

Monitoring SO₂ emission at the Soufriere Hills volcano: Implications for changes in eruptive conditions

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Abstract. Correlation spectrometer measurements of sulfur dioxide (SO₂) emission rates during the current eruption of the Soufriere Hills volcano, Montserrat, have contributed towards identifying different phases of volcanic activity. SO₂ emission rate has increased from <200 tonnes per day (td⁻¹; <2.3 kgs⁻¹) in the early stages of dome growth to >550 td⁻¹ (>6.4 kgs⁻¹) after July 1996, with the uncertainty associated with any individual measurement ca. 30%. Significantly enhanced SO₂ emission rates have been identified in association with early phreatic eruptions (800 td⁻¹ (9.3 kgs⁻¹)) and episodes of vigorous dome collapse and pyroclastic flow generation (900 to 1500 td⁻¹ (10.4 to 17.4 kgs⁻¹)). SO₂ emission rate has proved a useful proxy measurement for magma production rate. Observed SO₂ emission rates are significantly higher than those inferred from analyses of glass inclusions in phenocrysts, implying the existence of a S-rich magmatic vapour phase.

Introduction

The eruption of the Soufriere Hills volcano on Montserrat [Young *et al.*, 1998] has been closely monitored using a variety of geological and geophysical tools. Several different approaches for measuring volcanic gases have been employed through the course of the eruption to date. Filter packs [Allen, 1996], diffusion tubes and measurements of acidity levels of rainfall and surface water [Norton, 1997] have been used to assess the environmental impacts of SO₂ on the island. Correlation spectroscopy (COSPEC), open-path Fourier Transform Infra-Red spectroscopy (OP-FTIR) [Oppenheimer *et al.*,

1998] and direct sampling of both dome gases and the associated hydrothermal system [Hammouya *et al.*, 1998] have been used to monitor volcanic activity as part of the day-to-day hazard assessment undertaken by the Montserrat Volcano Observatory (MVO). In this paper we discuss the COSPEC data and links between SO₂ emission rates and dome extrusion rates. We also demonstrate the presence of an excess sulfur phase above that calculated using the petrological method.

Methodology

COSPEC measurements of SO₂ emission rates were conducted between late July 1995 and late June 1997 with some extended breaks resulting from instrument absence or problems. Trade winds in the eastern Caribbean typically carry the volcanic gas plume westwards out to sea at the height of the volcano summit (ca. 1,000 m above mean sea level (amsl)). Occasional boat, airborne and fixed-position scanning measurements have been made; details of these methods will not be given here. COSPEC data was primarily collected during road traverses from 28 April 1996 until 24 June 1997, after which time emplacement of pyroclastic flows made the route unviable. Road traverses were along the west coast road (Figure 1), approximately perpendicular to plume trajectory, at a distance of 4 to 6 km from the crater. Emission rates are quoted as the average of several both north- and south-bound traverses made in a single day. Windspeeds were measured by a hand-held anemometer from Windy Hill (until July 1996) and from the more representative St. George's Hill (Aug 1996 to June 1997). Systematic differences in windspeed for the two sites as determined by simultaneous measurements result in the Windy Hill windspeed data being adjusted by a factor of 1.78. Cumulative uncertainty is estimated as 30%.

Sources of uncertainty in SO₂ emission rate measurements include variation in vehicle speed during measurement, operator variability during data collection and reduction, chart reading error, and instrument calibration [Millan and Hoff, 1978]. Through different operators undertaking duplicate road traverses and analysis of COSPEC graphic traces, we found the uncertainty to be similar to that reported for other volcanoes [Stoiber *et al.*, 1983, Daag *et al.*, 1996]. Measurement independent variables such as fluctuations in wind speed, changes in cloud cover and solar angle and variation in plume opacity resulting from clouds or aerosol scattering also lead to considerable variability between individual COSPEC measurements [Millan, 1980]. A further source of variation may be scavenging of SO₂ gas by water droplets forming the orographic cloud that persistently caps the volcano [Seigneur and Saxena, 1988].

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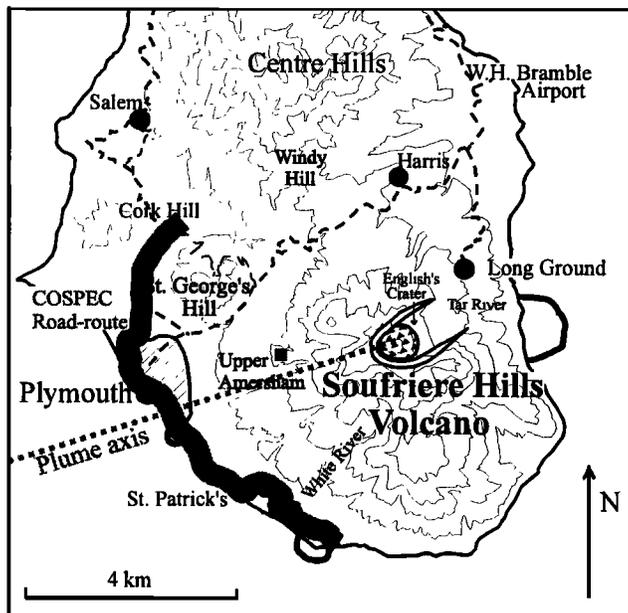


Figure 1. Montserrat showing the Soufriere Hills volcano, the capital town of Plymouth, the location of the coast road used for normal COSPEC runs, and other locations mentioned in the text.

Emission Rate Measurements Prior to Dome Extrusion

COSPEC measurements at Montserrat began on 29 July 1995 (Figure 2), a day after phreatic explosions opened the 28 July vent in English's Crater. Initial SO₂ values of 300 ± 150 td⁻¹ (3.5 ± 1.7 kgs⁻¹) were detected until 4 August when a value of 800 td⁻¹ (9.3 kgs⁻¹) was measured shortly after a seismically-defined steam emission event. This event heralded a seismic swarm on 5 to 6 August. Maximum SO₂ emission rate for a single run during the swarm was 1200 td⁻¹ (13.9 kgs⁻¹). The observed emission rates were greater than could plausibly result from oxidation of an hydrothermal system, and were used to infer that a magma body had been intruded to a shallow level in the volcano. This conclusion was reached well before the first extrusion of lava on 15 November 1995 [Young *et al.*, 1998] confirming the utility of COSPEC for volcano monitoring.

SO₂ Emissions During Dome Growth

SO₂ emissions associated with extrusive dome growth were measured between 28 April 1996 and 24 June 1997. Figure 2 summarises the time-series data set. Averaged over the period of observation, the daily average SO₂ emission rate (± 1 standard deviation) from Soufriere Hills volcano has been 436 ± 331 td⁻¹ (5.0 kgs⁻¹), ranging between 42 and 1933 td⁻¹ (0.5 and 22.4 kgs⁻¹). By contrast, emission rates of <200 td⁻¹ (2.3 kgs⁻¹) were observed during the comparable Mt Unzen eruption, Japan [Hirabayashi *et al.*, 1995]. There has been a general increase in SO₂ emission rate as the eruption has proceeded, consistent with an increasing magma extrusion rate (see below). Average emission rate between 28 April and 23 July 1996 was 202 ± 96 td⁻¹ (2.3 kgs⁻¹), increasing to an average of 554 ± 249 td⁻¹ (6.4 kgs⁻¹) from 24 July 1996 to 24 June 1997. Higher SO₂ emission rates cannot be closely related to periods of enhanced long period seismicity (Figure 2), even though

the likely cause of long-period seismicity is gas movement within the upper conduit and dome [Neuberg *et al.*, 1998].

Highest values of SO₂ emission rate were typically associated with large pyroclastic flow episodes (May, August 1996; January, April, June 1997; Table 1), as well as the major 17 September 1996 explosive event (implied from SO₂ diffusion tube data in the absence of COSPEC data). These elevated SO₂ emission rates are interpreted as resulting largely from the exposure of fresh, less degassed rocks in the avalanche scar, and from upwelling of new, largely undegassed, magma filling in the collapse scar volume. This was indicated by the major 17 September collapse, which led to a 45 minute-long sub-plinian explosive eruption [Robertson *et al.*, 1998].

Elevated SO₂ emission rates may also be the result of degassing of the pyroclastic flow deposits themselves. A traverse on 11 April 1997 within a few hours of major pyroclastic flows down the White River (Figure 1) showed two distinct peaks, indicating degassing from two separate sources; the summit region and the large area of freshly exposed hot pyroclastic flow deposits in the White River valley. Of the total measured emission rate ($1,524$ td⁻¹ (17.6 kgs⁻¹)), an estimated 35% emanated from the deposits. The unusual combination of wind direction and pyroclastic flow deposition on 11 April made these relationships clear; typically during the eruption, pyroclastic deposits were emplaced immediately up-wind of the vent, making it impossible to distinguish separate sources.

Data obtained from SO₂ diffusion tubes, which integrate ambient SO₂ concentrations over 2 week sampling intervals, quantitatively support the COSPEC data [Norton, 1997]. Average SO₂ concentrations for the period December 1995 to April 1997 at the Upper Amersham site (ground level, c. 2 km downwind of the dome) are 46 ppb, while the maximum recorded value (231.5 ppb) was for the period 17 September to 4 October 1996 which included a major explosive event. Routine measurements of enhanced acidity of rain water that has passed through the volcanic plume and of standing surface water beneath the plume [Norton, 1997] are also consistent with the COSPEC-derived SO₂ emission rate data.

Ratio of SO₂ vs. Magma Emission Rate During Dome Growth

During the course of the eruption to date, the extrusion rate of the dome has fluctuated, averaging ca. 2 to 3 m³s⁻¹ ($5,200$

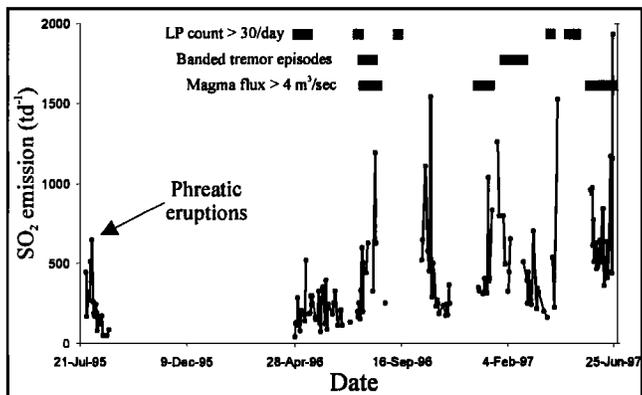


Figure 2. SO₂ emission rate (from COSPEC) and indication of periods of enhanced dome growth rate, occurrence of banded seismic tremor and enhanced long-period earthquake activity at the Soufriere Hills volcano, July 1995 to June 1997. Emission rate is the average of all runs for a single day.

Table 1. Elevated SO₂ Emission Rates Measured Following Partial Dome Collapses and Pyroclastic Flow Emplacement

| Date of emission rate measurement | Hours after collapse | SO ₂ emission rate above background ⁽¹⁾ | Volume of collapse (x10 ⁶ m ³) |
|-----------------------------------|----------------------|---|---|
| 13 May 1996 | ca. 24 | 259 % | 0.3 ⁽²⁾ |
| 12 August 1996 | ca. 24 | 275 % | 2 ⁽³⁾ |
| 13 August 1996 | ca. 48 | 128 % | un-measured |
| 13 August 1996 | ca. 51 | 159 % | un-measured |
| 9 January 1997 | 10 | 261 % | < 1 |
| 14 January 1997 | ca. 24 | 209 % | < 1 |
| 21 January 1997 | 20 | 317 % | 1.6 ⁽³⁾ |
| 11 April 1997 | 4 | 382 % | 2.3 ⁽²⁾ |

(1) Background calculated as average daily emission rate for 5 days before and after the date of the high post-collapse emission rate.

(2) Volume from surveyed deposits.

(3) Volume of collapse scar.

kgs⁻¹) up to May 1997 but generally increasing and reaching peaks in excess of 10 m³s⁻¹ (26,000 kgs⁻¹) [Sparks *et al.*, 1998]. Several different cycles of activity with variations in seismicity and eruptive style have occurred. SO₂ emission rate has increased consistently with magma extrusion rate so that during most of the eruption, an average of 680 ± 360 g of SO₂ have been erupted per tonne of magma (Figure 3).

SO₂ emission rates in October and November 1996 fell outside the general pattern as there was neither rapid dome growth nor enhanced seismicity to accompany the high observed gas:magma emission rate ratio (up to 3,000 g of SO₂ per tonne of magma). This period followed the explosion of 17 September, so the enhanced degassing may have been due to deep seated changes in the pressure regime of the magma storage area. Other periods of higher gas/magma emission rate ratio generally occurred immediately prior to collapse events (e.g. late-July 1996, early-April 1997 and late-June 1997; Figure 3). These periods of higher gas emission rates may correspond to periods of enhanced magma emission which were too short to be detected by the bi-weekly to monthly dome volume surveys.

For comparison, average magma emission rate at Mt. Unzen ranged between ca. 3 m³s⁻¹ (7,800 kgs⁻¹) in 1991 to ca. 1 to 2 m³s⁻¹ (2,600 to 5,200 kgs⁻¹) during 1992 and 1993, and 130 to 450 g of SO₂ were erupted per tonne of magma for most of the eruption, the ratio varying with time [Hirabayashi *et al.*, 1995]. Thus overall the Soufriere Hills lava dome has degassed significantly more SO₂ per volume of magma erupted than at Mt. Unzen and the ratio has stayed constant with time.

The generally consistent correlation between gas and magma emission rates has proven a useful monitoring tool, given the near-daily availability of COSPEC data and the difficulty of making visual observations and measurements of the dome during prolonged periods of cloud cover; SO₂ emission has acted as a general proxy for dome growth rate.

The Sulfur 'Budget'

The Soufriere Hills lava is a porphyritic andesite (58 to 60% SiO₂). Throughout the eruption of more than 250 x 10⁶ m³ of lava, the range of compositional variation has been very

limited [Devine *et al.*, 1998]. Any interpretation of SO₂ emission rates and especially their relationship to magma production rate necessarily requires knowledge of the potential sources of S and estimation of their possible contributions to the SO₂ emission rate measured at the surface. Sulfur concentrations measured in glass inclusions in quartz, orthopyroxene and plagioclase phenocrysts were uniformly less than the limits of resolution of the electron microprobe (<150 ppm) at the appropriate operating conditions. Chlorine concentrations were of the order of 2300 to 4200 ± 200 ppm. S and Cl concentrations were also measured in glasses from the August 1996 pyroclastic flows and 17 September sub-plinian pumice. These showed negligible S but Cl concentrations of 2000 ± 200 ppm. The S/Cl ratio (0.5 to 0.05) determined from comparison of glass inclusions and degassed glass is somewhat higher than the results of OP-FTIR observations (S/Cl ratios of <0.05 [Oppenheimer *et al.* 1998]). It is, however, consistent with filter pack data obtained within the plume (S/Cl ratios of 0.5 to 0.35 [Allen, 1996]) and with data from directly-sampled dome emissions (S/Cl ratio of ca. 0.2; [Hammouya *et al.*, 1998]).

Following the methodology of Gerlach and McGee [1994] the glass inclusion concentrations can be used to determine SO₂ degassing expected as the result of magmatic extrusion. A maximum value of 150 ppm in the melt and complete degassing of S during ascent gives ca. 95 g of SO₂ per tonne of extruded magma (assuming a melt fraction of 35% [M. Murphy, personal communication, 1998]). At the typical sustained extrusion rates of around 3 m³s⁻¹ calculated for this eruption, this equates to a daily emission rate of ca. 65 td⁻¹ (ca. 0.75 kgs⁻¹). This indicates the need for an additional source of SO₂ in the Soufriere Hills magmatic system. Similar 'excess' sulfur emission rates were observed during lava dome growth at Redoubt [Gerlach *et al.*, 1994] and Unzen [Hirabayashi *et al.*, 1995], but they are very much smaller than those observed for Pinatubo [Wallace and Gerlach, 1994] and El Chichon [Luhr, 1990].

While detailed analysis of these sources of S is beyond the scope of this current paper, potential sources include:

- presence of anhydrite/pyrrhotite as a minor phenocryst phase in the magma,
- the associated hydrothermal system,
- an S-bearing vapour phase co existing with the magma and
- a less evolved S-rich melt introduced into the system at depth.

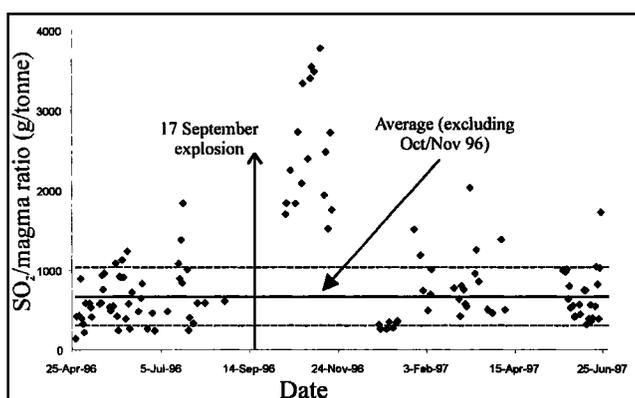


Figure 3. Ratio of SO₂ emission rate (from COSPEC) and magma emission rate (from bi-weekly to monthly dome volume measurements), plotted against time, excluding enhanced SO₂ emission rate after pyroclastic flow episodes (see Table 1).

No evidence has been found for the presence of anhydrite as a co-existing phenocryst phase and pyrrhotite only exists as small inclusions in Fe-Ti oxides [M. Murphy, personal communication, 1997]. Possible association of increased SO₂ emission rate with banded tremor (Figure 2) suggests some link to the hydrothermal system, although high SO₂ emission rates have been observed without the occurrence of banded tremor and vice-versa. Direct sampling of dome gases [Hammouya *et al.*, 1998] produced little evidence for hydrothermal input. Mafic inclusion glasses and associated phenocryst phases are also relatively sulfur poor, although S may have escaped into an associated vapour-phase during intrusion. Experimental phase equilibria [Barclay *et al.*, 1998] and glass inclusion studies have shown the magma to be vapour-saturated at depth and it is likely that this co-existing vapour phase contains a sulfur component.

It is postulated that some of the SO₂-enriched vapour phase remains stored in the dome after extrusion, and is released upon dome collapse as SO₂ emission rates commonly remain high for periods up to 50 hours following a collapse (Table 1). Thus overall correlation between magma extrusion and SO₂ emission rates can be accounted for by the partial degassing of the total S system during ascent, which may cause much of the seismic activity recorded at shallow depths within and beneath the dome [Neuberg *et al.*, 1998], and then continued degassing as collapse occurs and new magma is intruded to fill the scar. Work is in progress to further quantify the composition and degassing behaviour, and to determine the contribution of any co-existing vapour phase and its source to the eruption dynamics.

Conclusions

Practical difficulties dictate that direct samples of volcanic gases are impossible to obtain consistently during active dome growth; thus remote techniques such as COSPEC measurements provide unique insights into the role of volatiles in an eruption. Given the many sources of variation in measurements, a long time-series of SO₂ emission rate is essential to quantify changing styles of eruptive activity and to draw inferences concerning the sulfur budget of the volcano and changing conditions in the magma chamber. As part of a multi-disciplinary monitoring effort, we have found that remote SO₂ monitoring can provide a proxy for magma emission rate and an early-warning for changes in eruptive style. Consideration of sulfur budget for the volcano, along with seismic and deformation data, suggests that degassing of an excess of S in the magma occurs in the upper conduit and dome during ascent.

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