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SO_2 emissions from Soufrière Hills Volcano and their relationship to conduit permeability, hydrothermal interaction and degassing regime

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Abstract

The time series of sulphur dioxide (SO_2) emissions during the continuing eruption of Soufrière Hills Volcano, Montserrat, yields insights into conduit permeability and driving pressures, the role of the hydrothermal system and changes in magma flux both at depth and to the surface. On a time scale of years, an effectively constant supply of sulphur from a more mafic magma at depth permits evaluation of changes in the permeability of the plumbing system between 1995 and 2002 (due to magma rheology changes and hydrothermal sealing), most of which take place in the upper few hundreds of metres (dome and upper conduit). A broadly increasing SO_2 emission rate from 1995 to 1997 can be attributed to a constant or increasing supply of exsolving sulphur from depth, combined with a broadly increasing magma discharge rate at the surface. Decreases in SO_2 flux over three orders of magnitude, from July 1998 to November 1999, were due to a corresponding decrease in permeability of the upper conduit and dome due to cooling and 'sealing' by the precipitation of hydrothermal minerals and the closure of fracture and bubble networks. The second phase of dome growth, from November 1999 to the present, April 2002, has been associated with a similar range of SO_2 fluxes to the first phase. Large dome collapses in 1997 and during a period of zero magma flux in 1998 were associated with instantaneous SO_2 emissions of > 10 kt, which indicate a capacity for significant SO_2 storage in the conduit and dome prior to the collapses. SO_2 data suggest that the second phase of dome building, despite a similar sulphur budget in terms of supply from depth and mean SO_2 emission rate at the surface (around 500 t/d), is characterised by a higher bulk permeability at shallow depths and is a more 'open' system with respect to fluid through-flow than the first phase of dome building from 1995 to 1998. The lack of large SO_2 emissions after large dome collapses, in 2000 and 2001, suggests limited storage of SO_2 in the conduit system. The data suggest that the likelihood of a switch to explosive activity after a large collapse is more unlikely now than during the first phase of dome building. Over shorter time scales, permeability changes may be recognised from the SO_2 flux data prior to the onset of dome growth and during cycles of small explosions in 1999. On time scales of minutes to hours, pulses of SO_2-rich gas emissions occur after rockfalls and pyroclastic flows, due to the release of a SO_2-rich fluid phase stored in closed fractures and pore spaces within the dome. Long period and hybrid seismic events may be associated with changes in SO_2 emission rate at the surface at various times of the eruption, although only when the temporal

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resolution of SO_2 monitoring is improved, will it be possible for these short-term changes to be correlated and evaluated effectively. Monitoring SO_2 emission rates from Soufrière Hills Volcano is, at this stage, of primary value in the long run, on the time scale of years, where the relationships between deep supply and surface emissions can be used to evaluate whether the eruption might be waning, or has merely paused, which is of considerable value for hazard assessment.

Keywords: sulphur dioxide; Soufrière Hills Volcano; degassing; permeability; COSPEC

1. Introduction

The Soufrière Hills Volcano has been active since July 1995. Volcanic unrest began in 1992, when an intense earthquake swarm occurred beneath Montserrat, followed by another intense swarm in late 1994 (Aspinall et al., 1998). Similar swarms had occurred frequently during the last two centuries, with seismic crises recorded in 1897, 1933 and 1966 (Perret, 1939; Shepherd et al., 1971; Robertson et al., 2000). These swarms are considered to represent magma rising beneath the volcano but ponding before reaching the surface (Perret, 1939; Shepherd et al., 1971). The most recent (1992–1994) seismic crisis comprised 15 episodic swarms of volcano–tectonic earthquakes located at around 10–15 km depth beneath Montserrat (Aspinall et al., 1998) and culminated in November 1994, when some events were felt locally (Robertson et al., 2000).

Extrusion of non-juvenile material began in the summit region in September 1995 and sustained extrusion of andesite lava began in November 1995 (Young et al., 1998a,b). The lava is a porphyritic, hornblende–hypersthene andesite (Murphy et al., 2000), which comprises 45–55 vol% phenocrysts (30–35% plagioclase, 6–10% amphibole, 2–4% titanomagnetite, < 0.5% quartz, < 0.5% clinopyroxene), set in a partly crystalline groundmass (Barclay et al., 1998; Devine et al., 1998a; Murphy et al., 1998). Interstitial glass is rhyolitic, and represents 5–35% of the rock by volume. It has been proposed that the abundant disequilibrium features of the phenocryst population (reversely zoned crystals, wide ranges of mineral compositions, some without crystal–melt equilibrium, resorbed quartz) are interpreted as the products of a chemically homogeneous, convecting andesite magma that was heated from below and cooled from above (Couch et al., 2001). This model explains the intermingling of crystals with contrasting thermal histories. Mafic enclaves in the andesite, from sub-cm to tens of cm in dimension, are thought to represent a mafic input, which is considered to be a possible thermal and chemical trigger for eruption (Murphy et al., 2000). The mafic enclaves have a basaltic–basaltic andesite whole-rock composition and are rounded to sub-rounded (Murphy et al., 1998, 2000). The depth to the top of the magma chamber has been estimated at 5–7 km, based on phase equilibrium studies and the water content of the pre-eruptive melt (Barclay et al., 1998) and on hypocentral data (Aspinall et al., 1998). The vertical extent of the magma reservoir is unknown.

This eruption has been characterised by sustained periods of lava dome growth with occasional dome collapses (November 1995 to July 1997, December 1997 to March 1998 and from December 1999 to present, December 2001), periodic explosive activity (August–October 1997) and intervals with little or no dome growth at all (notably from March 1998 to November 1999). This latter period was characterised by 'residual' activity, including gravitational collapses of the cooling dome and fluid-pressure driven explosions as the melt crystallised and second boiling occurred (Norton et al., 2002). The erupted volume thus far is 450×10^6 m^3.

SO_2 emissions have been monitored episodically during the period 1995–2001 by use of a correlation spectrometer (COSPEC). Elsewhere, changes in SO_2 emission rate at numerous volcanoes have been shown to relate to volcanic activity (Rose et al., 1982; Casadevall et al., 1983; Malinconico, 1987; Chartier et al., 1988; Andres et al., 1993; Kyle et al., 1994). At Soufrière Hills Volcano, COSPEC data reveal a complex relationship between SO_2 emission rate and eruptive activity (Young et al., 1998a,b; Watson et al., 2000; Edmonds et al., 2001). Increases in SO_2 emission rate accompany increasing magma extrusion rate during some periods of the eruption (Young et al., 1998a,b) and correlate with seismic activity over short time scales during cycles of inflation and deflation associated with upper conduit pressure fluctuations (Watson et al., 2000). Whilst SO_2 emissions may sometimes mirror changes in magma flux rate during dome building, we also see large SO_2 emissions during non-eruptive periods. Likewise, whilst cyclic hybrid earthquakes and banded tremor may be correlated with SO_2 emissions in 1997 and in 1999 (Young et al., in press), similar cycles in 2001 were not associated with cyclic SO_2 emissions. There are clearly several competing controls on SO_2 emission rate, linked to the magma's rheological properties, magma supply and extrusion dynamics and changes in driving pressure throughout the system. For example, undercooling, degassing and crystallisation of microlites at shallow depths in the conduit and dome give rise to changes in the rheological properties of the magma (Sparks, 1997; Cashman and Blundy, 2000) which have an influence on the permeability of the dome and modulate the emission of gases by closing fracture and bubble networks. This will cause fluid-pressure increases at shallow depths and, thereafter, pressure dissipation phenomena such as conduit earthquakes and edifice deflation (Voight et al., 1999). As another example, during periods when magma flux to the surface is limited, cooling of the upper few tens of metres of the system may be enough to enable rising magmatic and hydrothermal fluids to precipitate salts and silica minerals in fractures, cracks and pore spaces, thereby retarding the flow of these fluids to the surface (Boudon et al., 1998). All of these processes have been important to varying degrees at various stages of the eruption and all of them will affect the ability of SO_2 to reach the atmosphere.

This paper investigates features of the SO_2 flux time series from Soufrière Hills Volcano, in order to place constraints on the origin and mechanisms of sulphur degassing and to assess the role of SO_2 monitoring at this and other dome-building volcanoes. Features of the SO_2 time series are presented on different time scales and are described with respect to other magmatic processes, specifically permeability change of the conduit, wall-rock and dome system. A better understanding of the controls on SO_2 emissions will enhance the value of gas emission data as a monitoring tool, both over short (days to weeks) and longer (years) time scales. As an example of a common and persistent style of volcanic activity, results from Montserrat have important implications for the surveillance of dome-building eruptions elsewhere.

2. Measurements of SO_2 at Soufrière Hills Volcano

SO_2 was measured using a Barringer Research COSPEC at Soufrière Hills Volcano, up to December 2001. The COSPEC is a spectrometer that operates in the ultra-violet region of the blue sky (Millan, 1980; Stoiber et al., 1983). It consists of a system of optics to collect the light, a chopper disc and signal processing electronics, which may be connected to a data logger or chart recorder. The chopper disc allows two wavelength bands between 300 and 315 nm to pass through to the electronics system, which measures wavelengths both on, and off, characteristic SO_2 absorption features in this wavelength band. The ratio between these measurements is assumed to be directly proportional to column SO_2 concentration. The voltage output is calibrated with a cell containing a known SO_2 concentration that is rotated into the field of view of the instrument. Several different instruments have been used over the course of the eruption, including a mini-COSPEC during 1998 and a COSPEC V from 1999 to 2001. The authors are satisfied that the differences in precision and accuracy between these different instruments is not significant. The COSPEC has been used in horizontal traverse and in stationary mode (Stoiber et al., 1983) and both sets of measurements are discussed here. A detailed discussion of the use of the COSPEC in these configu-

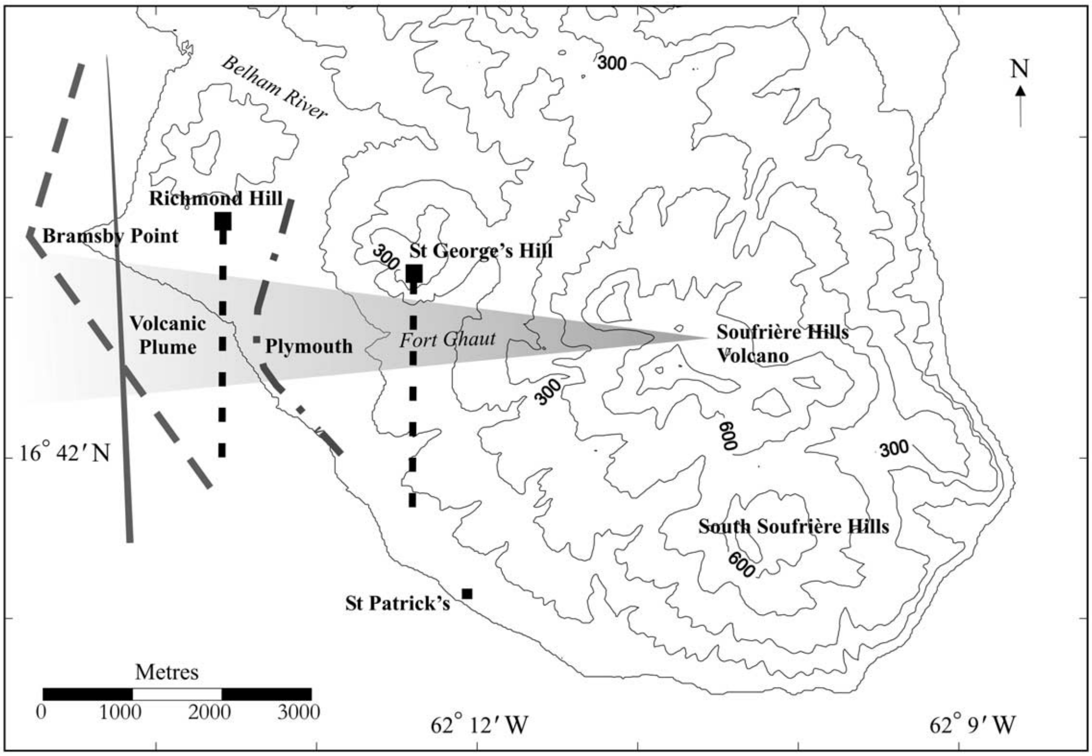

Fig. 1. Map of the south of Montserrat, West Indies. This map shows geometries of COSPEC traverses and where stationary scanning measurements were taken. The grey solid line represents horizontal traverses by helicopter, the dashed grey line a boat traverse route and the dot-dashed line a road vehicle route. Black squares show the location of stationary scanning measurements and the attached dotted lines show typical scan azimuth. Contours are shown in metres above sea level.

rations may be found elsewhere (Stoiber et al., 1983; Young et al., in press). For the horizontal traverse measurements, various platforms have been used (car, boat, helicopter). Traverse geometries and the configuration for stationary measurements are shown in Fig. 1. The plume azimuth is generally westerly to northwesterly (accounting for 87% of the days in 1 year on average), with a mean speed of 14–16 m s^{-1}.

Errors in COSPEC measurements have been discussed in detail elsewhere (Stoiber et al., 1983; Williams-Jones et al., 2001) and range from ± 13 to $\pm 42\%$, incorporating operator and methodological errors. The main source of error for SO_2 emission rate measurements using this method is widely regarded to be the uncertainty in plume speed, which accounts for up to 75% of this calculated error. This error has been reduced elsewhere, for example at Mt Erebus, Antarctica, with simultaneous video camera images being used to calculate more accurately the speed of the plume (Kyle et al., 1994). Errors associated with measurements of wind speed from ground-level anemometers alone are calculated to be +30% −5%, the error used here and elsewhere for COSPEC measurements on Montserrat (Young et al., 1998a,b). Additional errors accrue when the plume is opaque (ash or aerosol-laden) and become significant when the plume opacity exceeds 50% (Andres and Schmid, 2001).

The TOMS (Total Ozone Mapping Spectrometer) instrument is a spaceborne spectrometer also operating in the ultra-violet spectral region. It has been widely used to quantify and map SO_2 clouds (Krueger et al., 1995). The TOMS Volcanic Emissions Home Page is at http://skye.gsfc.nasa.gov/.

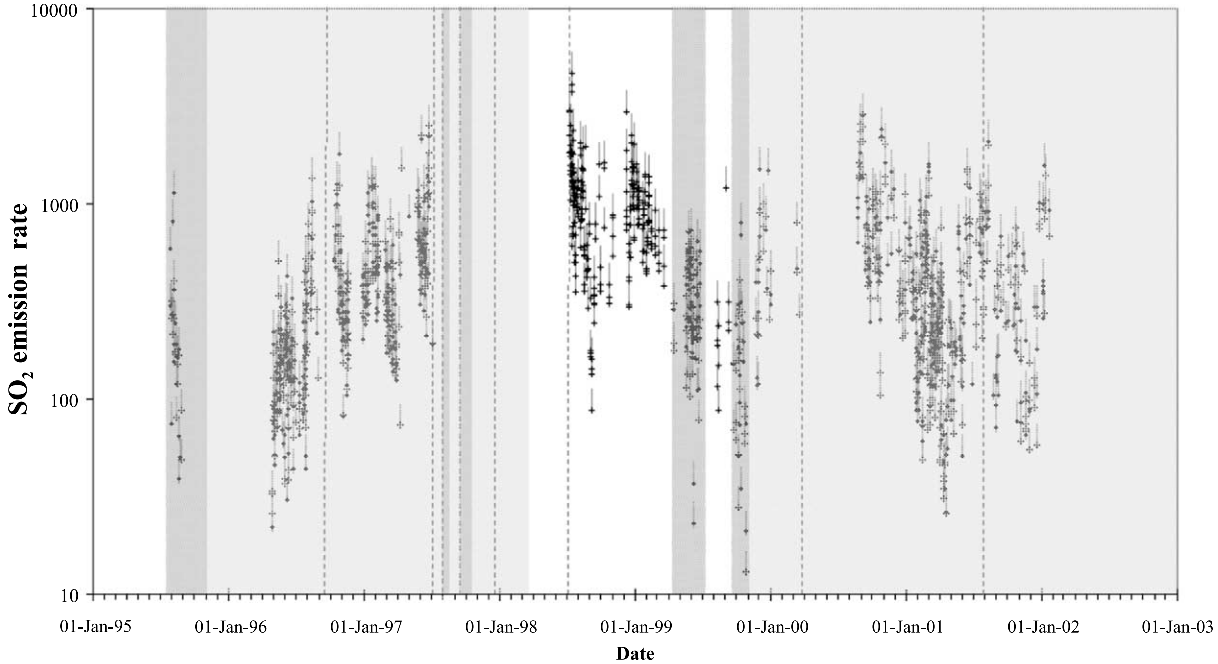

Fig. 2. SO_2 time series from Soufrière Hills Volcano, Montserrat, West Indies, from COSPEC measurements. Errors are typically +30% −5% on each data point. The major volcanic events are indicated. The pale shading shows the two main periods of lava extrusion and dome growth. Vertical dashed lines show large dome collapses. Dark shading represents periods of sustained explosive activity.

TOMS has detected SO_2 clouds 0–2 days after large volcanic events at the Soufrière Hills Volcano, with practical detection limits of slightly more than 2 kt SO_2 (Carn, personal communication).

2.1. SO_2 emission rates over the entire eruption, 1995–2001

Fig. 2 shows the COSPEC SO_2 time series for Soufrière Hills Volcano, Montserrat, from July 1995 to November 2001. Emission rates of SO_2 have been highly variable over both long time scales (months) and short time scales (days to weeks), with mean daily fluxes ranging from < 100 t day^{-1} (August 1995, November 1996, September 1999) up to > 2000 t day^{-1} (June 1997, July 1998, December 1998, August and November 2000). For a set of *x–y* COSPEC traverses made in a single day, the standard deviation as a percentage of the mean flux ranges from 8% up to 81%, with an average of 27%. The upper range of this variability is far in excess of measurement error and is considered to be real (due to either variations in source flux; or meteorological or topographical factors controlling near-source plume dispersion).

A broad trend of increasing SO_2 emission rate characterised the time series from July 1995 up to July 1997. The peak in SO_2 emission rate (4654 t day^{-1}) measured by COSPEC occurred during a period of no dome growth, following a large dome collapse on 3 July 1998. From July 1998 and throughout 1999 SO_2 emissions decreased, to a minimum of 50–150 t day^{-1} during September 1999. SO_2 fluxes increased again at the onset of the second phase of dome growth in November 1999. During this second phase of dome growth SO_2 emissions peaked in September 2000 (> 2000 t day^{-1}) and subsequently declined to between 100 and 700 t day^{-1} in September and October 2001.

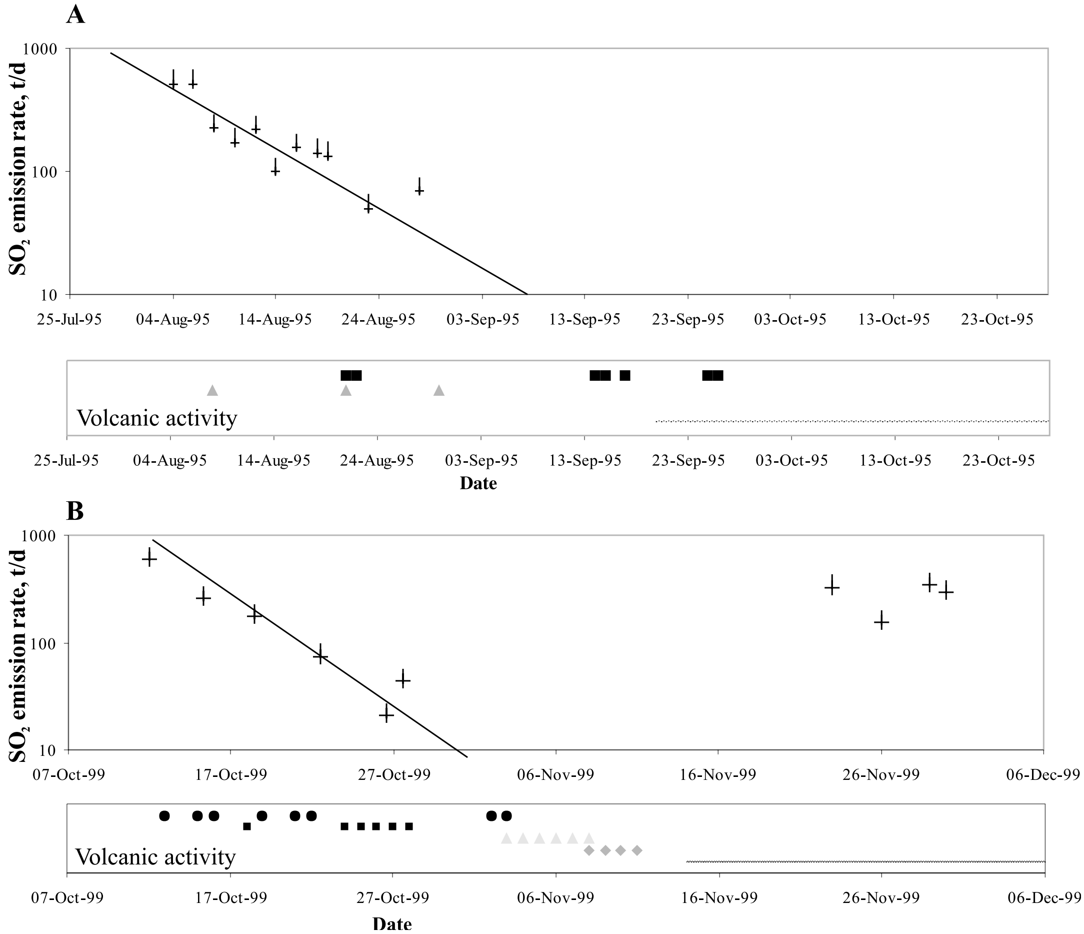

Fig. 3. SO_2 emission rate against time prior to the onset of lava extrusion in (A) 1995 and (B) 1999. Each plot shows a log plot of SO_2 over time. Beneath each is a time line of volcanic events prior to, and synchronous with, the onset of lava extrusion. Black circles represent periods of vigorous ash venting, black squares represent the occurrence of hybrid earthquakes, grey triangles are phreatic explosions, grey diamonds are periods of magmatic explosions and the dotted grey line shows the timing of the onset of lava extrusion.

Table 1
Table to summarise features of large dome/sector collapses at Soufrière Hills Volcano, Montserrat, 1997–2001

Date	Event description	Volume	TOMS
26 December 1997	Sector collapse and blast	90×10^6 m^3, talus, wallrock and dome material (Sparks et al., 2002)	33 kt (Mayberry et al., 2002)
3 July 1998	Dome collapse with basement failure	30×10^6 m^3, dome rock and basement	11 kt (Mayberry et al., 2002)
29 July 2001	Dome collapse	45×10^6 m^3 dome rock	2 kt (30 July 2001; Carn, personal communication)

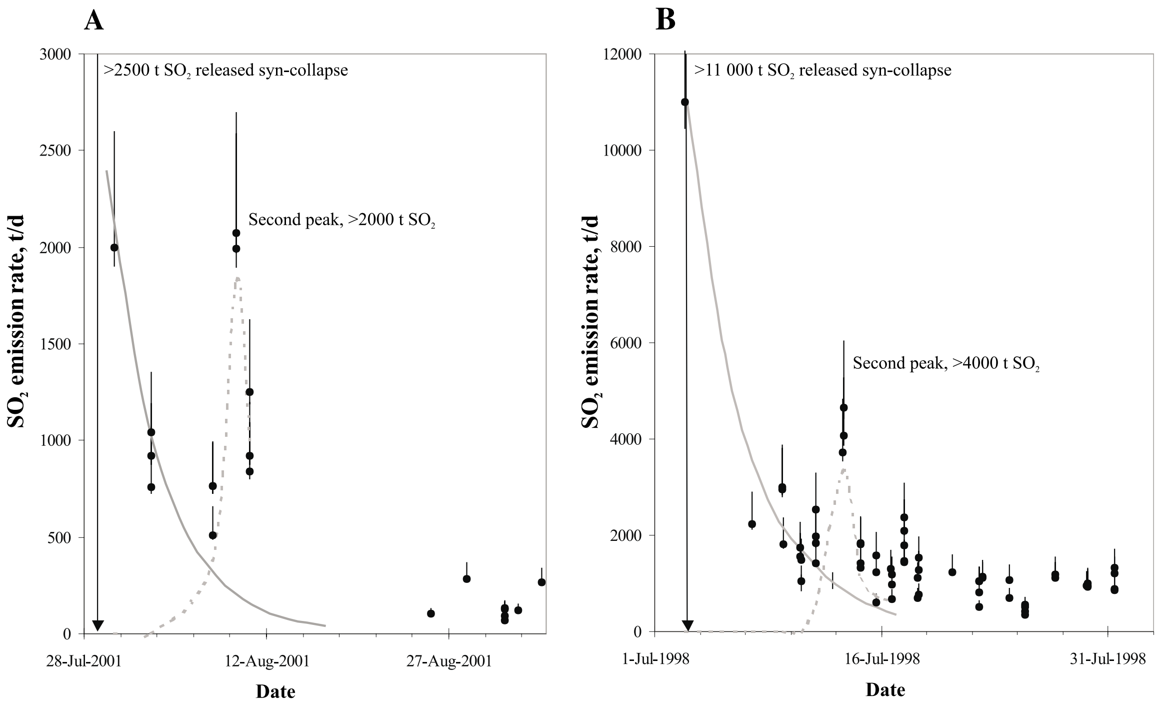

Fig. 4. SO_2 emission rates after dome collapses on (A) 29 July 2001 and (B) 3 July 1998. The dome collapses are indicated by vertical arrows on the time series. The solid line is an exponential decay fitted to the instantaneous SO_2 emission, the dotted line is generated by subtracting the first fit from the actual measurements. The SO_2 emission rates measured therefore represent the superposition of two SO_2 emission profiles. The instantaneous SO_2 emissions for 3 July 1998 and 30 July 2001 are derived from the TOMS data (Bluth and Carn, personal communication).

2.2. *Weeks–months changes in SO_2 emission rates*

2.2.1. *Decrease in SO_2 emission rate prior to the start of magma ascent, after a pause in dome growth*

The onset of both major phases of dome growth at Soufrière Hills Volcano were preceded by a decrease in SO_2 emissions over the weeks prior to the first extrusion of lava (Fig. 3). In both cases, phreatic explosions and hybrid earthquake swarms also accompanied the onsets of lava extrusion (Fig. 3).

2.2.2. *SO_2 emissions after large dome collapse events*

SO_2 emission rates are typically high after large dome or sector collapse events (Table 1). COSPEC measurements after two large events (3 July 1998 and 29 July 2001), reveal two peaks in SO_2 emission: one immediately after the event and a second 10 or 11 days later (Fig. 4). These secondary peaks both occurred during seismically quiet, post-collapse phases. Unfortunately, COSPEC data are unavailable for the other large collapse events between 1996 and 2001.

2.3. *SO_2 emission rates over hours–days time scales*

2.3.1. *Steam explosions during May and June 1999*

Short-term variations in SO_2 emission occurred during a period of phreatic explosive activity in 1999. From November 1998 to January 1999 (during a period of no lava extrusion to the surface), a series of collapses reduced the volume of the dome, forming an east–west gully and exposing a vent on the crater floor. Shortly after, from April to June 1999, small explosions, or venting episodes involving fine ash and steam emissions, occurred at frequent intervals (typically every 2–6

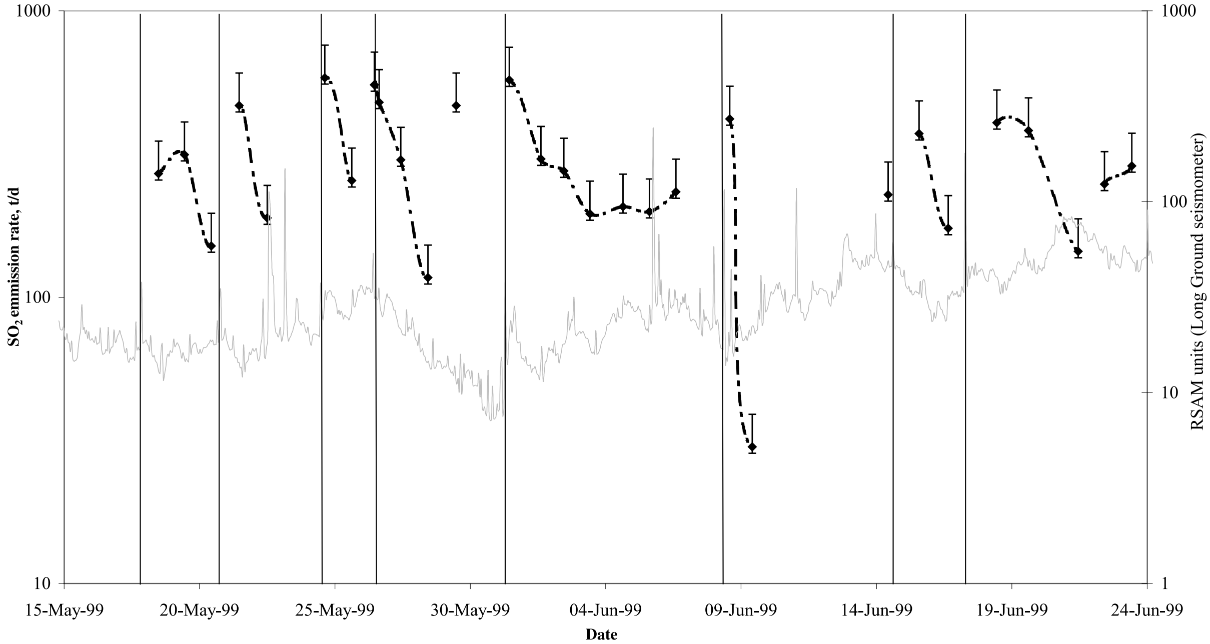

Fig. 5. SO_2 emission rates in May and June 1999 (black symbols and dashed lines). Errors on these measurements are +30% −5%. The RSAM data from the Long Ground seismometer on Montserrat are shown as a grey solid line. The occurrence of explosions and vigorous ash venting episodes are marked as black vertical lines.

days). The COSPEC data for the period 17 May to 26 June reflect this venting activity. Gas fluxes were high immediately after each explosive event and decreased over the following days down to emission rates of < 200 t day^{-1} (Fig. 5), soon after which the next venting or explosive episode took place.

Gravitational collapse events during this period, involving the excavation of small scars in the outer northern flank of the dome, appeared to have little effect on fluxes.

2.4. SO_2 emission rates over minutes–hours time scales

2.4.1. Cycles of enhanced seismicity and banded tremor

Cyclic seismicity, with a period of 6–20 h, is a phenomenon seen frequently at the Soufrière Hills Volcano. Periods of heightened seismicity are characterised by hybrid and long-period earthquakes, rockfalls and, occasionally, harmonic tremor (Baptie et al., 2002; Luckett et al., 2002; Neuberg and O'Gorman, 2002).

From April to June 1997, a cyclic pattern of SO_2 emissions correlated with cycles of hybrid earthquake, tilt and pyroclastic flow activity (Watson et al., 2000). During December 1999 and January 2000, when the cycles were dominated by harmonic tremor, SO_2 emissions appeared to correlate with the RSAM (relative seismic amplitude measurement; Endo and Murray, 1991), detected on the short-period and broadband seismometer network around the volcano. Peaks in SO_2 emission corresponded with RSAM peaks (Young et al., in press).

In August, September and October 2001, when rockfalls and long-period events dominated the seismicity, there was no clear relationship between SO_2 emission rate and RSAM level, and SO_2 emissions ranged from 50 to 1000 t day^{-1} during both peaks and troughs in seismic intensity (Fig. 6). Higher SO_2 emissions (600–700 t day^{-1}) occurred during and after rockfalls and pyroclastic flows, whilst low-frequency long-period and hybrid events seemed to have little effect on mass fluxes of SO_2. This increase in SO_2 with rockfalls has been seen in stationary scanning data

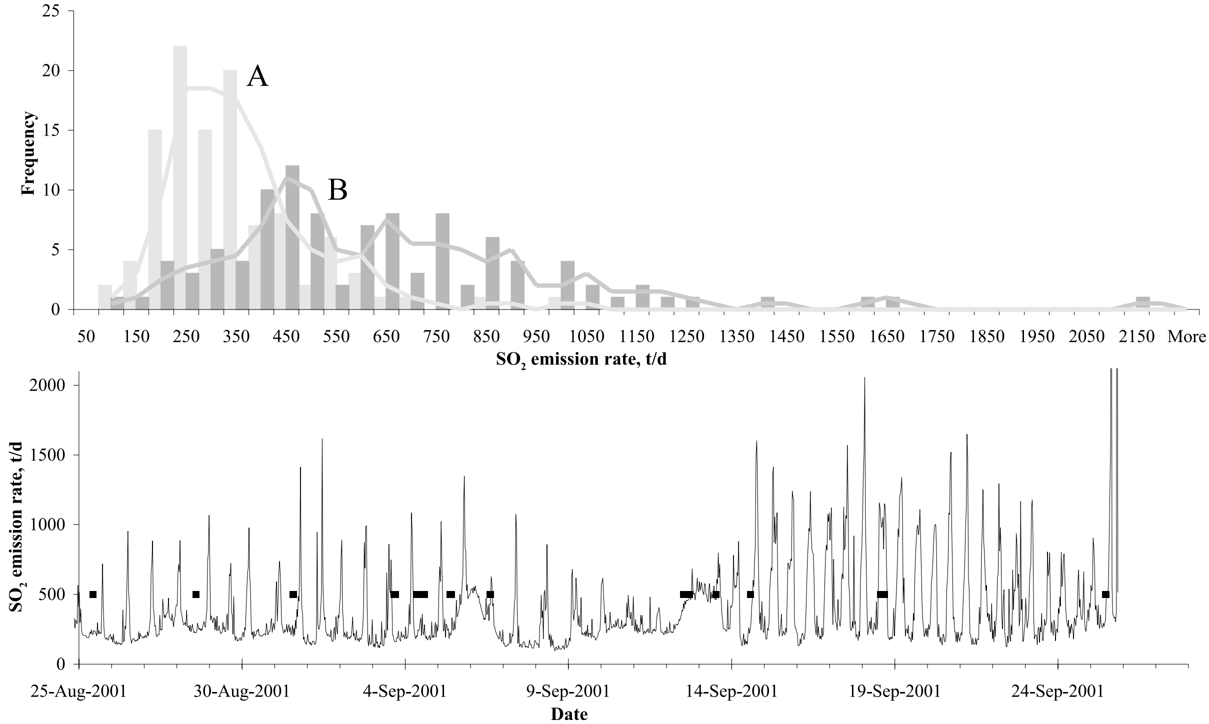

Fig. 6. Top: histogram to show the distribution of SO_2 emission rates throughout August and September 2001, through a period when the seismicity was dominated by 6–20 h cycles of rockfalls and long period events. (A) Measurements of SO_2 taken on 25, 28, 31 August, 3, 4, 5, 14, 18, 25 September 2001. (B) Measurements taken on 12 and 13 September. The SO_2 emission rates measured on 12 and 13 September were elevated and coincided with long tremor periods (over 24 h). Bottom: the Long Ground RSAM data for this period, showing the cycles in the seismicity and when COSPEC measurements were taken.

throughout the eruption. On 12 and 13 September 2001, a prolonged 36-h period of high RSAM occurred, mainly composed of rockfalls and long-period events. Over two 4-h measurement periods, fluxes had a larger range (100–2200 t day^{-1}) and modal value (450 t day^{-1}), than those measured on 25, 28, 31 August and 3, 4, 5, 6, 14, 18, 25 September 2001, which were characterised by a range of 50–1000 t day^{-1} and a modal value of 250 t day^{-1} (Fig. 6).

3. Discussion

3.1. The origin of the degassed sulphur at Soufrière Hills Volcano

Evidence from plagioclase-hosted melt inclusions in the erupted andesite indicates that dissolved sulphur concentrations were below 150 ppm in the pre-eruptive andesite and degassing of the andesite can only account for a small part of the observed SO_2 emission rate at the surface (Barclay et al., 1998; Edmonds et al., 2001; Table 2). This phenomenon has been documented at many intermediate-silicic volcanoes and is commonly referred to as the 'excess sulphur' problem (Stoiber and Jepson, 1973; Rose et al., 1982; Luhr et al., 1984; Andres et al., 1991; Wallace and Gerlach, 1994; see Wallace, 2001, for a recent review).

The source of this 'excess' sulphur has been a common subject for debate and has been identified as various processes, including: the degassing of unerupted magma (Rose et al., 1982); the breakdown of sulphur-bearing phases such as anhydrite and pyrrhotite (Devine et al., 1984); mixing between reduced and more oxidised magmas

(Kress, 1997); and the presence of a sulphur-bearing fluid phase at depth prior to eruption (Wallace and Gerlach, 1994). The mafic input to the andesite system and the formation of a fluid phase at depth are proposed to be the primary source of this sulphur at Soufrière Hills Volcano during this eruption. Basaltic–basaltic andesite melts are capable of dissolving a greater mass of sulphur than more silicic melts (Carroll and Rutherford, 1985) and this is therefore a plausible candidate for the sulphur source, although much experimental work remains to be done on the capacity of basaltic–andesite compositions to hold sulphur and other volatiles under these conditions. Sulphur fluid–melt partition coefficients have been found to be strongly dependent on redox conditions (Scaillet et al., 1998). In the mafic enclaves within the andesite, the glass contains very small (sub-μm to few μm size) droplets of a sulphur-rich immiscible liquid phase (∼ Fe–S–O; Murphy, personal communication, and authors' own work), whilst the rest of the glass contains little sulphur (< 100 ppm; Table 2). The presence of this sulphide phase in the mafic melt suggests that the melt was saturated with respect to sulphur prior to contact with the mid-crustal reservoir of andesite magma. At this stage, the mafic melt existed under high-temperature, reducing conditions and the fluid–melt partition coefficient for sulphur in this melt was low (Scaillet et al., 1998). As the magma began to cool and crystallise on contact with the cooler andesite at mid-crustal levels, an increase in the Fe content of the melt may have increased the solubility of S in the melt and therefore prevented the further formation of immiscible liquids or sulphide phases in the mafic melt. However, the relatively oxidising conditions of the andesite-bearing magma reservoir will in turn have countered this solubility increase by increasing the fluid–melt partition coefficient for S (Scaillet et al., 1998) and may have led to the efficient stripping of sulphur from the mafic magma, partitioning into a fluid phase (with CO_2) at the time of intrusion into the andesite storage area.

3.2. A model to describe SO_2 degassing

Having developed the hypothesis that the sulphur is sourced from a more mafic, non-erupted magma at depth, and partitioned into a SO_2-rich fluid phase at magma storage region depths (5–7 km), we now address the manner in which this fluid migrates upwards and reaches the surface.

SO_2 emissions at the Soufrière Hills Volcano vary over two orders of magnitude and over a range of time scales. If we assume that there is a constant supply of mafic magma from depth throughout the eruption, regardless of the rate of extrusion of andesite lava at the surface, then it follows that the system is not degassing freely all of the time. This assumption is supported, in part, by long-term differential GPS measurements, from a network of GPS receivers positioned around the volcano. Throughout the first phase of dome building, a net subsidence was recorded. From March 1998 to November 1999, when there was no lava extrusion, inflation of the volcanic edifice occurred. When lava extrusion began once more in November 1999, the subsidence trend returned (Mattioli et al., 2002). The simplest interpretation of the GPS data is that the andesite extrusion to the surface emptied the storage region at a faster rate than the mafic supply during extrusive periods, while during the residual period the mafic supply exceeded the escape of andesite from the storage region. The assumption

Table 2
Representative sulphur concentrations (columns 1–7) of melt inclusions inside plagioclase, matrix and mafic glass and the mean of those samples analysed (column 8, numbers in brackets denote number of analyses), measured by CAMECA electron microprobe (Cambridge University), in ppm (Edmonds et al., 2001)

	1	2	3	4	5	6	7	8
Plagioclase melt inclusions	62	b.d.	5	b.d.	140	b.d.	108	46 (40)
Matrix glass	b.d.	b.d.	36	149	89	b.d.	54	70 (61)
Mafic glass	62	15	68	43	98	104	b.d.	88 (48)

b.d., below detection.

used here is that the mafic supply (i.e. sulphur supply) has been effectively constant throughout the eruption thus far, notwithstanding small variations in magma flux to the surface on the short time scales of days to weeks that have been recognised through estimates of lava dome volume.

Given the assumption of an approximately constant supply of sulphur from depth, the pattern of SO_2 release at the surface can be interpreted as reflecting the ease of passage of gas out of the magma chamber and through the conduit, dome and shallow portions of the volcanic edifice. The COSPEC time series shows that the SO_2 fluxes are highly variable, on all time scales. We propose that the variability in SO_2 flux at this volcano is principally a function of permeability changes in the conduit and dome, over both short and long time scales. A simple model is developed using a modified version of the Darcy equation, relating the flux Q_S^{atm} ($m^3\ s^{-1}$) out of the system, with a cross-sectional area A (m^2), characterised by a length scale L (m) and across which there is a pressure drop ΔP (Pa). The fluid is uniform with a viscosity μ (Pa s) and flows through a network with a permeability k (m^2):

$$Q_S^{atm} = (Ak\Delta P)/(\mu L) \quad (1)$$

This assumes a constant flux of gas from below; that the gas is mainly composed of steam (SO_2 is only a small component of the fluid phase); the viscosity is of the order of 10^{-5} Pa s at the elevated pressures and temperatures within the dome; the range in pressure drop across the system is of the order of tenths to tens of MPa and the characteristic length dimension is of the order of 1500 m. This is the region in which much of the changes in magma properties occur (Sparks, 1997) and will be the dome and upper conduit. The upper conduit cross-sectional area is taken as approximately 1000 m^2 (from observations of the eruptive vent and characteristic lava spine dimensions). With a driving pressure of 0.1–10 MPa and a gas flux of several thousand tonnes per day (water emission rates at the *surface* have been estimated at 22 000–1 000 000 t day^{-1} based on gas sampling (Hammouya et al., 1998) and numerical modelling (Edmonds et al., 2002)), the effective bulk permeability of the dome and upper conduit region will be of the order of 10^{-9}–10^{-12} m^2 (Fig. 7). This estimate is larger than others; e.g. Voight and Elsworth (2000) quote a range of 10^{-13}–10^{-18}, but it incorporates large-scale fractures, interconnected bubble networks and wall-rock paths of gas escape to the atmosphere.

This estimate is relatively insensitive to uncertainty in L and A. Notice also that if the pressure drops or the permeability across the system changes, there will be a corresponding change in gas flux out of the system. For example, if an increment of magma exsolves volatiles (mainly water) and gas pressure increases, gas escapes (if it is able to), pressure drops and the flux of gas out of the dome system will again decrease. The decrease in pressure at the base of the system may cause another increment of magma to degas. This oscillation in pressure drop across the system may induce a time-dependent behaviour involving feedback between tremor, permeability and gas supply. A plug of relatively degassed (with respect to water), viscous magma may cause a reduction in the permeability of the upper conduit and dome. If there is a constant or increasing supply of exsolved gas building up beneath the blockage, then a situation may arise in which this sealing will cause an increase in gas pressure, resulting in a change in gas emission rate, determined by the relative magnitudes of gas pressure increase and tensile strength of the conduit and edifice.

The SO_2 emission rate may provide a proxy for detecting variations in total gas flux from the volcano, related to sealing and levels of overpressure. This process can act over minutes-to-years time scales, depending on the process causing the permeability change. For instance, rheological properties can change over hours as magma ascends the conduit, whereas the cooling and hydrothermal precipitation of minerals along fractures may take months to years to reduce permeability.

It is expected that the ability of other deep-derived volcanic gases, such as CO_2 and H_2O, to degas through the conduit and dome system may be similarly affected by the permeability constraints discussed here. HCl, however, is thought to degas at much shallower depths than SO_2 and CO_2 in this system (Edmonds et al., 2002) and

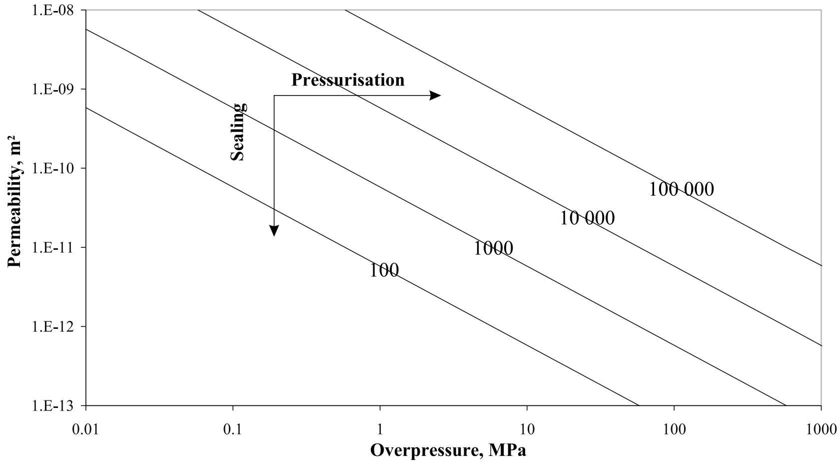

Fig. 7. A plot of overpressure (Mpa) and permeability (m^2) for the dome and upper conduit region derived from Darcy's Law. Fluxes of gas (steam) out of the system are contoured and labelled (tonnes per day).

once magma extrusion to the surface has ceased, the emission rate of HCl is expected to decay rapidly owing to its high solubility and 'scrubbing' by the hydrothermal system.

3.3. Sealing processes and permeability change

3.3.1. Hydrothermal sealing

The cessation of dome growth in March 1998 was followed by a period of cooling of the emplaced magma in the upper conduit and dome. After the 3 July 1998 dome collapse, fumarolic activity was high and vigorous steaming was observed within the collapse scar caused by this event for months afterwards. The addition of hydrothermal fluids to heated, silicic rocks has been well documented and has the effect of bringing about large changes in permeability due to the precipitation of silica in fractures and pore spaces once cooling begins (Tenthorey et al., 1998; Olsen and Scholz, 1998; Renard et al., 2000). This process has also been studied experimentally. Moore et al. (1994) investigated time scales of sealing and permeability change by passing fluid through heated granite at temperatures of 300–500°C with a pore pressure drop of 2 MPa in order to investigate time scales of sealing and permeability change. The permeability of the granite at the start of the experiments was calculated as 5×10^{-19} to 1×10^{-20} m^2 (the room temperature permeability at 50 MPa of the granite). It was found that permeability of the granite samples decayed over time after the heating episode according to a relationship of the form:

$$k = c(10^{-rt}) \quad (2)$$

where k is permeability, c is a constant and t is time. It was found that the parameter r, which describes the time dependence of the permeability decrease, varied with the temperature attained at the end of the initial heating episode. Samples heated to 500°C experienced a much faster decay in permeability, k, than those heated to 300°C. The relationship between temperature and the parameter r is shown in Fig. 8.

This figure shows that for experiments conducted at elevated temperatures, the permeability changes occur much faster and r values are higher, than for lower-temperature experiments. The implications of this work are that, if the experimental time scales (up to 60 days) and the change in r with initial temperature can be extrapolated, the permeability of granitic rock to fluid flow can be reduced by three orders of magnitude on time scales of 2–6 days (when cooling from 700°C) to 12–20 months (when cooling from 400°C). The higher-temperature example may be applicable to the 2–6 day repetitive explosive activity in April to June 1999, whilst the lower-temperature example may explain the decrease in SO_2 emissions during the period July 1998 to November 1999.

Thus it is to be expected that 'sealing' of dome rocks may be attained rapidly, and that sealing rates might be influenced by the prevailing eruptive conditions. This experimental work was intended to provide insights into sealing mechanisms in active fault zones, and hence prove that sealing mechanisms may be rapid compared to earthquake recurrence times. In the same way, we can use these results to illustrate that sealing may occur in cooling, shallow, volcanic systems on similar time scales, and that a well-connected crack network, necessary to transmit a fluid phase to the atmosphere from depth, may be impossible to maintain for a length of time comparable to the residual period from March 1998 to November 1999, and this could explain the decrease in SO_2 flux during this time.

The sealing mechanism is likely to involve the precipitation of secondary hydrothermal minerals from solution in the fluid phase. Silica minerals are the only significant stable alteration minerals to occur in acidic systems, comprising opaline silica, tridymite, cristobalite and quartz, which deposit within surficial hydrothermal environments inside vesicles, cavities and fractures (Boudon et al., 1998; Corbett and Leach, 1998). Hydrothermal minerals in pre-eruption fumarole rocks and early 1995 phreatic ashes have a large sulphate

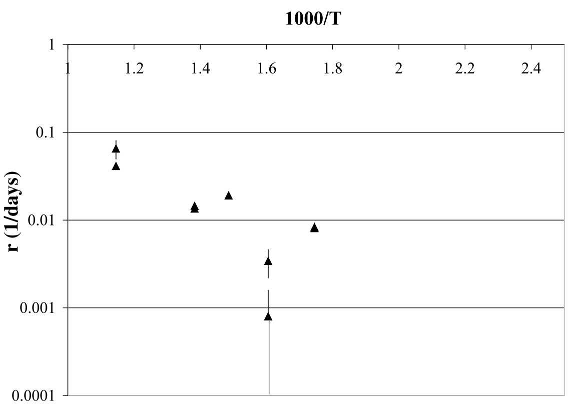

Fig. 8. A plot of *T*/1000, where *T* is the temperature in °C, against the parameter *r* (for definition, see text and Moore et al., 1994).

(and also polymorphs of silica) component, indicating an extensive hydrothermal system (Boudon et al., 1998). It has been suggested that the deposition of silica by hydrothermal or magmatic fluids may considerably reduce the porosity and the permeability of the country rocks of the magmatic conduit, preventing further development of the hydrothermal system around these areas and isolating the magmatic from the hydrothermal systems (Boudon et al., 1998). This has also been documented at Mount St Helens (Komorowski et al., 1997) and Galeras, Colombia (Stix et al., 1997). This is likely to be the sealing mechanism that, at times, prevents the escape of fluids from the magmatic conduit.

In epithermal environments (< 150°C), such as a cooling outer-dome carapace, fumaroles, a cooling open vent, cooling pyroclastic flow deposits and wallrocks, amorphous silica, cristobalite and tridymite are deposited. At Soufrière Hills Volcano, lava clasts in pyroclastic flow deposits contain 3–6% silica on average, mainly cristobalite (Horwell et al., 2001). For epithermal/mesothermal environments (< 300–350°) in the dome or upper conduit, quartz will deposit on cooling, with a maximum solubility at 350°C (Corbett and Leach, 1998). Therefore, rapid quenching of high-temperature fluids in the upper regions of a fluid up-flow conduit wall will produce a silica cap to seal the system. This could happen on a time scale of approximately months to years (Moore et al., 1994). For still higher temperatures (300 to > 400°C), fluid pressure exerts a large control on quartz solubility, as well as temperature. Rapid pressure drops can result in silica oversaturation and give rise to quartz vein development. This process can occur on shorter time scales and could give rise to cyclic sealing and pressure dissipations such as the activity in May and June 1999.

3.3.2. *Permeability decrease during magma ascent*

During periods of active extrusion, the high temperatures in the conduit and dome are likely to have allowed the shallow-level system to maintain a high permeability in the shallow-level system due to thermal cracking (Moore et al., 1994) and the development of a three-dimensional bubble network as volatiles, principally water, exsolve and expand on ascent. Permeability changes during periods of dome building most likely reflect rheological changes in the magma as it ascends, degasses and crystallises, in the top few hundreds of metres of the system (Sparks, 1997; Cashman and Blundy, 2000). It has been proposed that magma extrusion from the conduit is controlled by two competing processes (Voight et al., 1998): exsolution and degassing of water at the top of the magma column increases magma viscosity and yield strength in this region, leading to the development of a stiff plug of degassed magma and retarding magma flow. Pressurisation of the more water-rich and less viscous magma beneath the degassed plug acts to push the magma plug out of the conduit. The cycles of deformation, seismicity and SO_2 emission seen in May to August 1997 (Voight et al., 1998; Watson et al., 2000) are consistent with this model. Inflation of the edifice occurs as the magma beneath the plug becomes pressurised, accompanied by hybrid earthquakes or tremor. When the tensile strength of the degassed plug is exceeded, the plug is forced from the conduit along with a pulse of more volatile-rich magma. The pressure build-up is dissipated and deflation of the upper edifice occurs. This pressure drop introduces a feedback mechanism by promoting exsolution of volatiles from the next increment of magma, thereby increasing its viscosity and causing the cycle to begin once more. A model of stick and slip between the magma and conduit walls has also been proposed to describe cyclicity of this type at silicic volcanoes (Denlinger and Hoblitt, 1999). A steady supply of viscous magma from a reservoir into a conduit with non-smooth walls may experience fluctuating flow rates, resulting in a 'pulse' of rising magma. Pressure oscillations at the base of the magma pulse will also promote oscillatory volatile exsolution from the magma.

Since most SO_2 is exsolved at depths greater than those at which this cyclicity originates, its emission at the surface is dependent on the physical properties of the medium through which it travels to reach the surface. Therefore, the time-dependent variation in permeability of the upper reaches of the conduit, and of the pressure drop across the conduit and dome, are expected to lead to time-dependent variations in SO_2 emissions. The measurement of emission rate of SO_2 to the atmosphere during this style of activity therefore provides a means to examine the changes in permeability that arise during rheological changes induced in andesite magma at shallow levels. These pulses in magma flow rate are evident on a variety of time scales at Soufrière Hills Volcano. Short cycles over time scales of hours to days are reflected in cycles of increasing and decreasing seismicity and episodes of banded tremor. On a longer time scale, cycles of 6 and 14 weeks have been recognised at different times throughout the eruption. These cycles are reflected in an increasing and decreasing SO_2 emission rate.

4. An interpretation of the Soufrière Hills Volcano SO_2 time series

The broadly increasing SO_2 emission rate up to July 1998 (Fig. 2) indicates that this is not merely the progressive degassing of a single batch of mafic magma emplaced in a shallow crustal storage area, as appeared to be the case at other volcanoes, such as Mt Unzen (Japan) in 1992–1993. At Unzen, SO_2 emissions were not detected prior to the onset of extrusion. SO_2 emission rate correlated closely with extrusion rate of andesite lava (Hirabayashi et al., 1995) and two distinct magma pulses were recognised, which corresponded to clear peaks in SO_2 emission rate, followed by subsequent decay as extrusion rate decreased. In contrast, the SO_2 time series at Soufrière Hills Volcano shows a steadily increasing SO_2 emission rate throughout the first phase of dome building, with the peak in SO_2 emission rate occurring in July 1998, after extrusion had ceased. This indicates a constant or increasing supply of the mafic magma that supplied the sulphur to the system from depth, accompanied by a progressive 'opening' of the system by fracture propagation and 3-D bubble network formation. A broad trend of decreasing SO_2 throughout the residual period implies a permeability decrease in the edifice and conduit region due to the precipitation of hydrothermal minerals along fractures in the conduit and dome and the closure of 3-D bubble networks, progressively sealing the system, thereby retarding gas loss to the atmosphere.

The second phase of dome growth has been associated with similar SO_2 emission rate mean ranges and peaks to the first phase, with a general progressive increase up to September 2000 and several smaller peaks since then. This pattern suggests a constant or increasing supply of magma from depth, whilst the duration of this pattern suggests that the volcano may be evolving into a persistently active phase of activity, whereby the supply from depth and the ascent of magma to the surface has reached a steady state and shows no sign of ceasing in the short term. This is similar to the eruption of Santiaguito, Guatemala, a dacitic dome-building volcano, which has been semi-continuously active since 1922 (Rose, 1973) and to the activity at Bezymianny, Kamchatka, which began to erupt in 1955 and is characterised by andesite dome building, interspersed with periods of explosive activity (Simkin and Siebert, 1994).

Fig. 3 shows that the magnitude of the decreases in SO_2 emission rates prior to the onset of extrusion in both 1995 and 1999 is similar, diminishing in each case from 600–700 t day^{-1} to <100 t day^{-1} over 2–3 weeks. This suggests a similar mechanism of permeability reduction for both periods. When the SO_2 emission rates reached levels of <50 t day^{-1} hybrid earthquakes and phreatic explosions occurred, heralding the appearance of andesite lava inside the crater. This is consistent with the presence of an impermeable plug capping the ascending magma, which was breached as the magma reached the surface. The hybrid earthquakes and phreatic explosions were a manifestation of pressure dissipation, the pressure increase having been caused by water degassing without escape on magma ascent.

The 3 July 1998 dome collapse event almost instantaneously released at least 11 kt of SO_2 into the atmosphere, 3.5 months after extrusion of andesite lava had ceased (Table 1). The 26 December 1997 sector collapse event also released a large amount of SO_2, approximately 33 kt, detected 10 or 11 h after the event. These large SO_2 emissions and the similar gas-to-collapse-volume ratio ($\sim$0.4 kt SO_2 per 10^6 m^3 andesite), suggests that the conduit and dome systems were 'saturated' with respect to SO_2 trapped in fracture and bubble networks. These data suggest that the controls on SO_2 flux to the atmosphere, regardless of the magma supply and sulphur flux from depth and overpressures within the conduit and dome, are almost entirely within the dome, wallrock and upper few tens of metres of the conduit. The collapse of this material may therefore have been, in part, fluid-pressure driven, which instantaneously liberated this trapped SO_2-rich fluid. It is noteworthy that the only two events to generate large SO_2 clouds (3 July 1998 and 26 December 1997, with emissions of >10 kt) were events in which not only dome material, but basement and wallrock were involved, which might have implications for a more efficient 'opening' of the conduit system to fluid throughflow. The Vulcanian explosions of August–November 1997 were not accompanied by large SO_2 emissions (G.J.S. Bluth, personal communication). These events followed a large dome collapse on 21 September 1997, which may have effectively cleared any excess stored vapour phase.

During the second phase of dome building, from November 1999 onwards, the typical SO_2 emission for dome collapses is rather different. Major dome collapses occurred on 20 March 2000 (20×10^6 m^3 material) and 29 July 2001 (45×10^6 m^3 material). TOMS did not detect an SO_2 cloud after the dome collapse on 20 March 2000. A cloud containing around 2 kt SO_2 (close to TOMS detection limits; Carn, personal communication), was detected over Puerto Rico 20 h after the 29 July 2001 dome collapse event. The lack of large SO_2 emissions suggests that these collapses were associated with low internal gas pressures and may have been partly driven by external factors, such as intense rainfall (Matthews et al., 2002). During the second phase of dome building, although the range and average SO_2 emission rate have been similar to the first phase, the regime seems to have been characterised by higher permeabilities (of the dome, wallrocks and upper conduit) and lower overpressures. This implies a more 'open' system, where gas is able to escape relatively easily and not build up within the upper 1000 m or so of the conduit and dome system. Evidence from the numerical modelling of melt chlorine content in the first and second phases of dome building corroborates this (Edmonds et al., 2002). The 'open' system with respect to gas through-flow may, in part, explain the apparent change in nature of the pyroclastic flows during the second phase of dome building thus far. The surge component per total flow volume and the overall mobility of the pyroclastic flows have generally been lower from November 1999 to the present, whereas pyroclastic flows during 1996 and 1997, most notably 25 June and 21 September 1997, were undoubtedly associated with a large surge component (Loughlin et al., 2002).

The 3 July 1998 and 29 July 2001 dome collapses both had an associated second peak in SO_2 emissions, 10–11 days after the collapse. These are the only collapses for which COSPEC data were obtained immediately afterwards. The strong peaks in SO_2 are inexplicable in terms of magmatic activity, as there was little or no seismicity at these times. The removal of 30 and 45×10^6 m^3 andesite, repectively, during these events, may have transmitted a pressure drop to magma chamber depths and caused a spontaneous exsolution event in the mafic magma, liberating a mass of SO_2 which then migrated to the surface. A period of 10–11 days represents the time required for this slug of SO_2 to reach the atmosphere. This time scale broadly agrees with other estimates of magma ascent rate (Devine et al., 1998b; Sparks, personal communication). This suggests too, that the differential movement between the gas and magma phases is very close to zero during this phase of dome building, which also corroborates the idea that only very small changes are necessary in the system to switch from effusive to explosive activity.

The mafic magma at depth requires approximately 1030 ppm sulphur in order to supply the total emission of SO_2 throughout the eruption thus far (approximately 1.3 Mt), assuming the volume influx of chamber-intruding mafic magma is approximately equal to the total erupted volume (approximately 450×10^6 m^3). The second peak and the emission of around 10 kt SO_2 from 13 July to the end of July 1998 (Fig. 4) therefore require the spontaneous degassing of around 10 or 11 Mt, or about 4×10^6 m^3 mafic melt (or less pervasive degassing of a greater melt volume).

The cyclic activity in May–June 1999 (Fig. 5) followed the large dome collapses of November–December 1998 and January 1999, when the dome mass directly over the vent area collapsed away entirely, exposing the vent and upper conduit. As the top of the magma body in this region became sealed (through hydrothermal alteration and fracture closure), gas pressure increased until it overcame the tensile strength of the dome rocks or forced its way through closed fractures, resulting in the vigorous venting episodes or small explosions or gas jets from the dome surface. These events caused ephemeral changes in permeability, as fracture networks opened and closed, which was reflected in the SO_2 flux record. From the experimental data on the through-flow of fluids in granites, we know that if the top of the conduit magma body began to cool during this period, possibly due to being exposed through dome collapse, then order of magnitude decreases in permeability may occur on the time scale of days. This may have formed the driving force for these small explosions.

Short-term variations (on the time scale of days to weeks) in SO_2 emission rate relating to explosive activity have also been identified at Galeras Volcano, Colombia (Fischer et al., 1996; Stix et al., 1993; Zapata et al., 1997). Abrupt decreases in SO_2 flux, measured by COSPEC, were seen prior to explosions in 1992 and 1993, e.g. SO_2 emission rate dropped from 1600 t day^{-1} on June 25 to 400 t day^{-1} on July 7 prior to the July 16 1992 eruption. A decline in SO_2 flux also occurred prior to the 23 March 1993 explosion. These decreases have been attributed to the retainment of volcanic gases inside the volcanic edifice by a sealing process. Increased SO_2 flux immediately after an eruption is ascribed to the reopening of the conduit. Fischer et al. (1994) and Stix et al. (1997) have shown that the seismicity associated with these events is consistent with brittle failure of the sealed carapace at the top of the magma body. Once the strength of the impermeable seal has been exceeded, gas can freely escape through the permeable conduit and interaction of the gas with the hydrothermal system is minimised. Retainment of gases beneath an impermeable seal will allow the hydrothermal system to dominate the gas chemistry by selective absorption of more water-soluble gas species such as HCl.

On the short time scale of hours and minutes, SO_2 fluxes appear to be highly variable. The observation that SO_2 flux increases are seen after rockfalls is consistent with small slugs of gas released from pore spaces in the dome, bubble and fracture networks. Hybrid and long-period earthquakes and harmonic tremor do not appear to be *consistently* associated with any immediate changes in SO_2 emission rate. The RSAM is composed of many different seismic signals, some of which have no near-surface expression, i.e. conduit earthquakes may not be associated with immediate emission of gas at the surface, for example. In order to confirm a relationship between high- and low-frequency seismic signals, specifically long-period and hybrid earthquakes and rockfall events, with SO_2 emission, a higher temporal resolution and more accurate SO_2 emission rate measurements are required. On this short time scale, SO_2 fluctuations have been associated with Strombolian explosions on Pacaya Volcano, Guatemala (Andres et al., 1993), among other volcanoes, with SO_2 peaks being measured after the start of a Strombolian eruption and decreasing afterwards on time scales of minutes.

5. Sulphur dioxide monitoring as a long-term forecasting tool

Sustained, or post-collapse, SO_2 emission rates of > 500 t day^{-1} during the residual period (July and December 1998; January and September 1999) provided one of the clearest indications that the eruption of the Soufrière Hills Volcano was not waning. In the absence of magma reaching the surface and significant seismicity, SO_2 emissions remained high and variable through 1998 and 1999, indicating the continued degassing of a sulphur-rich source at magma chamber depths (albeit inhibited in its passage to the surface by the sealing of the edifice). The variability of the SO_2 emission rate during this period was directly related to conduit and dome permeability. It seems likely that SO_2 emissions will decrease once the magma supply from depth has ceased. After a length of time determined by the volume of residual magma in the chamber, its sulphur content and the permeability of the edifice, SO_2 emission rates will decrease (to less than 50 t day^{-1}) and large gravitational collapse events from the cooling dome will be associated with decreasing SO_2 emissions. This will indicate that the sulphur supply from depth is decreasing or has become less constant and more sporadic. This is a criterion for recognising the imminent end of the eruption.

6. Conclusions

The variability of the SO_2 flux from the Soufrière Hills Volcano, both before and after major eruptive episodes, points to a volcano that is not degassing freely for all of the time and is subject to periodic episodes of sealing and pressurisation. A constant supply of sulphur from depth is seen at the surface as a highly variable emission of SO_2. This variability arises from magmatic and, at times, hydrothermal processes occurring at shallow depths, up to 1 or 2 km but probably mainly in the few upper hundreds of metres of the conduit and dome. This variability allows us to evaluate how 'open' the system is on time scales of days to years (Fig. 9).

The first phase of dome building exhibited periods of 'closed' system behaviour with respect to gas through-flow, which is reflected in SO_2 emissions after large dome and sector collapse events. The second phase of dome building appears to be exhibiting generally more 'open' behaviour, reflected in the lack of large SO_2 emissions after large dome collapses. Explosive activity after these events appears to be unlikely at this stage of the eruption, although the data suggest that the system requires only small changes in conduit permeability and overpressure in order to revert back to this style of activity. The controls on SO_2 emission to the atmosphere appear to be almost en-

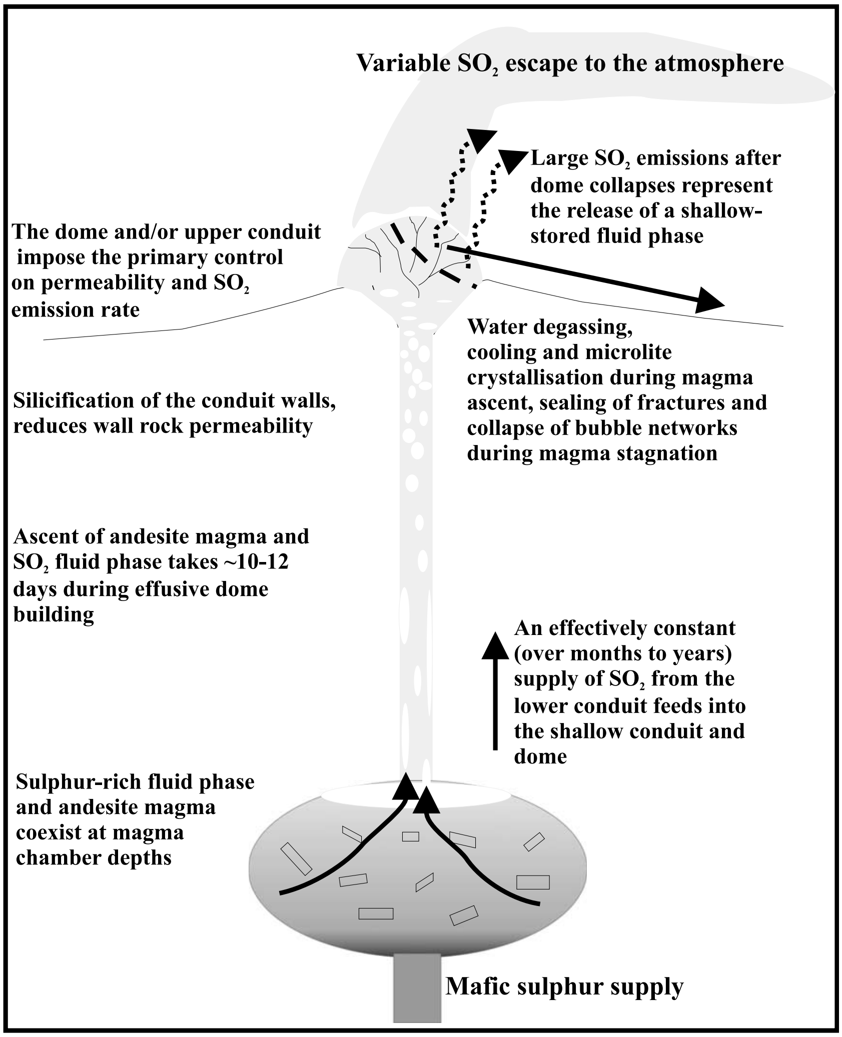

Fig. 9. A schematic diagram to show the controls on SO_2 emission rate to the atmosphere at Soufrière Hills Volcano, Montserrat.

tirely within the dome during phases of dome growth and in the upper conduit during periods when the vent is exposed (during the explosions in August–October 1997 and in April–June 1999).

Until the temporal resolution of SO_2 monitoring is improved, SO_2 emission rate monitoring at Soufrière Hills Volcano is primarily most useful as a long-term indicator of activity (months to years). It should be possible to recognise a cessation of sulphur supply from depth easily, once dome collapses and other large events cease to be associated with large emissions of SO_2. This implies that we can identify, through the monitoring of SO_2, the cessation of mafic magma supply from depth. Once this has ceased we can reasonably expect the surface extrusion of andesite lava to cease. Specifically, we will be able to evaluate whether the eruption has stopped or has merely paused, an important goal of hazard assessment and land use planning around an active volcano.

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