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Volcanic gas emissions from Soufrière Hills Volcano, Montserrat 1995–2009, with implications for mafic magma supply and degassing

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[1] Volcanic gas emissions illustrate a complex volatile budget for Soufrière Hills Volcano, Montserrat. Fluxes of sulphur dioxide, carbon dioxide, hydrogen sulphide and hydrogen chloride (and probably water) are substantial from this arc volcano. Additional sources of volatiles in addition to the erupting andesite are required to satisfy the mass balance defined by gas emissions and petrological constraints. Mafic magma is intruded at depths of >10 km, supplying volatiles by quenching, crystallising and vesiculating at the andesite-mafic magma interface. Sulphur, carbon dioxide and other volatiles supplied to the system migrate to the surface and their fluxes remain high while mafic recharge at depth is sustained. Decompression degassing of porphyritic andesite supplies chlorine and water to the gas plume. Chlorine partitions into vapour from the melt during periods of active extrusion, making the HCl/SO_2 ratio of volcanic gases a reliable indicator of the onset of eruptive activity. Studies of this volatile budget highlight the role of deep degassing and vapour production at arc volcanoes, which are important mechanisms for driving the eruption due to its effects on magma buoyancy and the advection of heat through the system. **Citation:** Christopher, T., M. Edmonds, M. C. S. Humphreys, and R. A. Herd (2010), Volcanic gas emissions from Soufrière Hills Volcano, Montserrat 1995–2009, with implications for mafic magma supply and degassing, *Geophys. Res. Lett.*, *37*, L00E04, doi:10.1029/2009GL041325.

1. Introduction

[2] The Soufrière Hills Volcano has been a prodigious emitter of volcanic gases since the onset of the current eruption in 1995. The magma, prior to eruption, is saturated with volatiles in an upper crustal magma reservoir [*Barclay et al.*, 1998]. The importance of volatile species in controlling the longevity and style of the eruption has been recognised through a number of important studies. The effects of H_2O degassing on the rheological properties of the magma have been shown to influence eruptive style [*Melnik and Sparks*, 1999; *Sparks et al.*, 2000], which ranges from lava dome-building to vulcanian explosive activity. Volcanic gas data have been instrumental in hazard assessment: the continued emission of SO_2 gases has been a reliable indicator of eruptive potential; and HCl/SO_2 molar ratios provide a useful confirmation of the onset and rate of andesite decompression and degassing at the start of eruptive episodes [*Edmonds et al.*, 2001]. The sustained emission of volcanic gases over the duration of the eruption [*Edmonds et al.*, 2003], combined with geophysical data indicating magma chamber recharge [*Elsworth et al.*, 2008] and petrological evidence for disequilibrium and heating of the erupting, porphyritic andesite (which is comprised of a rhyolitic interstitial melt [*Murphy et al.*, 2000; *Couch et al.*, 2001]) have led to a model of mafic magma recharge and degassing at depth, which is likely to have both triggered and sustained the current eruption. The mafic magma clearly mingles with the andesite to some degree and is erupted in the form of rounded enclaves of basaltic andesite bulk composition [*Humphreys et al.*, 2010; J. Barclay, Experimental phase equilibria constraints on pre-eruptive storage conditions of the Soufrière Hills magma, manuscript in preparation, 2009] and there is evidence that some of the microlites in the andesite are derived from this basaltic andesite [*Humphreys et al.*, 2009]. The basaltic andesite is not clearly related geochemically to the erupting rhyolitic melt [*Zellmer et al.*, 2003]. The proportions of CO_2 and H_2S gases relative to SO_2 in the volcanic plume were measured for the first time in 2008 during an eruptive pause and are consistent with the degassing of unerupted magma at depth [*Edmonds et al.*, 2010]. Given the longevity of the eruption and the new geophysical and petrological data showcased in this special issue and in other recent papers, which shed new light on the mafic magma recharge process and plumbing system, we here present complete time series for the major volatile species in volcanic gases and a summary of the volatile data for Soufrière Hills Volcano. We discuss their significance in terms of mafic magma supply and degassing processes and explore the evidence for the depth and form of the plumbing system derived from volatile data, outlining the significance for hazard assessment.

2. Methodology and Results

2.1. Sulphur Dioxide Emissions

[3] SO_2 fluxes at Soufrière Hills Volcano were measured using correlation spectroscopy during 1995–2001, whereby traverses were undertaken beneath the volcanic plume and the unique absorption spectrum for SO_2, using the background sky as a source of ultra-violet radiation (UV), was detected by the spectrometer. The mass of SO_2 in a column above the spectrometer was integrated across the traverse

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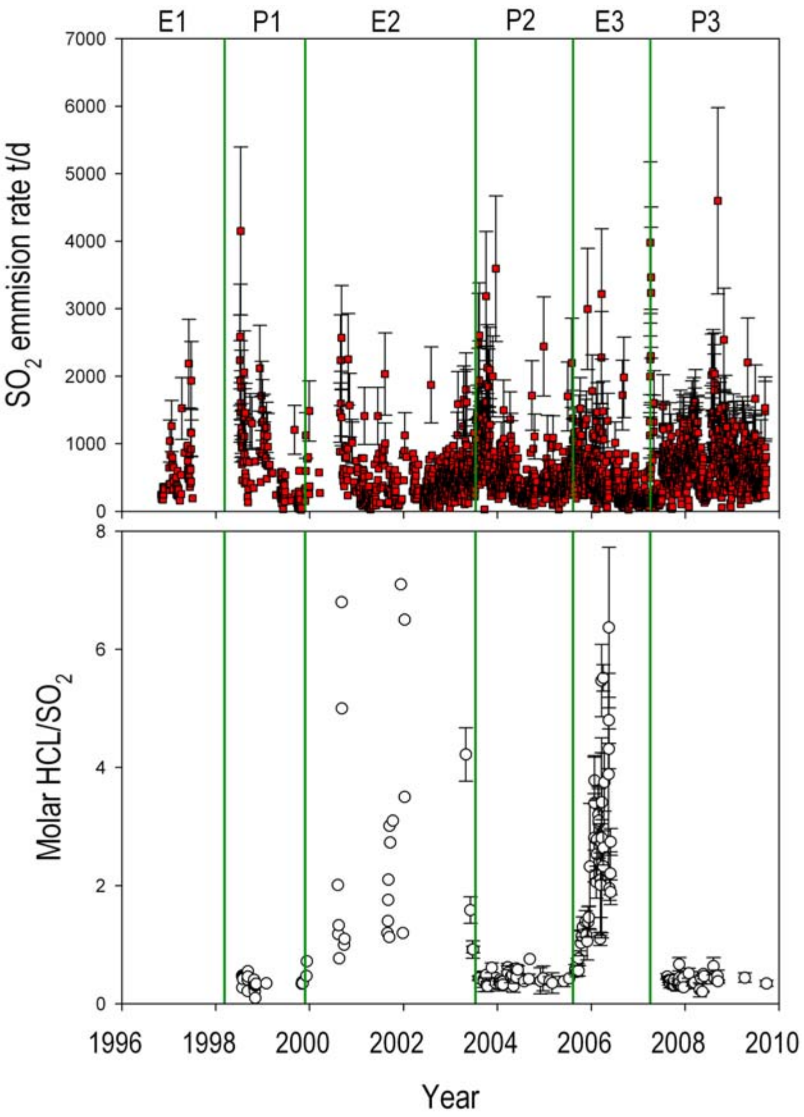

Figure 1. (top) SO_2 emission rate (in metric tonnes per day) 1995–2009 at Soufrière Hills Volcano, Montserrat. The mean emission rate for the entire eruption is 574 t/d. (bottom) Molar HCl/SO_2 in volcanic gases measured by FTIR from 1996 to 2009. E1, 2, 3: eruptive periods; P1, 2, 3: pauses in the eruption.

and the flux calculated by multiplying by the plume speed. In this way SO_2 flux measurements were carried out using a car, boat or helicopter as a platform. Since January 2002, SO_2 fluxes measured using a network of fixed UV spectrometers, utilising a different optical method for acquiring SO_2 column amounts: Differential Optical Absorption Spectroscopy (DOAS). The spectrometers receive UV light from the sky via an optical-mechanical system that scans the volcanic plume in two dimensions. The SO_2 column amounts are derived from the absorption spectrum, a function of the measured and background spectra, which is fitted to the known cross-section to derive a mass column amount. This is integrated over the scan and corrected for the scan geometry and multiplied by plume speed to calculate SO_2 flux [*Edmonds et al.*, 2003]. Since the development of this monitoring system, similar systems have been established elsewhere [e.g., *Salerno et al.*, 2009]. The daily SO_2 flux time series from 29 July 1995 to 30 December 2008 is shown in Figure 1. The scanning UV spectrometer network can measure the SO_2 flux ~85% of the time; during the remainder of the time the plume is either out of range of the spectrometers, below detection or ash attenuation prevents optical measurements. Practical concentration pathlength detection limits are on the order of 2–5 ppm.m which, when combined with typical plume speeds and dimensions, corresponds to fluxes of 30–80 t/d. Ash attenuation occurs during periods of heightened volcanic activity, which is of course often when SO_2 emissions are at their highest, which is a frustration, although satellite-based methods can potentially fill this gap (S. A. Carn and A. J. Prata, Explosive degassing at Soufrière Hills Volcano, Montserrat, submitted to *Geophysical Research Letters*, 2009).

2.2. Hydrogen Chloride Emissions

[4] Open path Fourier Transform Infra-Red (FTIR) spectroscopy has been used throughout the eruption to measure the molar HCl/SO_2 ratio, using the sun as a source of infrared radiation and utilising the unique absorption features

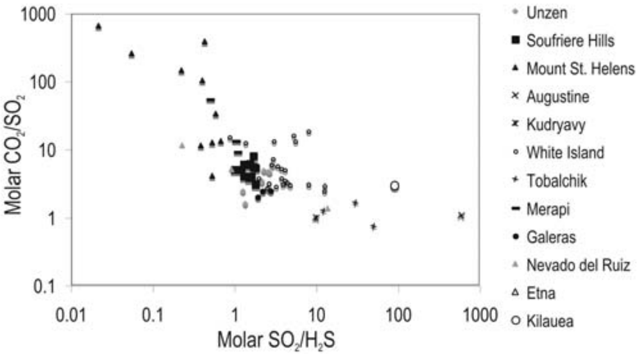

Figure 2. The proportions of CO_2, SO_2 and H_2S in the volcanic plume at Soufrière Hills Volcano, compared to other volcanic gas data [*Giggenbach*, 1975; *Menyailov and Nikitina*, 1980; *Gerlach*, 1982; *Giggenbach et al.*, 1990; *Alfaro and Zapata G.*, 1997; *Churakov et al.*, 2000; *Aiuppa et al.*, 2007; *McGee et al.*, 2008; *Ohba et al.*, 2008; *Edmonds et al.*, 2010, and references therein].

of these molecules in the acquired spectra, analysed using a retrieval code developed by M. Burton (INGV, Italy). A 20 cm Newtonian telescope is used with a MIDAC spectrometer, with spectra acquired using Autoquant at a sampling rate of 1 Hz, with 4 scans co-added to improve signal-to-noise. The molar HCl/SO_2 of volcanic gases has proved, during the early phases of the eruption, to be an excellent proxy for magma ascent, with little Cl degassing occurring during eruptive pauses, and Cl degassing beginning rapidly at the onset of andesite decompression, ascent and eruption, making this parameter an excellent monitoring tool [*Edmonds et al.*, 2001; *Oppenheimer et al.*, 2002]. The complete time series of FTIR-derived mean molar HCl/SO_2 ratios is shown in Figure 1 (bottom).

2.3. Carbon Dioxide Emissions

[5] Volcanic gases have been measured in a variety of other ways throughout the eruption. Gases were sampled directly from the lava dome during January 1996, prior to the onset of significant pyroclastic flow activity using Giggenbach bottle sampling and wet chemical analytical methods [*Hammouya et al.*, 1998]. More recently, CO_2, SO_2 and H_2S concentrations were measured in the plume using a non-dispersive infrared (NDIR) spectrometer and electrochemical sensor package (a Multi-GAS sensor) in July 2008 [*Edmonds et al.*, 2010]. The Multi-Gas sensor integrated a set of commercially-available sensors, including: a LI-840 NDIR closed-path spectrometer for CO_2 and two electrochemical sensors specific to SO_2 and H_2S. Figure 2 shows the gas composition data measured in July 2008 with data from a range of other volcanoes, which display a linear array ranging from CO_2-rich and H_2S-rich compositions to SO_2-rich and CO_2-poor compositions. These end-members correspond most commonly to either a) those dominated by hydrothermal systems (where SO_2 is scrubbed), and those which are primarily magmatic; or b) to high pressure- (which stabilises H_2S) and low pressure-equilibrated gases. Other variations in the gas compositions for the different volcanoes and settings are primarily due to differences in pre-eruptive volatile concentrations in the magma, oxygen fugacity and temperature. The Soufrière Hills gases have a composition very similar to those measured at Merapi (Indonesia) and Unzen (Japan) volcanoes, which are both intermediate-silicic arc volcanoes with similar eruptive styles.

3. Discussion

[6] The total output of sulphur from the volcano can be calculated by linear extrapolation between gaps in the SO_2 emission rate time series and by adding the sulphur contributed by discrete SO_2 releases caused by lava dome collapses and vulcanian explosions that have been measured by the TOMS and more recently OMI satellite-borne sensors (Carn et al., submitted manuscript, 2009). The contribution of sulphur from H_2S emissions can also be added if we assume that the molar SO_2/H_2S ratio measured in July 2008 [*Edmonds et al.*, 2010] is representative of the entire eruption.

[7] The cumulative S emission with time is shown in Figure 3. We can model this time series by calculating the amount of sulphur that would be supplied by considering two end-member degassing scenarios: a) complete degassing

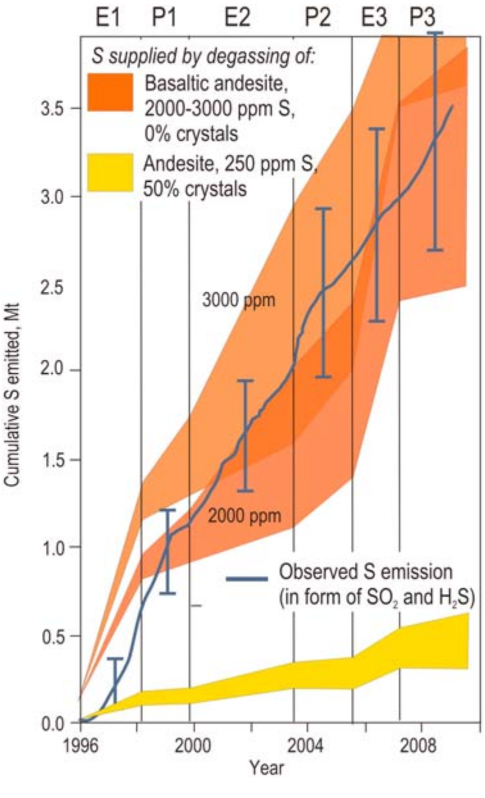

Figure 3. Models and observations of degassing of sulphur at Soufrière Hills Volcano, Montserrat. Observed cumulative sulphur emitted during the 1995–2009 eruption is shown in dark blue, with error bars at intervals. Orange shaded areas show the range in S emissions resulting from models of largely unerupted mafic magma degassing at depth, whereby mafic magma of basaltic andesite composition, containing 2000–3000 ppm sulphur, quenches against the andesite interface, vesiculating and crystallising. The yellow shaded area represents the range in sulphur emissions resulting from a model of degassing of erupting andesite only. E1, 2, 3: eruptive periods; P1, 2, 3: pauses in the eruption.

of porphyritic andesite magma during decompression, and b) degassing of unerupted, crystal-free mafic magma supplied to the system, whereby mafic magma quenches, vesiculates and crystallises against the interface with andesitic magma, causing complete degassing. Water-rich mafic magmas in arc settings may contain up to around 3000 ppm S [*Wallace*, 2005]. Melt inclusions from the plagioclase phenocrysts in the erupted andesite typically contain less than 250 ppm S [*Edmonds et al.*, 2001]. Ground deformation throughout the eruption, measured by continuous GPS, has been modelled in terms of temporal changes in fluxes of magma between different reservoirs in the system [*Elsworth et al.*, 2008]. In particular, a deep and a shallow reservoir can be resolved, with changes in volume of the deeper reservoir dominating the deformation signal. We use the model-derived mean surface magma flux throughout the three eruptive periods and pauses (which ranges from 0.1 to 6.5 m^3s^{-1}) to calculate the yield of sulphur supplied by degassing of andesite during eruption and the model-derived basement fluxes (which ranges from 0.2 to 4.1 m^3s^{-1}) to represent the volume flux of mafic magma entering the volcanic system from the lower crust [*Elsworth et al.*, 2008]. The intrusion of mafic magma into more evolved magma at depth is widely considered to be the driver of the eruption by transferring heat and volatiles to the overlying magma, thus causing it to erupt [*Murphy et al.*, 2000; *Devine et al.*, 2003]. Figure 3 shows the modelled cumulative sulphur emissions compared to the actual sulphur emissions and illustrates clearly that the observed sulphur emissions cannot be derived from degassing of andesite and are compatible with the complete degassing of mafic magma containing 2000–3000 ppm sulphur at the mafic magma-andesite interface at depth.

[8] This mechanism for supplying volatiles to the Soufrière Hills volcanic system is corroborated by other degassing studies. The composition of the gas phase at the surface in terms of SO_2, H_2S and CO_2 is sensitive to pressure, oxygen fugacity and the H_2O content of the magma [*Moretti and Papale*, 2004]. The gases measured during July 2008 were H_2S-rich and CO_2-rich and not in equilibrium with andesite at low pressures [*Edmonds et al.*, 2010]. The gas composition is consistent with a mixture of gases derived from deep degassing and more shallow, andesite-derived gases, with a CO_2- and H_2S-rich deep vapour in equilibrium with mafic magma at depth mixing with H_2O-rich and SO_2-rich vapour at low pressures. The exsolved gases require a mafic end member with a pre-eruptive CO_2 content of 2000–5000 ppm and up to 8 wt% H_2O and a rhyolitic end-member containing 2000 ppm and 4–6 wt% H_2O and indicate an equilibration pressure >400 MPa, or depths greater than around 13–15 km [*Edmonds et al.*, 2010], i.e., slightly deeper than the lower magma reservoir derived from inverting deformation data [*Elsworth et al.*, 2008]. An independent estimation of pre-eruptive melt CO_2 content can made using the measured mass CO_2/SO_2 ratio of 2.5–3.6 (molar ratio of 3.5–5.1 [*Edmonds et al.*, 2010]), a total cumulative sulphur emission of 3.5 Mt (Figure 3), a total erupted volume of 1 km^3 [*Wadge et al.*, 2010] and a phenocryst content of 50% by volume. We obtain a maximum pre-eruptive CO_2 content of 6000–9000 ppm for the andesite, or a lower CO_2 content if the CO_2 gases are sourced from a mixture of the intruding mafic magma and the andesite; this estimate falls within the CO_2 ranges estimated by thermodynamic modelling [*Edmonds et al.*, 2010].

[9] Chlorine degassing is effectively modelled by assuming that chlorine partitions from the rhyolitic melt to the H_2O-rich vapour during decompression of the andesite [*Edmonds et al.*, 2001]. A pre-eruptive Cl content of >5400 ppm is required to produce the highest HCl/SO_2 ratios measured in the gas phase using mass balance (assuming a phenocryst content of 50% by volume and that most of the Cl is degassed in the form of HCl), which is consistent with the higher end of the range of Cl contained in melt inclusions [*Humphreys et al.*, 2010]. Chlorine may also be contributed to the rhyolite melt by mingling with a mafic magma end-member, which also contains significant amounts of chlorine [*Humphreys et al.*, 2010].

[10] These degassing studies have important implications for the interpretation of other petrological and geophysical data at this volcano and for assessment of hazard and risk. The intrusion of mafic magma is important for driving the eruption, both in terms of supplying heat and for supplying volatiles to the overlying andesite. There is petrological evidence that of mafic magma is physically incorporated to some extent into the andesite at a variety of scales, through macroscopic mingling, whereby mafic enclaves are formed at the interface by quenching, crystallisation and vesiculation, forcing the crystals apart and in doing so forming discrete, rounded enclaves (Barclay et al., manuscript in preparation, 2009); and through enclave disaggregation, involving mingling of melt and transfer of crystals [*Humphreys et al.*, 2009, 2010]. The mafic magma exsolves volatiles and generates a deep vapour, which migrates to shallow levels independently of melt and also contributes volatiles to the andesite via the magma mingling process, which enriches the bulk volatile concentration of the andesite. The volcanic gas flux and composition at the surface record, to some extent, the shifting balance of these processes through time. The transfer of volatiles affects magma buoyancy and also advects heat to shallow levels in the system, which is important for sustaining the eruption. The SO_2 emissions can be used as a first-order indicator of the efficiency and rate of mafic injection at depth and can be expected to decline rapidly (within a few weeks) should this magma supply at depth cease. CO_2 emissions, if a high temporal resolution measurement system be established at this volcano, are also expected to show a deep magma supply signature, with fluxes remaining high even during eruptive pauses until the deep supply begins to wane. The HCl/SO_2 ratio in the gas plume remains an effective method for establishing the timing of the onset of andesite eruption and a potential proxy for eruption rate.

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