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Key Points:
- SO_2 fluxes from the Soufriere Hills volcano are decoupled from magma
- SO_2 gas is sourced at depth and emerges in pulses associated with seismicity
- Magma systemssegregate fluids and melt then destablize to erupt

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Crustal-scale degassing due to magma system destabilization and magma-gas decoupling at Soufrière Hills Volcano, Montserrat

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Abstract Activity since 1995 at Soufrière Hills Volcano (SHV), Montserrat has alternated between andesite lava extrusion and quiescence, which are well correlated with seismicity and ground deformation cycles. Large variations in SO_2 flux do not correlate with these alternations, but high and low HCl/SO_2 characterize lava dome extrusion and quiescent periods respectively. Since lava extrusion ceased (February 2010) steady SO_2 emissions have continued at an average rate of 374 tonnes/day ($\pm$ 140 t/d), and incandescent fumaroles (temperatures up to 610°C) on the dome have not changed position or cooled. Occasional short bursts (over several hours) of higher ($\sim$ 10x) SO_2 flux have been accompanied by swarms of volcano-tectonic earthquakes. Strain data from these bursts indicate activation of the magma system to depths up to 10 km. SO_2 emissions since 1995 greatly exceed the amounts that could be derived from 1.1 km^3 of erupted andesite, and indicating extensive partitioning of sulfur into a vapour phase, as well as efficient decoupling and outgassing of sulfur-rich gases from the magma. These observations are consistent with a vertically extensive, crustal magmatic mush beneath SHV. Three states of the magmatic system are postulated to control degassing. During dormant periods (10^3 to 10^4 years) magmatic vapour and melts separate as layers from the mush and decouple from each other. In periods of unrest (years) without eruption, melt and fluid layers become unstable, ascend and can amalgamate. Major destabilization of the mush system leads to eruption, characterized by magma mixing and release of volatiles with different ages, compositions and sources.

1. Introduction

It is commonly observed that sulfur dioxide (SO_2) fluxes from arc volcanoes are greatly in excess of the amounts predicted from the amount of dissolved sulfur in the associated erupted silicic magmas [*Wallace*, 2001; *Wallace and Edmonds*, 2011; *Fiege et al.*, 2014]. This observation is commonly explained as the consequence of unerupted basaltic magmas supplying the excess sulfur, particularly when vesiculation accompanies quench crystallization of the basalt against cooler silicic magma in an upper crustal magma chamber. A sulfur-rich exsolved volatile phase could be released from deeper underplated basalt and migrate upward into more silicic magmas by permeable flow [*Bachmann and Bergantz*, 2006] without requiring direct mafic-silicic magma interaction. The silicic magma is then erupted and the SO_2 is advected to the surface in an exsolved vapour phase and vented to the atmosphere. Evidence for underlying basalt magma commonly occurs in the form of small proportions of quenched mafic inclusions and dispersed crystals from the mafic magma mixed into the silicic magma [*Martel et al.*, 2006; *Humphreys et al.*, 2009]. Invariably, the proportion of admixed mafic magma is, in itself, insufficient to account for the released SO_2, but is invoked as circumstantial evidence for the presence of larger volumes of such magmas at depth.

The ongoing eruption of Soufrière Hills Volcano (SHV), Montserrat (1995 to present) has been interpreted within this model framework [*Humphreys et al.*, 2009; *Barclay et al.*, 2010; *Christopher et al.*, 2010; *Edmonds et al.*, 2010; *Plail et al.*, 2014]. However, the existing paradigm does not explain many of the detailed observations that have arisen from the SHV eruption. In particular, magma and gas fluxes are largely decoupled, with large gas fluxes continuing for many years after eruptive activity has ceased [*Christopher et al.*, 2014a].

If the gases are sourced from magmas intruding the system syn-eruptively, then these observations seem to require a mechanism for rapid separation of melts and fluids. Here we summarize SO_2 flux data and recent observations from SHV, and develop a novel alternative model of magma system destabilization with major implications for interpreting arc magma degassing and volcano behavior.

It is envisaged that SHV is underlain by a vertically extensive large igneous mush system (crystals + melt ± gas) formed over tens of thousands of years and perhaps extending from the shallow crust (> 4 km) to the Moho. During long periods of quiescence between eruptions the mush evolves by additions of hydrous basalt magma from the mantle, igneous differentiation (including fractionation, assimilation and partial melting) and compaction-driven upward melt migration. In some regions of the mush the melts will be vapour-saturated and so magmatic fluids will coexist. Multiphase crystal-melt-fluid system compaction, with upward movement of melt and fluid and downward movement of crystals, leads to the gradual formation of discrete layers of melt and fluid [*Solano et al*., 2012; *Connolly and Podladchikov*, 2013]. The large differences in viscosity and density between melt and exsolved fluids make it likely that, as the solid matrix compacts, melts and fluids will migrate upward at different rates. Large-scale destabilization of the magmatic system occurs when melt and fluid layers amalgamate and accumulate toward the top of the system to form shallow-level magma chambers. We propose that the current SHV eruption is the consequence of such a destabilization event. Because volatiles have exsolved and segregated from the melt prior to eruption, the eruption of magma and discharge of volatiles become partly or wholly decoupled in space and time. The large differences in solubility and melt-fluid partitioning behavior of different volatile species as functions of pressure and melt composition lead to major variations in emitted gas chemistry. Gas flux data, geochemistry, petrological data and geophysical observations are summarized to provide evidence for this alternative model.

2. Observations

The main observations from monitoring of the SHV eruption are summarized in Figures 1 and 2. Seismic unrest started in 1992 [*Wadge et al*., 2014] and culminated in the start of the eruption in July 1995. Magma (porphyritic hornblende andesite) first reached the surface in November 1995. Since then about 1.1 km^3 of andesite has been erupted [*Wadge et al*., 2014] in activity characterized by alternating periods of lava dome extrusion and quiescence (i.e., lack of surface magmatic activity). Occasional Vulcanian explosions have contributed only a minor fraction (<1%) to the total inventory of SO_2, as shown by satellite measurements [*Carn and Prata*, 2010]. Seismicity and ground deformation correlate well with these alternations, with copious seismicity and deflation during periods of dome extrusion contrasting with ground inflation and limited seismicity during quiescent periods [*Mattioli et al*., 1998; *Norton et al*., 2002; *Elsworth et al*., 2008].

The SO_2 flux data show marked fluctuations, with a long-term average of 490 tonnes per day ± 196 t/d up to February 2010 [*Christopher et al*., 2010]. In a cumulative plot (Figure 2) the flux has been quite steady, albeit with marked fluctuations. Between 1996 and 2010 several maxima in SO_2 flux were observed, three of which are quite prominent after 2002 when the quality and resolution of the measurements improved considerably (from one measurement per day to one every few minutes) due to replacing COSPEC with mini-DOAS automated UV spectrometer measurement systems. Details of the instrumental design, retrievals procedure and error analysis are given by *Edmonds et al*. [2003], expanded upon more recently by *Kern et al*. [2012]. Uncertainties (2σ) on individual measurements are not likely to exceed ±40%, with most of the uncertainty attributable to estimations of plume speed and to attenuation of the signal with distance away from the plume [*Kern et al*., 2012]. Measurements of SO_2 flux were carried out by both traverse methods and scanning methods throughout 2002 and there was good agreement between the two (see discussion by *Edmonds et al*. [2003]). Three peaks in SO_2 flux between 2003 and 2010 suggest a 2 year cyclicity, a period similar in duration to, but entirely out of phase with, the first three phases of extrusion [*Christopher et al*., 2014a]. The SO_2 flux fluctuations are not well correlated with lava dome extrusion, periods of inflation or deflation nor with seismic event rate.

After the last dome collapse in February 2010 there has been no lava extrusion and the volcano has entered into the longest period without dome growth since the onset of the eruption. Since February 2010 seismicity has been low and has been characterized by a declining number of rockfalls. There have been sporadic bursts of volcano-tectonic (VT) earthquakes, termed VT strings, associated with surges in SO_2 flux, a

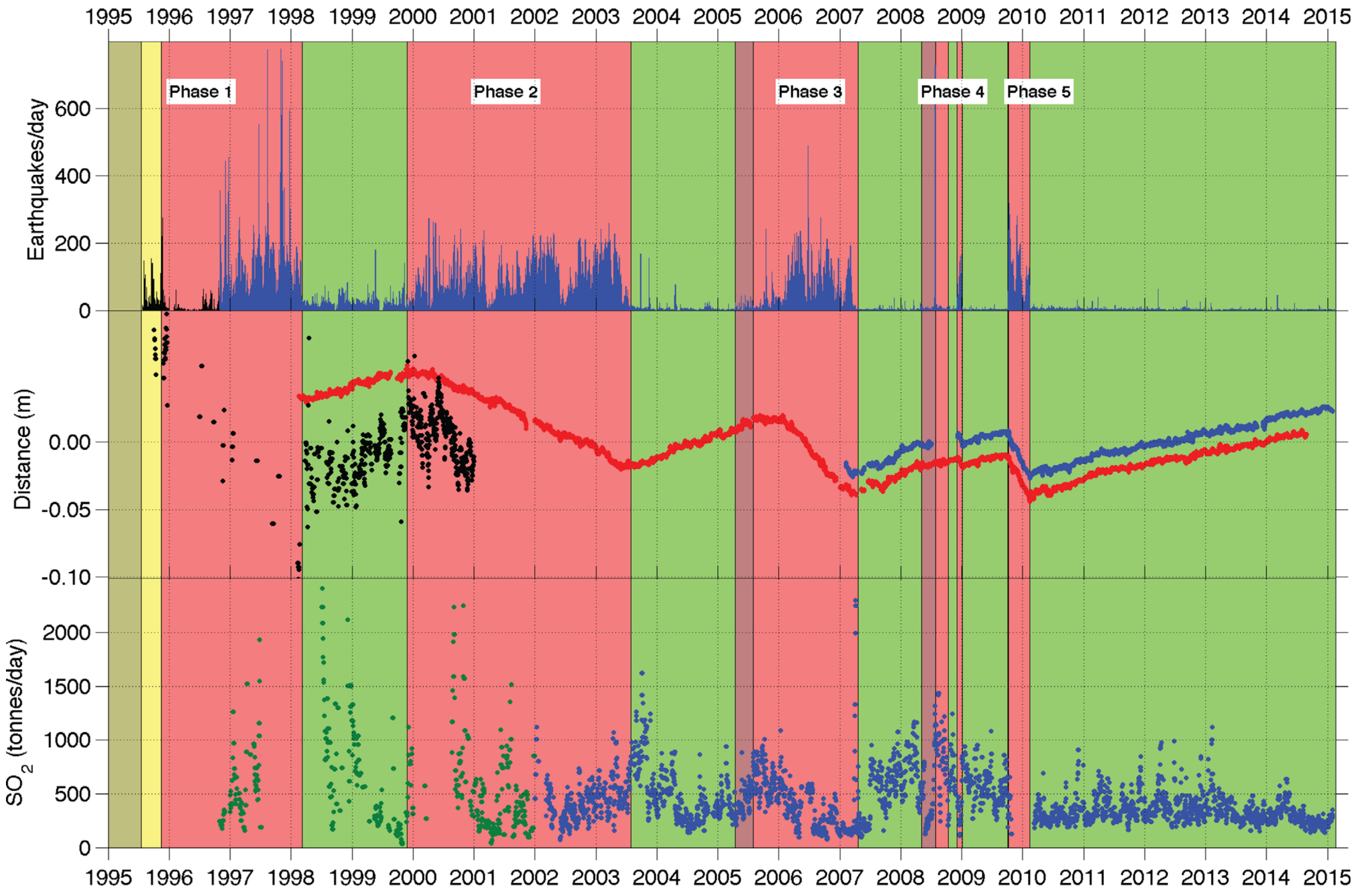

Figure 1. Seismic, GPS and SO_2 monitoring data at the Soufrière Hills Volcano, Montserrat for the period 1 January 1995 to 31 August 2014. Extrusive phases and pauses are shown in red and green respectively. (top) Number of seismic events detected and identified by the seismic system. (middle) GPS data smoothed with 7 day running mean filter. Red: radial displacement of station MVO1 (GAMIT processing) located 6 km north of the dome summit, Black: GPS height of HARR about 2.5 km NNE of the dome. Precise location of GPS stations can be found in *Wadge et al.* [2014]. (bottom) measured daily SO_2 flux, filtered with 7 day running median filter. Green: COSPEC data, Blue: DOAS data. All data shown are archived by the Montserrat Volcano Observatory.

phenomenon first recognized in 2007. Although inflation has continued throughout this quiescent period, SO_2 output has changed in its character. From February 2010 to December 2014 the average flux was 374 tonnes SO_2 per day ($\pm$ 140 t/d). There are large fluctuations in daily rate (Figure 2), which partly reflects measurement error and partly real variability, with the highest and lowest values being 4500 and 66 tonnes per day respectively. Overall the SO_2 output has been quite steady, with no indications of the marked cyclic variations recorded prior to 2010 (Figure 1). The changed gas flux patterns before and after February 2010 suggests a change in system behavior.

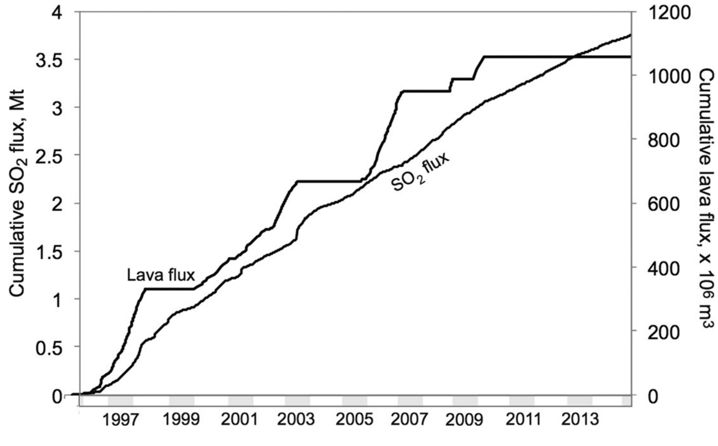

Figure 2. Cumulative SO_2 flux and lava volumetric flux versus time.

Gas flux since February 2010 (Figure 3) shows several short bursts of increased SO_2 flux over periods of a few hours to several days, which are associated with short VT strings in which rates of

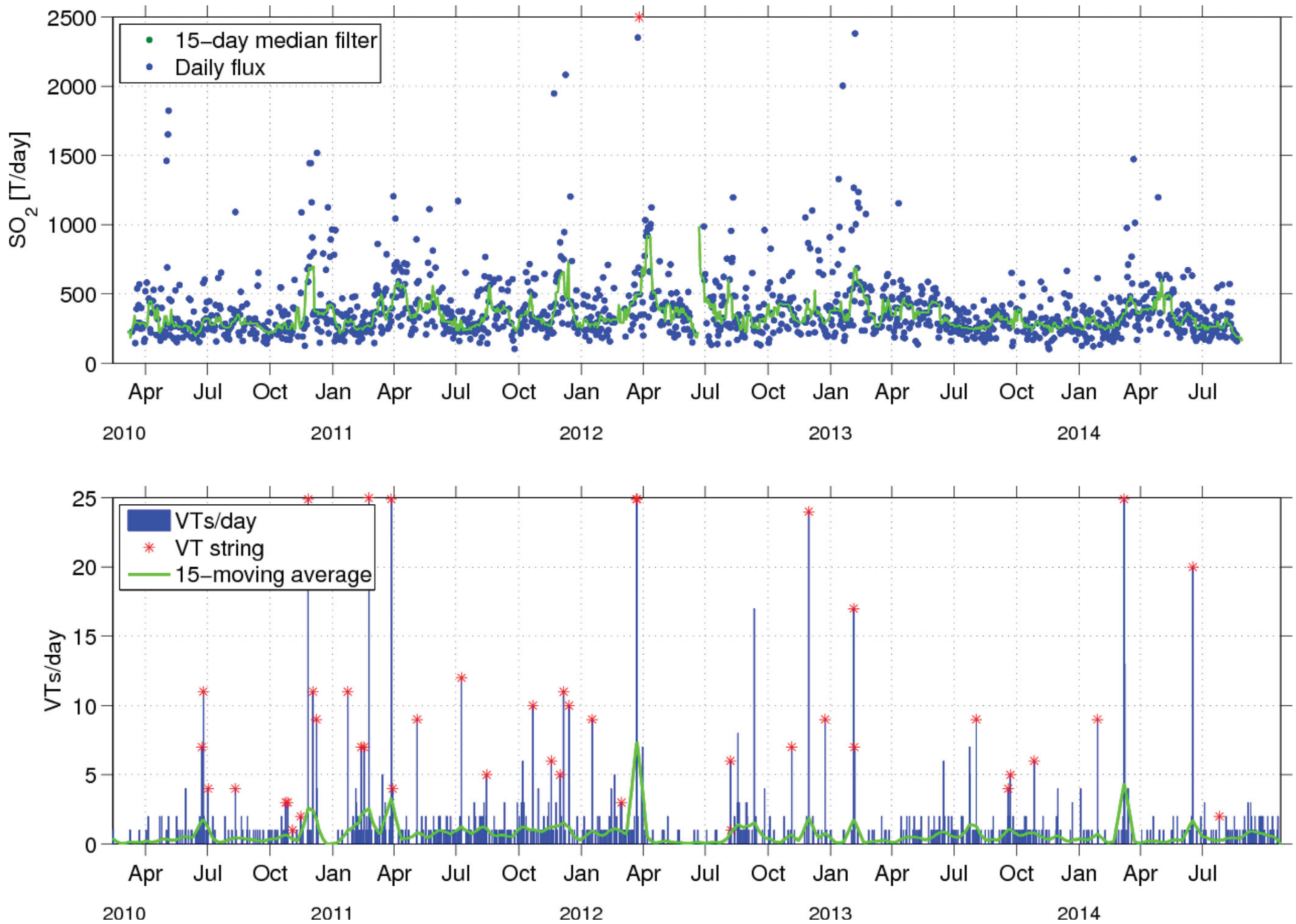

Figure 3. (top) SO_2 flux and daily count of volcano-tectonic earthquakes (VTs) since April 2010. A 15 day median moving filter has been applied to both data sets to indicate broad trends. Marked spikes in SO_2 and VT rate coincide and correspond to VT strings and associated pulses in SO_2 flux described in the text.

seismic events increase several fold. Gas bursts of this kind were also recognized from 2007 onward. Some of the bursts of SO_2 are also manifested by formation of surface fissures and ash venting. Since 2010 several high temperature, incandescent fumaroles have been observed on the northern part of the dome, mostly clustered around the dome collapse scar of February 2010 (Figure 4). These fumaroles have remained fixed, with temperatures of up to 610°C (Table 1) and there is no detectable cooling over time. Observations from three borehole strain meters [*Hautmann et al.*, 2014] indicate that the deformation associated with some of the larger VT strings and gas bursts characteristically lasts several tens of minutes and involves marked pressure changes in the shallow conduit system ($<$ 5 km), the shallow magma chamber (5–7 km depth) and a midcrustal magma chamber ($>$ 10 km) [*Hautmann et al.*, 2014]. The strain data suggest accumulation of magmatic fluids in the shallow conduit system accompanying deflation of the shallow magma chamber followed by deflation of the shallow conduit system during the gas bursts. On one occasion (23 March 2012) deflation was followed by inflation of the deeper magma chamber at 10 km or more depth. Estimates of overpressure [*Hautmann et al.*, 2014] are quite low (of order 1 to 2 MPa).

Taken together these observations indicate that the S-rich magmatic fluids are being released from depth (at least to 10 km), are spatially and temporarily decoupled from the parental magmas, and can be released without coeruption of andesite magma. Gas compositions recorded during pauses and periods of lava dome growth also support the notion that sulfur-rich gases are deeply derived. Large fluctuations in HCl/SO_2 have been documented, with high ratios during periods of dome growth and low ratios in the periods of no dome growth [*Edmonds et al.*, 2001; *Edmonds*, 2008]. The increase in HCl/SO_2 relates to increases in HCl flux while SO_2 flux remains unchanged. The chemistry of gas released from magmas is a sensitive function of degassing pressure, magma composition and degassing history [*Lesne et al.*, 2011; *Fiege et al.*,

Figure 4. Incandescent fumaroles on the lava dome in long-exposure photograph taken from MVO, 4 September 2011. The location of the fumaroles remain unchanged to November 2014 and there has been no sign of cooling.

2014]. Carbon dioxide has the lowest solubility in silicate melts and melts typically become vapour-saturated at low to midcrustal depths, depending on the initial melt CO_2 content, giving rise to exsolved fluids [*Dixon and Stolper*, 1995; *Blundy et al*., 2010]. H_2O and sulfur species (H_2S and SO_2) partition into the vapour phase. Fluid-melt Nernst partition coefficients ($D_S^{\frac{f}{m}}$) for total sulfur (i.e., $H_2S + SO_2$) are sensitive to pressure melt composition and redox state. In the pressure range 50–400 MPa $D_S^{\frac{f}{m}}$ under oxidized conditions (fO_2>NNO) are in the range 50–400 for andesites [*Fiege et al*., 2014] and 7–80 for basalts [*Lesne et al*., 2011] with the highest values corresponding to the lowest pressures. For haplogranite at 200 MPa [*Keppler*, 1999, 2010] gives $D_S^{\frac{f}{m}}=47\pm4$. At more reduced conditions $D_S^{\frac{f}{m}}$ increases significantly. For example, an increase from 47 to 468 as fO_2 is reduced from NNO to CoCoO (approximately 2 log units lower than NNO) has been reported [*Keppler*, 1999, 2010].

At a given fO_2 the degassing behavior of S is controlled strongly by pressure and the total exsolved fluid fraction [*Keppler*, 2010]. The latter is controlled principally by the initial H_2O content, as this is the dominant magmatic volatile species. H_2O solubility is strongly pressure dependent. As magmas ascend and degas, so H_2O exsolves from the melt (Figure 5a) and the fluid fraction increases, sequestering S to an ever greater extent because of the increase in $D_S^{\frac{f}{m}}$ as pressure drops [*Witham et al*., 2012]. The result is a strongly sigmoidal variation in S content of the melt as pressure falls, leading to an abrupt loss of sulfur from the melt over a small pressure range, as demonstrated by the experimental data (Figure 5b) on andesite [*Fiege et al*., 2014] and basalt [*Lesne et al*., 2011]. The behavior of S in andesitic melts differs from that in basaltic melts with the onset of sulfur loss from the melt taking place at slightly higher pressure (200 MPa) for andesite compared to basalt (150 MPa), although the experiments of *Fiege et al*. [2014] lack the resolution to quantify this difference. The principal controls on sulfur degassing are $D_S^{\frac{f}{m}}$ and the exsolved gas fraction, which is in turn related to the initial H_2O content. The parental basaltic andesites on Montserrat are estimated to have contained at least 9 wt% H_2O initially [*Edmonds et al*., 2014]. This is somewhat higher that the values used in the experiments shown in Figure 5 (3−7 wt%), which will serve to drive the maximum sulfur release to higher pressures.

Table 1. Fumarole Temperatures Recorded in Thermal Images in the Period 1 October 2013 to 31 March 2014

Location	Temperature (°C)	Last Image
Gas Vent, 2010 collapse scar	390–580	25 March 2014
Headwall, 2010 collapse scar	271–618	25 March 2014
23 March 2012 crater	57–80	25 March 2014
Tar River	217–429	25 March 2014
Summit 2006–2007 dome	619	11 March 2011
E and N flank of Galway's Mtn	60–84	
South flank of dome	334	11 March 2011

The strong partitioning of sulfur into vapour results in vapour-saturated melts containing low concentrations of sulfur, a feature common in arc intermediate magmas [*Wallace*, 2001; *Wallace and Edmonds*, 2011], with most of the sulfur mass being

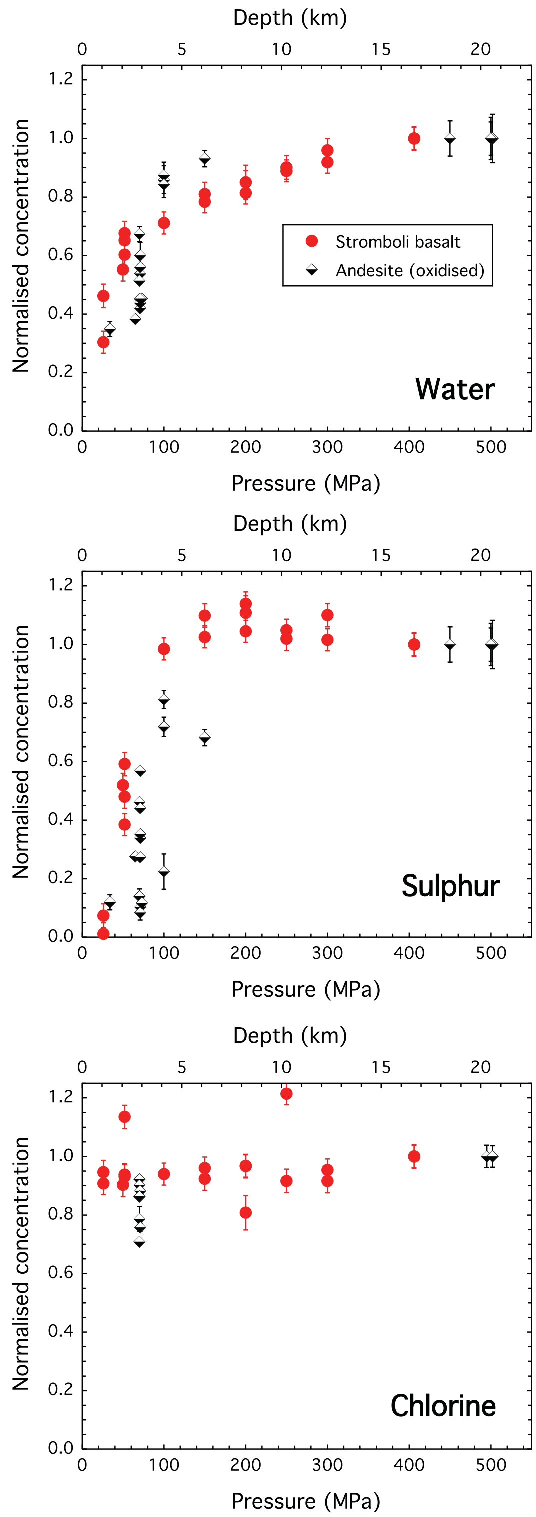

Figure 5. Evolution of dissolved volatile contents of quenched melt from experiments on andesite and basalt under relatively oxidized (>1 log unit above NNO) for (a) H_2O, (b) total sulfur (SO_2+H_2S), and (c) chlorine. Data are plotted normalized to the volatile content of the highest pressure experiment on the same bulk composition. Error bars are 1 s.d. propagated from uncertainties on starting and glass volatile contents. Andesite data at 1030°C from *Fiege et al.* [2014]; Stromboli basalt data at 1150°C from *Lesne et al.* [2011]. At these temperatures crystallization is negligible at all pressures. The initial H_2O content is 4–8 wt% for the andesites and 3 wt% for the basalts. Note the abrupt drop in melt S contents below ~150 MPa, but persistence of Cl contents to very low pressure. In plotting the andesite data no account has been taken of the kinetics of degassing, which exerts a second order effect on the behavior of S and Cl [*Fiege et al.*, 2014]. The depths on the upper axis have been converted from pressure using a density of 2500 kg m^{-3}, and are for illustrative purposes only.

present in an exsolved fluid phase. In contrast to sulfur, the partitioning behavior of chlorine is heavily dependent on melt composition, with little pressure dependency [*Webster*, 1992]. For typical hydrous arc magmas chlorine partitions into volcanic vapour much less strongly, $D_{Cl}^{\frac{f}{m}} \leq 5$ for basalts [*Lesne et al*., 2011] and <10 for andesites [*Fiege et al*., 2014]. Consequently, hydrous silicate melts retain their Cl contents until low pressures (Figure 5c) such that most of the HCl gas by mass being generated in the upper parts of the conduit system and during eruption itself [*Edmonds et al*., 2001]. It is evident from Figures 5b and 5c that Cl/S ratios of volcanic gases are sensitive indicators of degassing depths, with higher ratios signifying more shallow gas release.

At SHV gaseous Cl/S ratios increase sharply at the onset of lava extrusion, remaining high throughout eruptive periods [*Edmonds et al*., 2001; *Oppenheimer et al*., 2002; *Christopher et al*., 2010]. These observations reflect that the chlorine is sourced from ascending and decompressing andesite as it exsolves copious quantities of H_2O, with chlorine partitioning into the H_2O-rich vapour, resulting in large fluxes of H_2O and HCl-rich gases that are coupled to the erupted magma flux. Gases emitted during eruptive periods are therefore mixtures between deep, sulfur-rich gases, and shallow-derived H_2O and HCl-rich gases. Between eruptive periods, the deep-derived and typically S-rich gas dominates emissions and Cl/S ratios are low. At pressures of $\geq$10 MPa sulfurous magmatic fluids that are rich in H_2O contain both oxidized (SO_2) and reduced (H_2S) sulfur species in subequal proportions for most reasonable arc magma redox states [*Burgisser and Scaillet*, 2007] and it has been shown that SHV is no exception. Gas measurements (by Multigas) in 2008 to 2010 showed that SO_2 and H_2S were approximately equimolar [*Edmonds et al*., 2010] and it is likely that the equilibrium between these species acts as a dominant redox buffer in the gas phase [*Giggenbach*, 1987]. At pressures lower than $\sim$10 MPa the proportion of H_2S relative to SO_2 decreases rapidly due to homogeneous equilibria involving H_2O that produce H_2 [*Burgisser and Scaillet*, 2007].

There is increasing evidence that the SHV magmatic system extends into the middle crust and possibly even deeper. Ground deformation documented by GPS and strain meters indicates a magma system that extends from depths of 5 km to as much as 15 km [*Voight et al*., 2006; *Elsworth et al*., 2008, *Hautmann et al*., 2010, 2014]. Ground deformation [*Gottsmann and Odbert*, 2014], seismic tomography [*Paulatto et al*., 2012], thermal modeling [*Annen et al*., 2014] and strain data [*Hautmann et al*., 2014] indicate that the system is well connected. In fact, these new data suggest that a single, contiguous magmatic system might be a more appropriate model than the two discrete chambers previously postulated [*Elsworth et al*., 2008]. At the scale of seismic resolution no large melt-rich magma body can be discerned, suggesting that the magma system is in a dominantly crystal-rich or mushy state [*Paulatto et al*., 2012]. Melt-rich lenses, if present, are below the spatial resolution of the seismic imaging techniques. The average proportion of melt calculated from the P-wave velocity reduction is dependent on the geometry of the melt pockets, but for reasonable geometries is no more than 12 vol% in a mush region 4–6 km in diameter. Thermal modeling [*Paulatto et al*., 2012; *Annen et al*., 2014] shows that the seismic wave velocity structure can be reconciled with a smaller mush region with melt fractions of up to 35 vol%, consistent with the very high crystal content of the erupted andesite.

Petrological and geochemical data also support a vertically extensive magmatic system from 5 km to at least 10 km [*Edmonds et al*., 2014]. Phenocryst mineral assemblages and the compositions of most melt inclusions indicate that the SHV andesite magma assembled at low-pressure at the top of the magma system at 5 km [*Barclay et al*., 1998; *Edmonds et al*., 2001; *Rutherford and Devine*, 2003]. However, there is evidence of magma residence and crystallization at greater depths, including some plagioclase and pyroxene-hosted melt inclusions with $>$6 wt% H_2O as well as water contents of orthopyroxene that indicate melts with as much as 9% H_2O and depths of crystallization $>$10 km [*Humphreys et al*., 2009; *Edmonds et al*., 2014]. Finally, the andesite magma shows the signature of extensive amphibole fractionation [*Zellmer et al*., 2003a; *Christopher et al*., 2014b] at depths well below the shallow magma chamber.

Magma mixing is conspicuous in SHV andesites and is manifested by mafic inclusions with complex mixed phenocryst assemblages [*Murphy et al*., 2000; *Plail et al*., 2014] and cryptic mixing of crystals derived from mafic magma into the andesite groundmass [*Humphreys et al*., 2009]. Nonerupted mafic magma has also been inferred to be the source of the SO_2 which, in keeping with many arc volcanoes [*Wallace*, 2005], is greatly in excess of the amount of sulfur that could originate from the erupted andesite by a factor of at least 10 [*Edmonds et al*., 2001; *Christopher et al*., 2010]. The depth of mixing within the vertically extensive magma system is not well constrained. The mafic inclusions range from evolved basalt to basaltic andesite in composition with Mg numbers indicative of significant fractionation of a primitive basalt prior to mixing with the andesite magma [*Zellmer et al*., 2003a]. This petrogenesis indicates derivation of the mafic magmas from mantle-derived parent basalt at deeper levels in the crust than the observed magma system (5–15 km depth). Very water-rich and vapour saturated andesite magmas were resident in the middle crust [*Edmonds et al*. 2014] and could be an additional source for some of the excess sulfur.

Taken together, the observations suggest an extensive trans-crustal magmatic mush system extending from about 5 km depth into the middle crust and plausibly into the lower crust. The geometry of the magma system is constrained by geophysics, geodesy, thermal modeling and degassing behavior, while its history is constrained by stratigraphic, geochronological, geochemical and phenocryst zoning data. SHV has a history of short-lived dome eruptions, which alternate with long periods of dormancy. Large volume dome eruptions of compositionally indistinguishable andesite [*Zellmer et al*., 2003a] occurred at around 24, 18, 12, 6, and 2 ka with the small volume Castle Peak dome erupted at about 0.5 ka [*Harford et al*., 2002; *Trofimovs et al*., 2013]. There is no evidence for eruptive activity in the time periods between these events. Occasional magma or magmatic fluid transfer into the upper crust causes periods of unrest without eruption during the dormant periods, such as the seismic unrest periods of 1896–1897, 1933–1937, and 1966–1967 [*Wadge et al*., 2014]. These restless episodes are consistent with internal modifications to the mush system, such as melt and gas migration from one level to another, but without discharge to the surface.

Uranium-series isotope data indicate long crustal residence times of the andesite magma (10^5 years) yet data on zoning of trace elements in plagioclase phenocrysts suggests short magma residence times (10^2-10^3 years) in the shallow magma chamber at $\sim$ 5 km depth [*Zellmer et al*., 2003a; 2003b]. Thermal modeling [*Paulatto et al*., 2012; *Annen et al*., 2014] of magma chamber assembly in the upper crust requires time scales of a few thousand years to be consistent with seismic tomography and the short phenocryst residence times. These shorter time scales can be reconciled with the U-series time scales if the latter pertain to generation of differentiated melts in deeper parts of the system, whereas the former are indicative of preeruptive magma storage at higher levels of the system. Magmas formed by deep-seated differentiation ascend periodically to the shallow storage region, where they undergo further crystallization and magma mixing, without significant additional chemical differentiation. Thus the shallow storage region can be viewed as a relatively short-lived phenomenon where magmas assemble shortly prior to eruption. In keeping with *Annen et al*. [2006] we consider the chemical character of magmas to be developed at depth in the crust, whereas their textural characteristics are acquired in shallow, preeruptive storage regions. The long and short crystal residence time scales, respectively, reflect this dichotomy. Overall these observations imply active transfer of the fractionated andesite magmas from the middle crust (and below) to the shallow chamber during periods of dormancy that typically last thousands of years. Magma transfer may be manifested geophysically, but need have no surface expression in the form of magma or gas discharge.

3. Magmatic System Model

We propose a new model for the SHV system with an emphasis on the role of deeply generated volatiles, the idea of a mature magma system with magma and magmatic fluid accumulation and generation at multiple levels, and destabilization of the system leading eventually to a major eruption. Our model builds on: models of multiple magma chambers and igneous mush systems [*Hildreth*, 2004; *Marsh*, 2004; *van der Zwan et al*., 2013; *Walker et al*., 2013; *Jaxybulatov et al*., 2014; *Cashman and Giordano*, 2014]; concepts of polybaric differentiation [*Pichavant et al*., 2002; *Grove et al*., 2005; *Ulmer*, 2007; *Gertisser et al*., 2012; *Almeev et al*., 2013; *Hamada et al*., 2014; *Melekhova et al*., 2015]; separation of melts and fluids from low porosity source regions into inherently unstable melt-rich layers or lenses [*Solano et al*., 2012; *Connolly and Podladchikov*, 2013]; and segregation of magmatic fluids from underplated magmas including basalt and andesite derived by fractionation of basalt [*Bachmann and Bergantz*, 2006].

Figure 6 shows a schematic depiction of the envisaged trans-crustal magmatic mush system. During periods of dormancy (Figure 6a) the magma system extends throughout the crust and is mostly in the partially molten state, and stable. The system evolves slowly and is characterized by a steady flux of primitive mafic magma at the base and segregation of fractionated melts within the partially molten rock due to gravitational compaction, perhaps augmented by tectonic strain, to form multiple layers. The physics of layer production is a slow, creeping process governed by simple and pure shear [*Solano et al*., 2012; *Connolly and*

Podladchikov, 2013; *Shield et al*., 2014]. Multiple high-porosity melt layers may form within the same mush pile and migrate upward at different relative velocities. The melt in any of these layers will have a variety sources within the mush, providing a mechanism to mix together melts of different history and parentage. Migrating melts will evolve chemically as they react with crystals in the mush through which they pass [*Solano et al*., 2014]. Magmatic fluids will also segregate once the melts become volatile-saturated, either through crystallization ("second boiling") or decompression ("first boiling") or both. Second boiling could release large fluxes of volatiles over long time scales, particularly given that the total volume of melt contained within the mush underlying SHV is likely to exceed 40 km^3. Even small amounts of crystallization (10%) of a magma containing 250 ppm sulfur in the melt phase is likely to lead to significant sulfur release (2 Mt), which is roughly equal to the amount of sulfur that has been released during the current eruption. There is no evidence, however, that 10% of crystallization has taken place over the 15 years of the eruption, with erupted lavas having monotonous values of crystallinity close to the locking point [*Murphy et al*., 2000]. Trace element zoning of plagioclase indicates crystallization over decades to centuries [*Zellmer et al*., 2003b], so it is possible that second boiling fluids from the andesite could have been stored within the mush system. The composition of the magmatic volatiles released will be strongly depth dependent due to different solubilities of the key species (H_2O, CO_2, H_2S, SO_2, HCl) [*Dixon and Stolper*, 1995; *Zajacz et al*., 2011].

Magmatic fluids have radically different properties to melts (orders of magnitude lower viscosity and lower density) such that fluids have a strong tendency to separate and decouple from the melt from which they exsolved originally. During stable periods there is very slow decompression of melts so that if they are volatile-saturated fluids will exsolve and the pressure in the system can be expected to increase. However, this tendency may be counteracted by contraction of the magma upon crystallization and by volume expansion and deformation of surrounding ductile crust. Slow release of exsolving magmatic fluids can thus be expected at the top of the magmatic system. These fluids then interact with upper crustal hydrothermal systems to form fumaroles and hot springs with magmatic fluid components, as is observed in many dormant volcanoes.

The sill-like layers of melt and magmatic fluids that are predicted to develop are inherently unstable due to their buoyancy so that the system can reorganize, resulting in melt and fluid rising within in the system (Figure 6b) and amalgamating into larger magma bodies. This process contrasts with the background migration of melts through compaction described above, notably because it will operate on shorter time scales (perhaps months to years, or less, before eruption). A variety of instabilities have been recognized in two or three-phase systems when layers of buoyant melts and fluids segregate from a crystalline matrix and from each other, including Rayleigh-Taylor instabilities, fingering, porosity waves and shear bands [*Bremond D'Ars et al*., 1995; *Connolly and Podladchikov*, 2013; *Michaut et al*., 2013; *Shield et al*., 2014]. Periods of destabilization caused by these mechanisms need not lead to eruption, but may instead be the cause of volcanic unrest, as happened at SHV in 1896–1897, 1933–1937, and 1966–1967 [*Wadge et al*., 2014]. Since melts and magmatic fluids can decouple, destabilization events may involve predominantly melt or predominantly magmatic fluids or both. Volume and pressure changes related to upward transfer of melts and fluids will be largely related to the decompression of already segregated magmatic fluids and exsolution of volatiles in decompressing, vapor-saturated melts. Unlike passive gas exsolution resulting from background melt migration, exsolution triggered by abrupt rise of melt lenses will result in much higher volatile fluxes. Vapor-saturated, decompressing melts will crystallize (as a result of degassing) to form suspended crystals, exsolving further volatiles. Thus amalgamated melt layers can readily evolve into crystal-bearing magma chambers.

During eruptive phases (Figure 6c) the destabilization of melt and fluid layers generates sufficiently large overpressures that magma at the top of the system can connect to the surface and an eruption ensues. Connection to the low-pressure surface environment further enhances instability in the underlying mush and associated exsolution through pressure decrease. We suggest that this general process, with both melt and fluids derived from throughout the magmatic mush, accounts for the observations of SHV eruption in general, and the coeruptive release of large quantities of magmatic gas in particular. This mechanism has the potential to supply substantial volumes of deep magmatic gas rich in SO_2 (and CO_2) to the surface, in quantities that are out of proportion to the volumes of magma erupted. Thus mush destabilization, rather than mafic magma injection, can explain the sulfur-excess problem of arc volcanoes [*Wallace*, 2005].

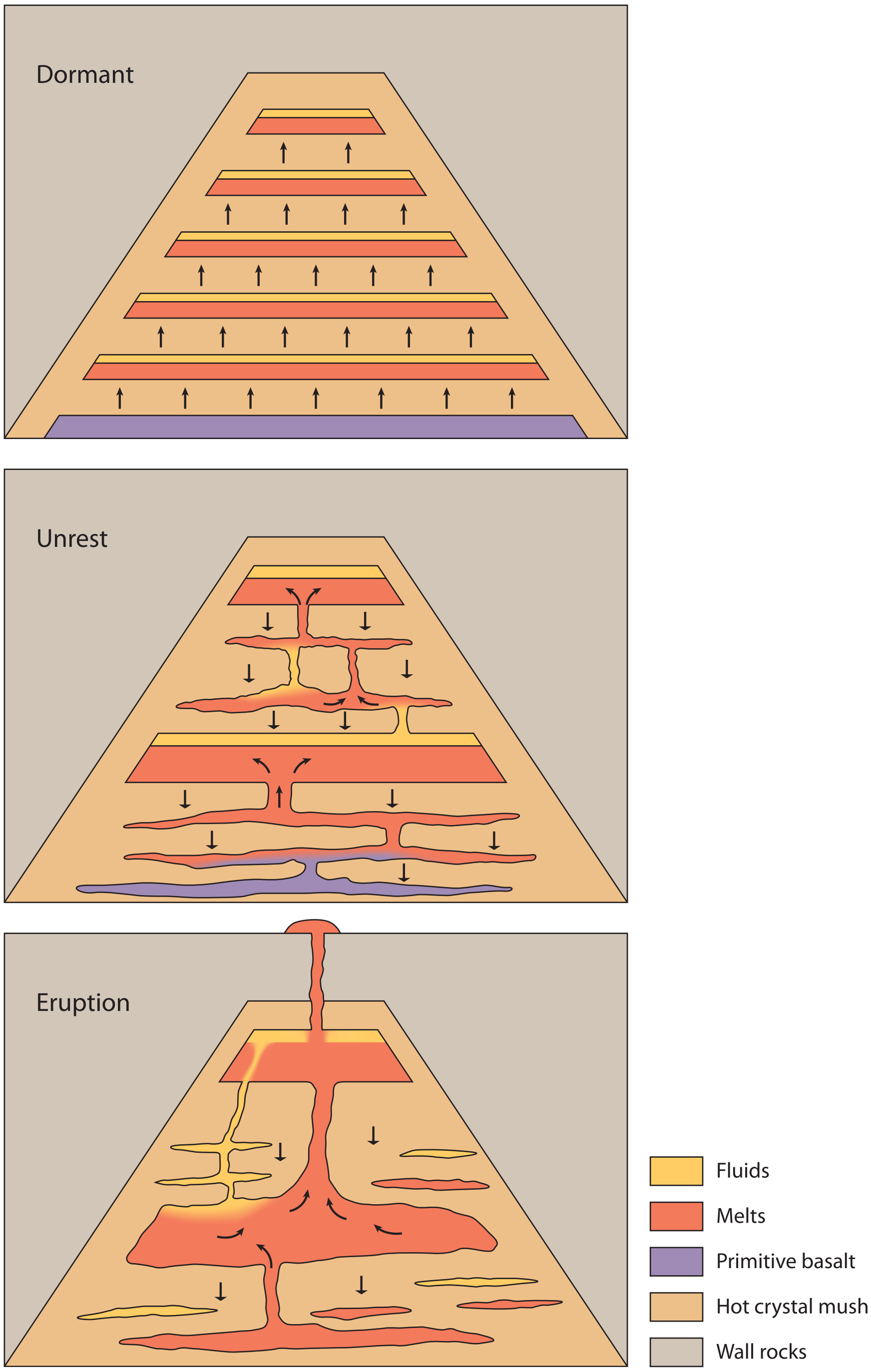

Figure 6. Schematic cartoons of trans-crustal magmatic mush system invoked beneath SHV in three principal states. (top) Dormant state: primitive basalt magma fluxes into the base of the crust; layers of fractionated melt and magmatic fluids slowly segregate from the igneous system above solidus temperatures to form layers. Primitive basaltic magma (shown schematically as a purple layer) fluxes from the mantle into the crust at the base of the system. (middle) Unrest state: layers of melt and magmatic fluids connect and move upward while the igneous, crystal-dominated framework (mush) collapses downward; the melts layers amalgamate to form magma chambers at physically favorable depths; decompression of melts and magmatic fluids results in crystallization, increases in magma and fluid pressure but insufficient for surface eruption. (bottom) Major destabilization of the trans-crustal layered igneous system leads to eruption; melts and magmatic fluids decompress causing increases in magma pressure; magmas, melts and magmatic fluids from different crustal depths can mix together; eruption has a positive feedback on the system increasing its instability. Tectonic processes may further enhance instability. Note that magmatic gas has three different sources: magmatic fluids generated and segregated from the igneous system in dormant periods (old gas), magmatic fluids generated by exsolution during melt and magma decompression during periods of instability, and gas generated from magma as it erupts at low pressure.

4. Implications

Our destabilization model has some major implications for interpreting volcano behavior, which we discuss in the context of the SHV as a case study. The widespread evidence for magma mixing being associated with many volcanic eruptions has led to the idea that mixing itself may trigger eruptions [*Sparks et al*., 1977]. The same model has been applied to explain the current SHV eruption [*Murphy et al*., 2000; *Christopher et al*., 2010]. In this model deeper more mafic magma ascends and disrupts more silicic magma residing in a shallow chamber, liberating volatiles and increasing magma pressure due to replenishment. As mafic magmas have higher liquidus temperatures than felsic magmas, mixing triggers crystallization of the mafic magma [*Sparks and Marshall*, 1986]. If these magmas are initially volatile-rich, crystallization will also trigger gas exsolution and an increase in pressure [*Tait et al*., 1989; *Huppert and Woods*, 2002]. In this scenario excess SO_2, as exemplified by SHV, has been attributed to mafic magma replenishment [*Christopher et al*., 2010]. Conversely in the destabilization model introduced here, magma mixing and upward transfer of mafic magma from deeper levels can be a consequence, rather than a cause, of eruption, although many of the same physical effects that contribute to eruption triggering will be obtained. It remains plausible that system destabilization may be initiated in the deepest part of the system, perhaps resulting from an influx of primitive basaltic magma (Figure 6a), causing ascent of deep mafic magma to the shallower parts of the system. However, instability could equally develop first in the shallower regions of the system as a consequence of large contrasts in bulk density between magma layers and surrounding mush caused by volatile exsolution. Downward propagation of instabilities would allow fractionated mafic magmas from the lower crust to rise and mix with more evolved, but cogenetic, silicic magmas. In the latter scenario the eruption is triggered by top-down destabilization, and magma mixing is the consequence rather than the cause of eruption. Instabilities propagating into the crust from the top down have been invoked for Icelandic volcanic systems, with progressive magma flow at depth responding to pressure changes in the shallow system [*Tarasewicz et al*., 2012]. The role of tectonics in enhancing, or even triggering, destabilization [e.g., *Davis et al*., 2007; *Gottsmann et al*., 2009] would benefit from closer study.

The replenishment model of eruption triggering has emphasized the role of incoming deeper magma in increasing the pressure in a shallow chamber [e.g., *Barmin et al*., 2002]. However, in the destabilization model, magma or melt transfer to shallower levels may not have a major role because upward transfer of melt (and any suspended crystals) is compensated by downward transfer of hot ductile, melt-poor crystal mush. There is therefore no net volume change related to the melt and crystal components of ascending magma. Any associated volume changes are almost entirely related to the decompression of exsolved and exsolving magmatic volatiles. Thus the common interpretation of preeruptive pressurization of shallow magma chambers prior to eruption in terms of influx of new magma may be misleading. We suggest instead that the relevant flux responsible for chamber pressurization and ground deformation is not the magma flux but the exsolved volatile flux. Degassing can account for deflation in several volcanoes [*Girona et al*., 2014].

The decoupling of magma and magmatic volatiles demonstrated at SHV has been explained as the consequence of the effects of variable solubility and partitioning of different volatile species and degassing systematics (notably CO_2, H_2O, H_2S, SO_2 and Cl) combined with their ability to segregate from their source melts in deep, mature igneous systems. If the processes of gas exsolution and segregation take place during long periods of dormancy, then significant amounts of exsolved volatiles could be generated and stored in the crust over a wide range of depths, perhaps in several discrete volatile-rich layers (Figure 4). Any or all of these stored volatile layers can then be released in an eruption or sequence of eruptions, or in periods of unrest without eruption. Volatile movements unaccompanied by magma can thus initiate unrest and eruption. A variant of this idea is the concept of gas sparging [*Bachmann and Bergantz*, 2006] in which volatile segregation from deeply emplaced mafic magma combined with heat transfer remobilizes silicic mushes at shallower levels in the system.

Magmatic volatiles can thus originate in different ways, each involving volatiles of contrasted chemistry: from shallow degassing (H_2O and HCl with minor S volatiles) of silicic magma chambers; from ascent of deeper more mafic magmas during system disturbances during eruptions (H_2O, SO_2, H_2S); from fractionation of wet basalt to form andesite magma that undergoes further crystallization and volatile exsolution in the middle crust (also H_2O, SO_2, H_2S); and by segregation and long term storage of deep, poorly soluble volatiles (CO_2, SO_2, H_2S) exsolved from intermediate to silicic melts that form by fractionation of basalt in the lower and middle crust [*Annen et al*., 2006]. The segregation and storage may occur via slow, background migration of volatile-saturated melts, or via abrupt melt lens coalescence events associated with system destablization. The release of volatiles from all of these sources can occur on very different time scales as volatiles become temporarily, spatially and chemically decoupled from magma.

Observations at SHV suggest that some of the volatiles are transferred from at least 10 km depth [*Hautmann et al*., 2014]. In that case some of the excess SO_2 may be old, predating the current eruption, and deeply stored. These old volatiles may be evolved from the middle crustal andesite reservoir or from deeper fractionating basalt. They mixed with vapour derived from mafic magma during shallow chamber magma mixing. An alternative interpretation then of the SHV observations is that magmatic volatiles have slowly accumulated throughout the crustal system over the last few thousand years and are now being released as a consequence of the proposed system destabilization and the eruption itself creating a pathway to the Earth surface. The continued high flux of SO_2 that has followed the last dome extrusion in 2010 can be explained by deep (>5 km depth) magmatic volatiles continuing to flux through the upper parts of the magmatic system.

The behavior of volatile species in trans-crustal magmatic mush systems also has implications for hydrothermal ore formation associated with arc volcanoes. Porphyry copper deposits are widely viewed as giant sulfur anomalies [*Hunt*, 1977] and are therefore likely to be related in origin to the excess sulfur phenomenon of arc volcanoes [*Wallace*, 2005]. Similarly, most porphyry copper deposits contain fluid inclusions of high salinity and metal content [*Bodnar et al*., 2014] testifying to the importance of chlorine-rich fluids in mineralization. *Blundy et al*. [2015] have proposed that interaction of deep-derived S-rich gases with shallow-stored metalliferous brines is a fundamental ore forming process in porphyry copper systems. Thus the mechanisms by which fluids (including brines) move through magmatic mushes and accumulate may be of economic as well as volcanological importance. It is striking that at SHV recent tephras contain Cu-Fe sulphide and sulphate (anhydrite) minerals [*Plail et al*., 2014] consistent with the gas-brine model of *Blundy et al*. [2015] and hinting that ore-forming processes may be ongoing beneath SHV today, in part due to the underlying magmatic mush that allows fluids and melts to migrate and accumulate separately for extended periods, before rapidly juxtaposing them during destabilization events. The difference between a volcano like SHV and a copper porphyry system may be the extent to which the latter fixes deeply sourced sulfur in shallow crustal mineralized zones through reaction with brines, whereas the former vents much of the sulfur to the surface.

5. Conclusions

The rich data set on the geological history and eruption phenomena at Soufrière Hills Volcano, Montserrat, during the 1995 to 2015 period provides an opportunity to reassess models of crustal magmatic systems in the context of volcanism. Observations of magmatic volatiles, in particular SO_2, together with data on solubilities of different volatiles [*Lesne et al*., 2011; *Fiege et al*., 2014] and geophysical data [*Hautmann et al*., 2014], provide evidence that volatiles become decoupled from magmas and that some volatiles are deeply sourced (perhaps from the midcrust). Geochemical, petrological and geophysical observations provide evidence for a magmatic system that extends down to the lower crust and perhaps the uppermost mantle. It also seems that the magma storage system down to at least10 km depth is well connected and contiguous. The recent geological history of SHV, together with modeling of chamber formation, indicates that this system tends to be dormant for typical periods of several thousand years and is interrupted by intermittent eruptions that last a few years or a decade or two. Upward migration of melt and gas by matrix compaction may occur continuously throughout periods of dormancy with little or no surface manifestation. Occasional, noneruptive periods of unrest lasting a few years and marked by increased seismicity and enhanced fumarolic activity can occur during dormant periods and could be related to migration and amalgamation of melt and fluid lenses.

Critically, observations at SHV can be explained by processes operating within a large trans-crustal magmatic mush system. During dormant periods primitive basalt magma is fluxed from the mantle into the crust where cooling leads to fractionation in a lower crustal hot zone [*Annen et al*., 2006]. Modeling of the dynamics of melt segregation in mushes and buoyancy-induced instabilities [e.g., *Bremond D'Ars et al*., 1995; *Connolly and Podladchikov*, 2013] indicate that buoyant fractionated melts segregate due to deformation of hot melt-bearing intrusive rocks to form layers, which eventually become unstable and produce melt-rich layers that ascend through the crustal magmatic system and amalgamate to form magma bodies where further crystallization and magmatic fluid exsolution occur. Magmatic fluids can exsolve and segregate from the parental melts at all depths and thereby become decoupled from magma. Unrest during dormant periods is attributed to destabilization of magma, melt and magmatic fluid layers (Figure 6b); decoupling of melts and fluids allows the possibility of some unrest being related to fluid movements alone. Eruptions are attributed to major periods of instability in which accumulation of magma and fluid at the top of the system leads to overpressures large enough for magma to reach the Earth's surface. Magmatic volatiles can thus originate in three different ways during eruptions: from shallow degassing of andesite magma during extrusion; from ascent of deeper more mafic magmas due to magmatic system instability; and by segregation and long term storage of deep, low solubility volatiles in the lower and middle crust.

We propose that the observed excess SO_2 at SHV is a consequence of both discharge from mafic magma replenishment and escape of magmatic fluids segregated from parent melts and accumulated throughout the trans-crustal magmatic mush system during dormant periods. Magmatic volatiles are the main source of overpressure in the shallow parts of the magma system, leading to unrest and eruption.

Our new model has important implications for the interpretation of volcano monitoring data at andesitic volcanoes. Specifically, large SO_2 gas fluxes might not necessarily be linked to shallow magma flow, transport or eruption processes and might instead be utilized, in conjunction with deformation records, to understand the fluid capacity of the shallow crust for driving destabilization and eruptive activity. The model also has implications for the formation of hydrothermal ore deposits, notably porphyry copper systems where interaction between different fluids (metalliferous brines and S-rich gases) may be a key ore-forming process [*Blundy et al*., 2015].

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