoning and textural complexity seen in the erupted magmas (Cashman and Blundy, 2013). These authors suggest that magma storage systems under most 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 to another. The presence of common enclaves of cumulate material, such as gabbro and pyroxenite, in the SSH lavas (Cassidy et al., 2014) is also consistent with the remobilisation 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 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 bulk compositions.

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 $f_{O_2}$ (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).

Zoning profiles in plagioclase crystals shows that anorthite contents are negatively correlated with Fe, Mg and Ti (Fig. 8), with magma crystallisation and differentiation yielding less An-rich compositions, and increases in magma temperature or water content raising An contents. Although Fe partitioning in plagioclase strongly depends on crystal composition, and melt temperature and $f_{O_2}$ (Longhi et al., 1976; Sugawara, 2001; Aigner-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 between Ti and An% that is largely independent of temperature, and Mg partitioning depends 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, $f_{O_2}$ alone cannot fully replicate the observed increases in Fe, Mg and Ti observed at the rim of the crystals (Fig. 8).

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 enriched in Fe, Mg and Ti. This interpretation is supported by the kernel density plots of anorthite content (Fig. 4), where two populations of cores are evident, as well as a large range of anorthite values at the rims. The population of cores with An$_{76-95}$ likely represents deeper, more stable plagioclase crystallisation, but the cores with lower anorthite contents (An$_{50-65}$) may represent plagioclase crystals that evolved in a shallower (lower PH$_2$O), more evolved, magma body. Zoning profiles (Figure 8a) show cores with high anorthite contents (An$_{86}$) and increasingly albitic rims (down to An$_{56}$) with a corresponding increase in Fe, Mg and Ti contents.. This zoning profile is consistent with a plagioclase from a wet mafic mush being mixed into a more evolved melt at lower pressures.
A history of mixing is supported by the presence of two distinct groups in olivine core compositions (Figs. 4 and 5): Group 1, Fo$_{72-80}$ and Group 2, Fo$_{56-68}$. These groups suggest mixing between two distinct magma batches, or with the entrainment of more forsteritic 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 rim of the crystal (Figs. 5c, 6, 7 and 12a). Many of the Group 1 olivines exhibit normal zoning at the rims, consistent with magma from a primitive mush entrained into a more evolved storage system. This hypothesis is illustrated by the zoning profile in Figure 6b, 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 while Ca and Mn partitioning are not directly affected by melt $f_{O2}$ and temperature (Dunn, 1987; Libourel, 1999), Ca concentration of olivines is strongly dependent on the alkali 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 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 exposed to more mafic compositions and hotter temperatures of the intruding magma.

Pyroxene Mg# can change in response to changes in melt composition or $f_{O2}$ (Streck et al., 2002). Thus, the saw-tooth major element zoning in Figure 10 is likely related to a 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 µm) of a fraction (~5%) of the pyroxenes are consistent with a change in the composition 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 (10–25 µm) (Murphy et al., 2000).
Timing of mixing events

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 crystal growth are maintained and that the mixing event results in an instantaneous, rather 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 composition. The resulting diffusion profiles may be used to estimate the timescales between 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 chronometric approach has been applied to reverse zoning profiles in our SSH samples (we cannot apply it to normal zoning profiles, because it is difficult to distinguish mixing-driven disequilibrium from fractionation-dependent growth zoning in this case). We use the DIPRA 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 consistent with relaxation of an initial compositional step over 10 to 60 days (Supplementary figures 1 and 2). This timescale is similar to that estimated from compositional profiles in Fe-Ti oxides induced by heating in SHV lavas, where andesite remobilisation by mafic intrusions occurred days to weeks prior to eruptions (Devine et al., 2003). A timescale of days to weeks between mixing and eruption is comparable to the short pre-eruptive mixing timescales calculated at Ceboruco, Quizapu, Nea Kamini and Mount Unzen volcanoes (days 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 longer timescales (months to decades; Coombs et al. 2000; Costa and Chakraborty, 2004; Millet et al., 2014). Our results imply a relatively short period between the assembly of the SSH magmas and their ascent and eruption at the surface.
Basalts are often thought to represent relatively unmodified primary melts from the mantle. However observations in this study from whole rock trends, melt inclusions, fractional crystallisation modelling and phenocryst zoning attest to a hybridisation model 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 compositions, following the recharge filtering model of Kent et al. (2010). However, the basaltic whole rock compositions of SSH are generated mixing components from multiple magma bodies, comprising andesitic to rhyolitic melt compositions and mafic mineral phases. The SSH preserves a wide range of melt inclusion compositions unlike the SHV which comprises only limited range of evolved rhyolitic melt inclusions. This is likely a consequence of the deeper mixing of multiple different magma bodies and the lack of a further shallow crystallisation stage, which would otherwise increase the likelihood of preserving silicic melt inclusions through the incorporation of crystals derived from a shallow crystal mush.

Tectonic control for the eruption of basalts

While many of the observations relating to magma mixing as a control over whole 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 basaltic and andesite lavas at SSH and SHV ~130 ka. In this context, it is noteworthy that the volatile contents of melt inclusions and geophysical investigations of SHV support the existence of two upper crustal magma chambers; one at 10-12 km that feed into a shallower 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., 2010; Mann et al., 2013; Edmonds et al., 2014). We hypothesise that the eruption of more
mafic rocks at SSH was because these lavas were assembled directly from a magma chamber of similar depth (8-12 km) to the deeper of the two chambers below SHV, but without passing through the shallower chamber. But what allows the SSH basalts to bypass this shallow density filter?

In general, eruption of basaltic compositions in dominantly andesitic settings requires a favourable stress field (Hildreth, 1981). Indeed, density is not the only factor which limits the ascent of mafic magmas; structural controls imposed by lithology and rheological boundaries within the crust can also act to slow and sometimes stall magma ascent (Eichelberger, 1978; Dufek and Bergantz, 2005; Karlstrom et al 2009; Kent et al., 2010). Faulting systems may promote the ascent of denser magmas, particularly within an extensional and therefore decompressional regime. Volcanoes are also commonly found 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., 1997; Cembrano and Lara, 2009). In these areas, local extensional features are associated with individual volcanoes, and it is suggested that a causal relationship exists between extension and volcanism or intrusion (Moore, 1979; Aydin and Nur, 1982; Hutton and Reavey, 1992; Tibaldi, 1992; Milia and Torrente, 2003; Spinks et al. 2005; Brogi et al. 2010; Davis et al. 2010). In addition, there is evidence that tectonics can strongly control the composition of magmas. For instance, at the Taupo volcanic zone basaltic volcanism occurs 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 crustal extension (Allan et al. 2013 ; Millet et al. 2014), following the recharge filtering 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 and may be the cause for the frequent sector collapses on the island (Mathieu et al., 2011).
Transtensional tectonics in this region may not only control the source of these magmas (Cassidy et al., 2012), but may also lead to localised faulting that thus provides a pathway for these higher density mafic magmas, that would otherwise be trapped within the crust (Fig. 12). Over time, however, the crust in these areas may impose lithostatic control as the eruption of the basalts thickens the crust. As a result, later magmas would be required to undergo differentiation by crystal segregation to become buoyant enough to erupt at the surface (Plank and Langmuir, 1988; Devine, 1995), thus increasing the likelihood of generating more evolved andesites. This is supported by numerical modelling from Pinel and 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, the eruption of basaltic lavas may be characteristic of the early products of new eruption 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 mature andesitic systems, including northern Japan (Katsui et al., 1978; 1979); central south 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 strengthened by the observation that both Redonda and Kahouanne, two adjacent islands to Montserrat which lie on the same transtensional fault systems (Fig. 1), also produce mafic volcanism. These seamounts represent the emergence of new volcanism in the Lesser 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, producing the commonly observed andesitic volcanoes. Although fault structures thus 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 identified at other periods in Montserrat’s history. The conditions favourable to basaltic
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 the sense of the birth of a new volcanic centre, since the event is bracketed by andesite 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 been enhanced relative to other periods in Montserrat’s history. Thus, although fault structures may have promoted ascent of dense mafic magmas at this location, this alone does not provide a satisfactory explanation for the timing of the SSH episode of basaltic volcanism. Other processes affecting crustal stress conditions, such as collapse of the volcanic edifice, may help explain the precise timing of SSH volcanism.

**CONCLUSIONS**

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
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 inclusion data, phenocryst chemistry and fractional crystallisation modelling suggests that 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 reservoir, but geophysical and petrological studies suggest that this deeper SHV system is much larger in volume than the shallow SHV reservoir. This is in contrast with the SSH, the results here show evidence for small, discrete pockets of crystal mushes with melt batches, 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 specific stress conditions (i.e. post collapse).

Acknowledgements

Iris Buisman, Ian Croudace, Richard Pearce are thanked for lab assistance. The authors wish to thank Adam Kent, Marc-Alban Millet and Jim Cole for their constructive reviews and for the editorial handling of John Gamble. MC and SFLW thank NERC for financial support via grant NE/K000403/1.

REFERENCES


Cassidy, M., Taylor, R. N., Palmer, M. R., Cooper, R. J., Stenlake, C., Trofimovs, J.


Mathieu, Lucie, Benjamin van Wyk de Vries, Martin Pilato, and Valentin R. Troll. "The Interaction between Volcanoes and Strike-Slip, Transtensional and Transpressional
Fault Zones: Analogue Models and Natural Examples." *Journal of Structural Geology*


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

<table>
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<td>SSH A</td>
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<td>SSH5B</td>
<td>SSH10</td>
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<td>SSH7B</td>
<td>SSH1F</td>
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**Major elements (wt %)**

| SiO₂ | 49.22 | 50.87 | 58.77 | 48.66 | 47.84 | 50.19 | 52.69 | 56.35 | 0.2 |
| TiO₂ | 1.00 | 0.87 | 0.59 | 0.97 | 0.90 | 0.77 | 0.87 | 0.66 | 1.7 |
| Al₂O₃ | 18.67 | 19.20 | 17.96 | 18.89 | 19.30 | 19.67 | 17.83 | 16.64 | 2.1 |
| Fe₂O₃ | 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 | 4.71 | 1.6 |
| CaO | 10.93 | 10.65 | 7.27 | 10.86 | 11.52 | 10.39 | 9.04 | 6.36 | 1.3 |
| Na₂O | 2.45 | 2.73 | 3.52 | 2.63 | 2.41 | 2.68 | 3.21 | 3.18 | 3.0 |
| K₂O | 0.63 | 0.69 | 0.60 | 0.59 | 0.53 | 0.34 | 0.77 | 1.85 | 3.3 |
| P₂O₅ | 0.12 | 0.13 | 0.16 | 0.10 | 0.09 | 0.11 | 0.21 | 0.16 | 3.7 |
| Total | 100.0 | 99.7 | 99.8 | 98.3 | 98.4 | 97.9 | 97.5 | 99.2 |   |


Fig. 2. Whole-rock variation diagrams from the Soufrière Hills, the mafic enclaves within the Soufrière Hills (SHH), and the SSH samples used in this study. Data sources include Murphy et al. (1998, 2000), Horwell et al. (2001), Zeltner et al. (2003) and Cassidy et al. (2012). Dashed lines indicate fractional crystalization modelling under variable pressure and fO₂ 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.
Fig. 6. (a) Normally zoned olivine profile (9SSH4_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) a normal zoning of 9_SSH4Plag02; (b) demonstrates complex oscillatory zoning (15_SSH78Plag02).
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_SSH5Bpx09); (b) normal zoning of clinopyroxene (16_SSH7Bpx02).
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

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<tr>
<th>Sample</th>
<th>Phase</th>
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<th>K₂O</th>
<th>CaO</th>
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</table>

All major element oxide concentrations are given in units of wt.%; CO₂ and Cl are given in ppm. FeO_total denotes the FeO oxide concentration assuming that all Fe in the sample exists as Fe⁷⁺. Mg/Mil is the magnesium-number of the melt inclusion (molar Mg/Mil = Fe/Mil); Mg# host is the magnesium-number of the crystal host (olivine, pyroxene—augite or enstatite); K₀ is given by \( \frac{X_{K_2O}/X_{MgO\text{host}}}{X_{H_2O}/X_{MgO\text{host}}} \) (in moles), which is equal to 0.3 ± 0.04. Values outside this range indicate disequilibrium between melt inclusion and host (Reeder & Emslie, 1970). b.d., below detection limit.
Table 3: Temperatures and pressures estimated using results of Putirka (2008)

<table>
<thead>
<tr>
<th>Sample</th>
<th>( T ) (°C)</th>
<th>An% or Mg# value</th>
<th>Method</th>
</tr>
</thead>
<tbody>
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<td>89.7</td>
<td>Plag–melt</td>
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<td>1166</td>
<td>94.0</td>
<td>Plag–melt</td>
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</table>

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)

<table>
<thead>
<tr>
<th>( \text{H}_2\text{O} ) (wt %)</th>
<th>( \text{CO}_2 ) (ppm)</th>
<th>( T ) (°C)</th>
<th>( \text{H}_2\text{O}_v ) (mol %)</th>
<th>( \text{CO}_2 ) (mol %)</th>
<th>( P ) (MPa)</th>
<th>Depth (km)</th>
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</thead>
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<td>266.6</td>
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</table>

Depths are estimated using a lithostatic pressure gradient of 23MPa km\(^{-1}\) (Hautmann \textit{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).

Montserrat

More evolved melts

Erupted melts intersect same cumulate reservoir

Faulting promotes ascent of basaltic melts

Transtensional stresses draw up isotopically distinct magma from the mantle (Cassidy et al., 2012)

SHV

SSH

1 km

LCHZ

10 km