

Periodic sulphur dioxide degassing from the Soufrière Hills Volcano related to deep magma supply

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Abstract: Soufrière Hills Volcano produced prodigious quantities of sulphur dioxide (SO₂) gas throughout 1995–2013. An unprecedented, detailed record of SO₂ flux shows that high SO₂ fluxes were sustained through eruptive pauses and for two years after the end of lava extrusion and are decoupled from lava extrusion rates. Lava extrusion rates have exhibited strong 1- to 2-year cyclicity. Wavelet analysis demonstrates periodicities of c. 5 months and c. 2 years within the SO₂ time series, as well as the shorter cycles identified previously. The latter period is similar to the wavelength of cycles in lava extrusion, albeit non-systematically offset. The periodicities are consistent with pressure changes accompanying deformation in a coupled magma reservoir system whereby double periodic behaviour may arise from limited connectivity between two reservoirs. During periods of lava extrusion SO₂ is released together with the lava (yielding the c. 2 year period), albeit with some offset. In contrast, when magma cannot flow because of its yield strength, SO₂ is released independently from lava (yielding the c. 5 month period). Our results have implications for eruption forecasting. It seems likely that, when deep supply of magma ceases, gas fluxes will cease to be periodic.

Measurements of volcanic sulphur dioxide (SO₂) degassing processes are an important tool for volcano monitoring (Casadevall *et al.* 1983; Bluth *et al.* 1994; Fischer *et al.* 1994; Young *et al.* 1998a; Aiuppa *et al.* 2009; Werner *et al.* 2013). Sulphur gas emissions from volcanoes have been used to forecast eruptions as well as to assess the level of activity, the extrusion rate, the characteristics of the plumbing system and the pressurization of shallow magma reservoirs during an eruption (Casadevall *et al.* 1983; Caltabiano *et al.* 1994; Gerlach & McGee 1994; McGee & Sutton 1994; Hirabayashi *et al.* 1995; Luckett *et al.* 2002; Zobin *et al.* 2008). In recent years extended, temporally resolved time series of daily SO₂ emissions have been generated

following new developments in low-cost automated spectrometer networks that operate in the ultraviolet region of the electromagnetic spectrum. This allows for the capture of spectra containing characteristic absorption features owing to SO₂ every few minutes during daylight hours, which are then processed using differential optical absorption spectroscopy and combined with plume speed to produce SO₂ fluxes (e.g. Edmonds *et al.* 2003a; Galle *et al.* 2003; McGonigle *et al.* 2003). Soufrière Hills Volcano was the first locus of this volcano-monitoring development worldwide (Edmonds *et al.* 2003a; Christopher *et al.* 2010).

A consequence of the automated spectrometer network at Montserrat is an unprecedented

14 year-long time series of SO₂ emissions from the Soufrière Hills Volcano, preceded and supplemented by seven years of correlation spectrometer (COSPEC) data from 1995 to 2002 (e.g. Young *et al.* 1998a; Watson *et al.* 2000; Gardner & White 2002; Young *et al.* 2003). Over the course of the eruption, the time series has been used for volcano monitoring and has lent insights into magma supply, as well as conduit and dome permeability (Edmonds *et al.* 2001, 2003b, 2010; Young *et al.* 2003; Christopher *et al.* 2010; Nicholson *et al.* 2013). The SO₂ data have also been used as a hazard assessment tool (e.g. Komorowski *et al.* 2010). There are aspects of the patterns of SO₂ degassing from this volcano, however, that are not well understood: the long time-scale periodicity that is apparent in the data and the lack of a coupling between degassing and lava extrusion rates. These features of the SO₂ flux time series are the focus of this paper and may be a critical part of understanding the deep plumbing system.

Geological setting

The Soufrière Hills Volcano is an andesitic lava dome complex on the island of Montserrat in the Lesser Antilles arc. The current eruption began on 18 July 1995 with ash venting, followed by phreatic explosions over the following weeks and months (Young *et al.* 1998b; Robertson *et al.* 2000). Juvenile material was erupted around 15 November 1995 (Young *et al.* 1998b), building the first lava dome of the eruption. Over its 19-year history, the eruption has been characterized by discrete periods of lava extrusion (extrusion phases numbered I–V) and associated dome growth separated by periods of no lava extrusion referred to as pauses (Table 1).

Extrusion episodes have been punctuated by dome collapses and periods of Vulcanian explosions, generating pyroclastic flows and ash columns and co-ignimbrite clouds. Volcanic activity has been described in detail elsewhere (e.g. Aspinall *et al.*

1998; Miller *et al.* 1998; Young *et al.* 1998b; Calder *et al.* 2002; Norton *et al.* 2002; Sparks & Young 2002; Herd *et al.* 2005; Loughlin *et al.* 2010; Ryan *et al.* 2010; Wadge *et al.* 2010; Odbert *et al.* 2014; Stinton *et al.* 2014; Wadge *et al.* 2014). The first three phases of dome building were characterized by extended periods (on the order of 20 months) of extrusion and pause (Fig. 1). Phases IV and V were much shorter: phase IV was characterized by two separate pulses of magma extrusion (IVa and IVb; Fig. 1; Table 1). Both subphases had low extrusion rates (Wadge *et al.* 2010, 2014) and were punctuated by explosions (Komorowski *et al.* 2010). Phase V was also short in duration (early October 2009 to mid February 2010) and was characterized by frequent explosive activity (Stinton *et al.* 2014).

During each period of lava extrusion, ground deformation measured by Global Positioning System (GPS) and seismic activity correlated strongly with lava extrusion (Fig. 1). Over the course of the eruption, inflation of the ground's surface has been observed during pauses and deflation during periods of lava extrusion, which has been interpreted as the result of a continuous deep supply to a lower magma chamber at *c.* 12 km, with a pulsed supply into and eruption rate out of a shallow chamber at *c.* 6 km (Elsworth *et al.* 2008).

The Soufrière Hills andesite is the result of a complex magma genesis, dominated by magma mixing, fractional crystallization and crustal contamination during its long residence in an upper-mid crustal magma reservoir (Murphy *et al.* 1998, 2000; Zellmer *et al.* 2003; Humphreys *et al.* 2009, 2012; Christopher *et al.* 2014). It is likely that mingling between crystal-rich mushes, felsic melts and intruding mafic magmas is important for generating andesite, with vertically protracted small bodies distributed through the upper crust (Cassidy *et al.* 2012). This has also been proposed for other andesitic systems (e.g. Fichaut *et al.* 1989; Sato *et al.* 1999; Martel *et al.* 2006; Kent *et al.* 2010; Ruprecht & Plank 2013).

Table 1. Overview of phases of lava extrusion and eruptive pauses 1995–2010 and the mean SO₂ flux for each

Eruptive phase	<i>n</i>	Mean daily flux (t/day)	Pause following	<i>n</i>	Mean daily flux (t/day)
I – 15 November 1995	68	569	I – 10 March 1998	112	699
II – 27 November 1999	563	471	II – 1 August 2003	698	561
III – 8 August 2005	604	429	III – 4 April 2007	482	620
IVa – 29 July 2008	77	923	IVa – 14 October 2008	50	800
IVb – 3 December 2008	32	451	IVb – 4 January 2009	258	627
V – 4 October 2009	7	379	V – 11 February 2010	1257	392

'Pause following' is the eruptive pause that immediately followed the eruptive phase in the same row; *n* is the number of days in the eruptive phase or pause.

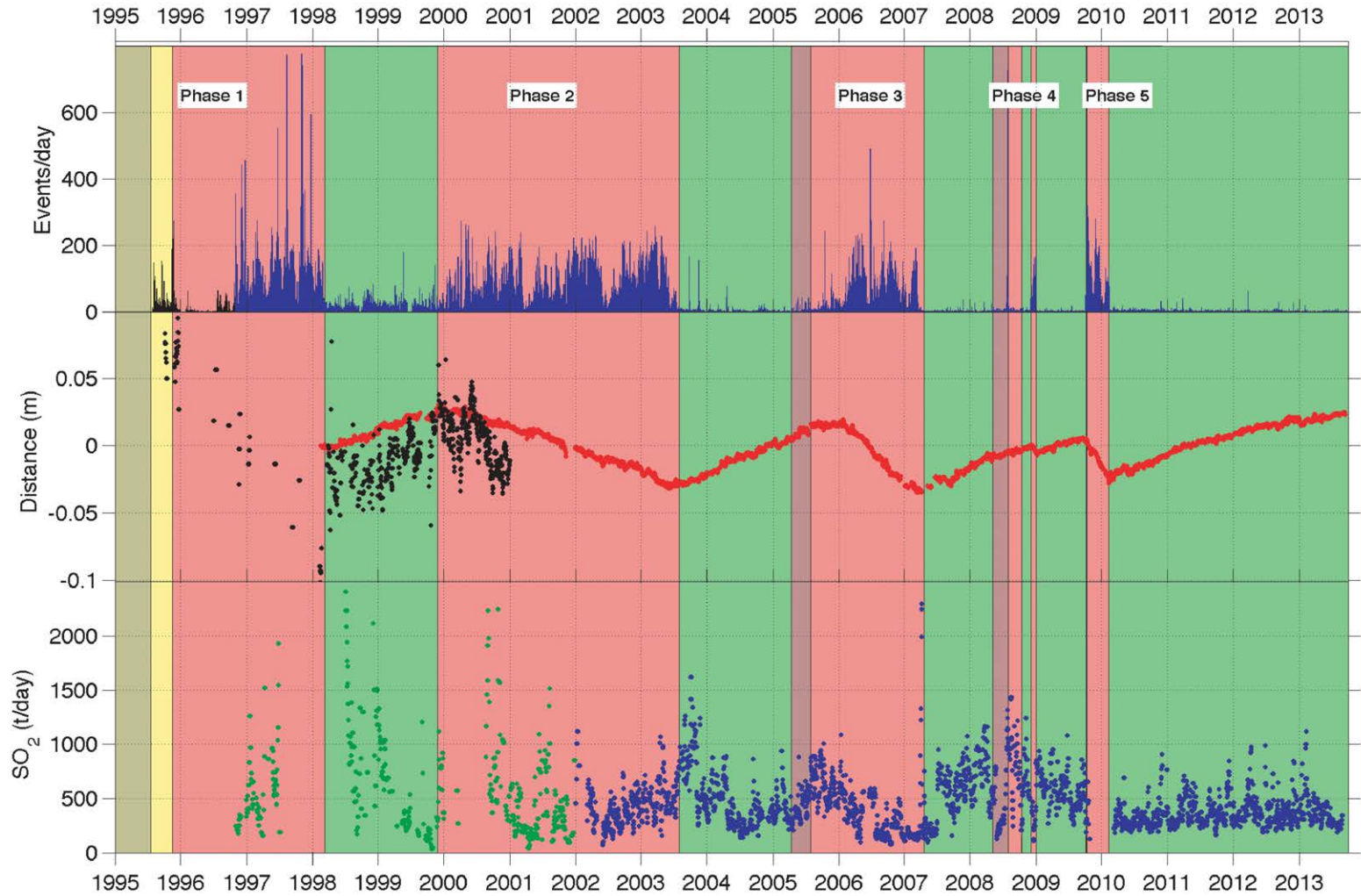


Fig. 1. Plots to show daily number of seismic events (top), height as measured by Global Positioning System (GPS) receivers, in metres (middle) and SO_2 flux (bottom) with time. Black dots are kinematic GPS data, red dots are fixed network GPS data. Green dots are data from COSPEC, blue dots are data from the UV spectrometer network. Extrusion phases are shaded in pink, pauses in green, transition periods in darker pink and phreatic activity in yellow.

The volatile budget of such an open system is likely to be complex. The low concentrations of sulphur in the plagioclase-hosted melt inclusions from the andesite (<100 ppm, Edmonds *et al.* 2001) are not sufficient to account for the mass of sulphur degassed during the eruption (which would require melt concentrations of >1000 ppm sulphur; Christopher *et al.* 2010), an observation common to many other intermediate-silicic volcanic systems, and referred to as 'the excess sulphur problem' (e.g. Westrich & Gerlach 1992; Wallace & Gerlach 1994; Wallace 2001; Wallace 2005; Shinohara 2008; Wallace & Edmonds 2011). Much of the sulphur in the system exists in a vapour phase prior to andesite magma ascent and eruption. A strong partitioning of sulphur into a hydrous vapour is consistent with thermodynamical and experimental studies for oxidized rhyolitic melts (Scaillet & Pichavant 2003; Moretti & Papale 2004). The vapour is probably replenished by intruding mafic magma, either through second boiling during crystallization of the basalt owing to the interaction of the two magmas or by overturn and mingling of a two-layer magma system, effectively distributing volatiles carried by deeper mafic magmas into the upper, more felsic levels of the system (Christopher *et al.* 2010; Edmonds *et al.* 2010, 2014).

The flux of SO₂ gas from the Soufrière Hills Volcano has varied on a range of timescales. The variability in lava flux and seismicity on timescales of hours to weeks has been attributed to a number of processes such as changes in shallow magma permeability during 'stick-slip' eruptive behaviour (Watson *et al.* 2000), changes in lava extrusion rate (Young *et al.* 1998a; Luckett *et al.* 2002) and sealing caused by the precipitation of silica in the conduit and dome during extrusive pauses (Edmonds *et al.* 2003b). Cycles with periodicities on the order of 6–8 weeks (50 days) and 10–14 days have been attributed to variations in lava extrusion and seismicity (Nicholson *et al.* 2013). Throughout the 1995–2010 eruption, however, SO₂ degassing has been high both during periods of no magma extrusion (pauses) and during periods of lava extrusion (Table 1). The degassing during eruptive pauses is clearly not due to ascending magma. In this case the gas must permeate upward, perhaps through a crystal-rich andesitic magma body, to the surface. A primary question we address here is the origin of the cyclic degassing.

Cyclic lava extrusion and degassing displaying periodicity on several different time scales are well documented at a number of different types of volcanoes (e.g. Denlinger & Hoblitt 1999; Voight *et al.* 1999; Voight *et al.* 2000; Barmin *et al.* 2002; Harris & Neri 2002; Sparks & Young 2002; Lautze *et al.* 2004; Harris *et al.* 2005; Sweeney *et al.* 2008; Oppenheimer *et al.* 2009; Wadge *et al.*

2010; Melnik & Costa 2014). Dome-building eruptions are commonly characterized by periodicity in magma discharge rates (Barmin *et al.* 2002) on timescales of hours to years. Cyclic patterns in seismicity, lava effusion and ground deformation were evident in phase I (1995–1998; see Table 1) (Miller *et al.* 1998; Voight *et al.* 1998, 1999; Robertson *et al.* 2000; Druitt *et al.* 2002; Lensky *et al.* 2008; Loughlin *et al.* 2010). For example, Voight *et al.* (1998) identified a 6–14 h inflation cycle during 1997, which correlated with degassing (Watson *et al.* 2000) while Druitt *et al.* (2002) reported cycles (10 h mean) of explosive activity in August 1997. More recently, Odbert *et al.* (2014) showed that cyclic seismicity and volcanic activity at the Soufrière Hills Volcano has periods ranging from sub-daily to multidecadal.

Aims of this paper

As of April 2014, the volcano is in its nineteenth year of quasi-continuous degassing and there remain aspects of the emission patterns of volcanic SO₂ from Soufrière Hills that are not well understood. The first is that, on a timescale of months to years, the emission of SO₂ is decoupled from the extrusion of lava (Fig. 1). Eruptive pauses can last up to >24 months, and during these relatively long periods SO₂ fluxes have been sustained at levels of >500 t/day for weeks to months. In other words, as much or even more SO₂ degasses when the volcano is not erupting as when it is erupting (Table 1); this observation requires some deep-seated process such as a change in permeability or advection of the gas phase to shallower levels to be operating, allowing gas to migrate through, or bypass, crystal-rich andesite regardless of whether or not andesite lava is being extruded. Lava extrusion ceased on 11 February 2010. It is unclear whether the present, continued high fluxes of SO₂ at the surface (>300 t/day; Table 1) since 2010 are indicative of a continued supply of mafic magma into the plumbing system, or whether they are sourced from a static, crystallizing or convecting magma body at depth.

In this paper, we present a time series of SO₂ data, from July 1995 to July 2014. We identify long timescale periodicity in the dataset that is independent of magma flux and we consider the source of such periodicity. In doing so we explore processes that may have influenced the flux of SO₂ that we measure at the surface, including gas scrubbing and modulation of degassing by the presence of lava domes, as well as deep magma supply, overturn and convection processes that may influence sulphur degassing at depth. We consider published numerical and conceptual models describing the

morphology of the plumbing system beneath the Soufrière Hills Volcano and what constraints the SO₂ emissions place on these schemes. We discuss the implications of the ongoing degassing for long-term hazard assessment.

Methodology

Sulphur dioxide emission rates

The SO₂ flux data were acquired using a COSPEC during 1995–2002 (Stoiber *et al.* 1987; Young *et al.* 2003) and using Ocean Optics UV spectrometers and differential optical absorption spectroscopy during 2002–2014 (Platt 1994). Two automated and telemetered UV spectrometers, coupled with scanning devices, were installed on the western flanks of the Soufrière Hills Volcano at Lovers Lane and Brodericks (Fig. 2), which acquire spectra from 8 a.m. to 4 p.m. each day, thus providing daily flux measurements with an estimated error of <35% (Edmonds *et al.* 2003a; Galle *et al.* 2003). Details on the installation of the network, instrument specifications, spectral evaluation, mass flux calculation, associated errors and other information relating to the Soufrière Hills UV scanning

network are described in detail by Edmonds *et al.* (2003a).

Time series analysis

Volcanic SO₂ flux time series typically contain error-induced noise that makes it difficult to recognize and constrain periodicities without a statistical analysis. The time series also contains gaps that render statistical analysis by simple Fourier transform methods non-effective. Nicholson *et al.* (2013) conducted an analysis of a subset of this time series and filled gaps in their dataset using two different techniques: gaps less than 14 days were filled using linear interpolation, while reconstructive analysis methods were used to fill the larger gaps. They used a short-term Fourier transform analysis approach, which employs a sliding window to measure how the spectral properties of the data change with time. Such analyses can be configured to have varying degrees of overlap between successive data windows. Window lengths of 256–128 days and overlaps of 50–99% were used.

We use wavelet analysis in order to preserve continuous information about the temporal variability of the periodicities in the analysis, which is not

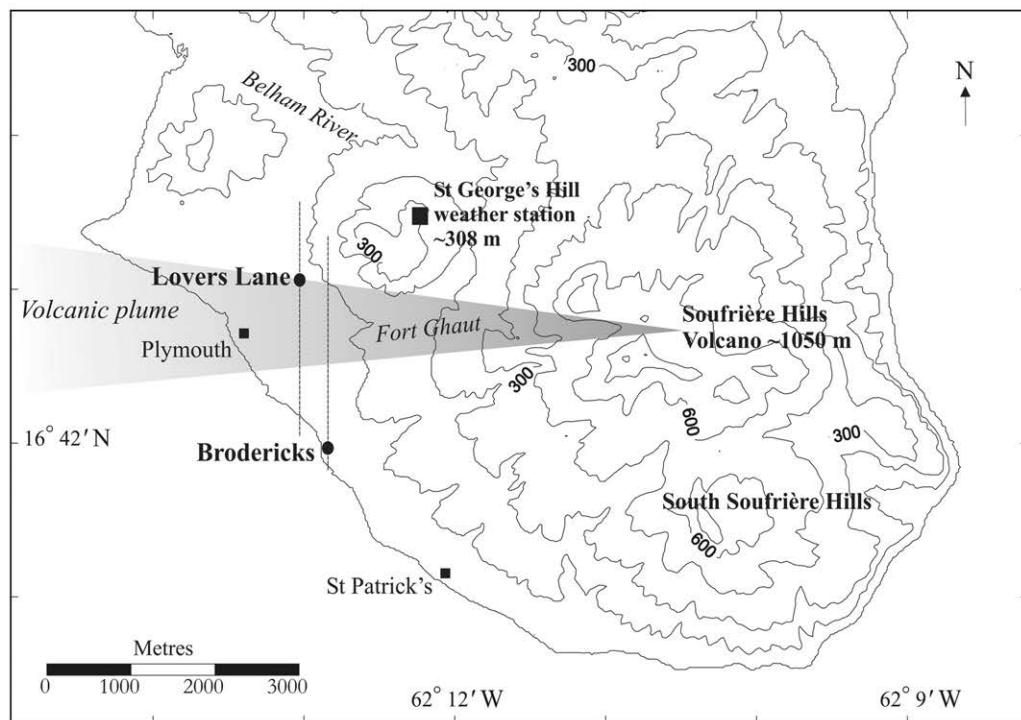


Fig. 2. Map of Montserrat showing the location of the two network spectrometers relative to the volcano: Lovers Lane and Broderick's. The prevailing winds are from the east and blow the plume over Plymouth. The bold line through each spectrometer position indicates the respective plane in which they scan.

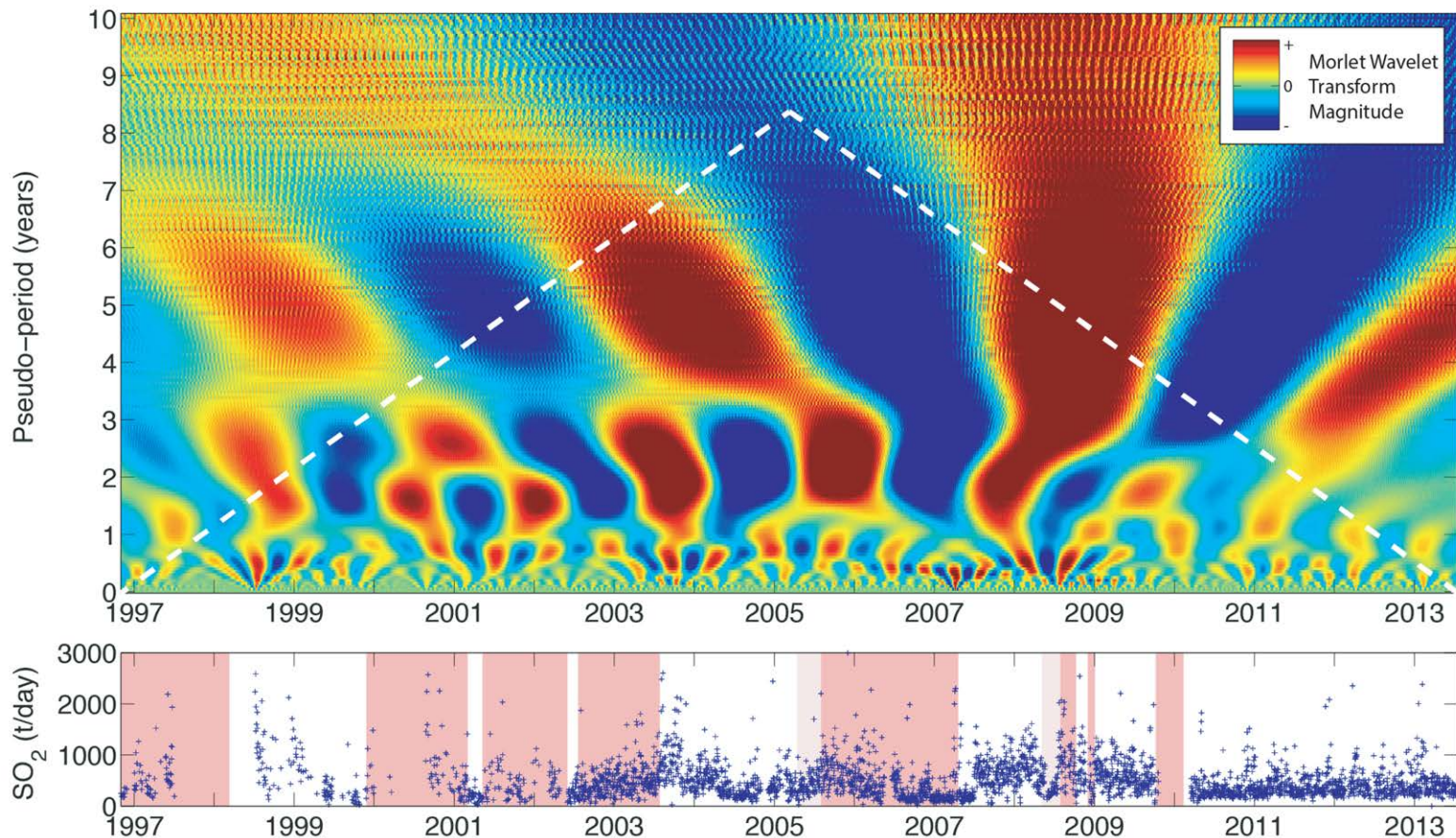


Fig. 3. Continuous wavelet analysis of the whole SO_2 dataset (including COSPEC and DOAS data) between 1997 and 2013. Transform values are obtained as an average of 100 versions of the original time series in which gaps are filled with random values, as described in the text. The upper panel indicates the wavelet transform value; strong positive values correlate strongly and positively with the Morlet wavelet, strong negative values correlate negatively. Pseudo-period, indicated on the left axis, corresponds to the average frequency of the convolved wavelet. White dashed lines indicate the distance from the region within 1 pseudo-period of the ends of the time series; regions of the transform prone to stronger edge-effects. Transform values outside the dashed lines should thus be regarded as less reliable. The lower panel shows the original dataset. Areas shaded in light and dark pink indicate phases of lava extrusion and transition phases, respectively.

possible using Fourier transform techniques. Previous studies have used wavelet analysis to constrain periodicities in degassing time series datasets obtained from other volcanoes (e.g. Oppenheimer *et al.* 2009; Boichu *et al.* 2010). Wavelet analysis breaks down a time series into time–frequency space, thus making it possible to determine both the dominant modes of variability and how those modes vary in time (Torrence & Compo 1998).

The continuous wavelet transform function (CWT) in Matlab was used, for our analysis. A CWT is computed by measuring the correlation between the signal and a wavelet (defined as a small wave with finite length and which integrates to zero). The wavelet is time-shifted and compressed or stretched to measure correlation as a function of time and frequency, respectively.

In our case, we choose the Morlet wavelet, which approximates to a sine wave and is suitable for analyses. A CWT may be illustrated as a 2D colour map that indicates the strength of correlation of the signal with the wavelet at different times and frequencies. Wavelet analysis was applied to the entire dataset after running the data through an 11-day median filter. The 11-day filter was chosen to smooth out any variations occurring on the order of a week or less. Gaps in the time series were filled using random values from a population with the same standard deviation as the data. The gaps in the COSPEC data (1995–2002) were too large to fill. A wavelet analysis of these random numbers (white noise) does not exhibit cycles and merely produces gaps in the plot. The plot generated by the wavelet analysis for the whole dataset (Fig. 3) confirms this, with the sparse nature of the COSPEC data producing a weaker signal in that part of the plot.

Periodicity of cycles in the sulphur dioxide degassing time series

General description of the SO₂ flux time series

Over the dataset as a whole, there are five long period cycles of SO₂ on the order of years that

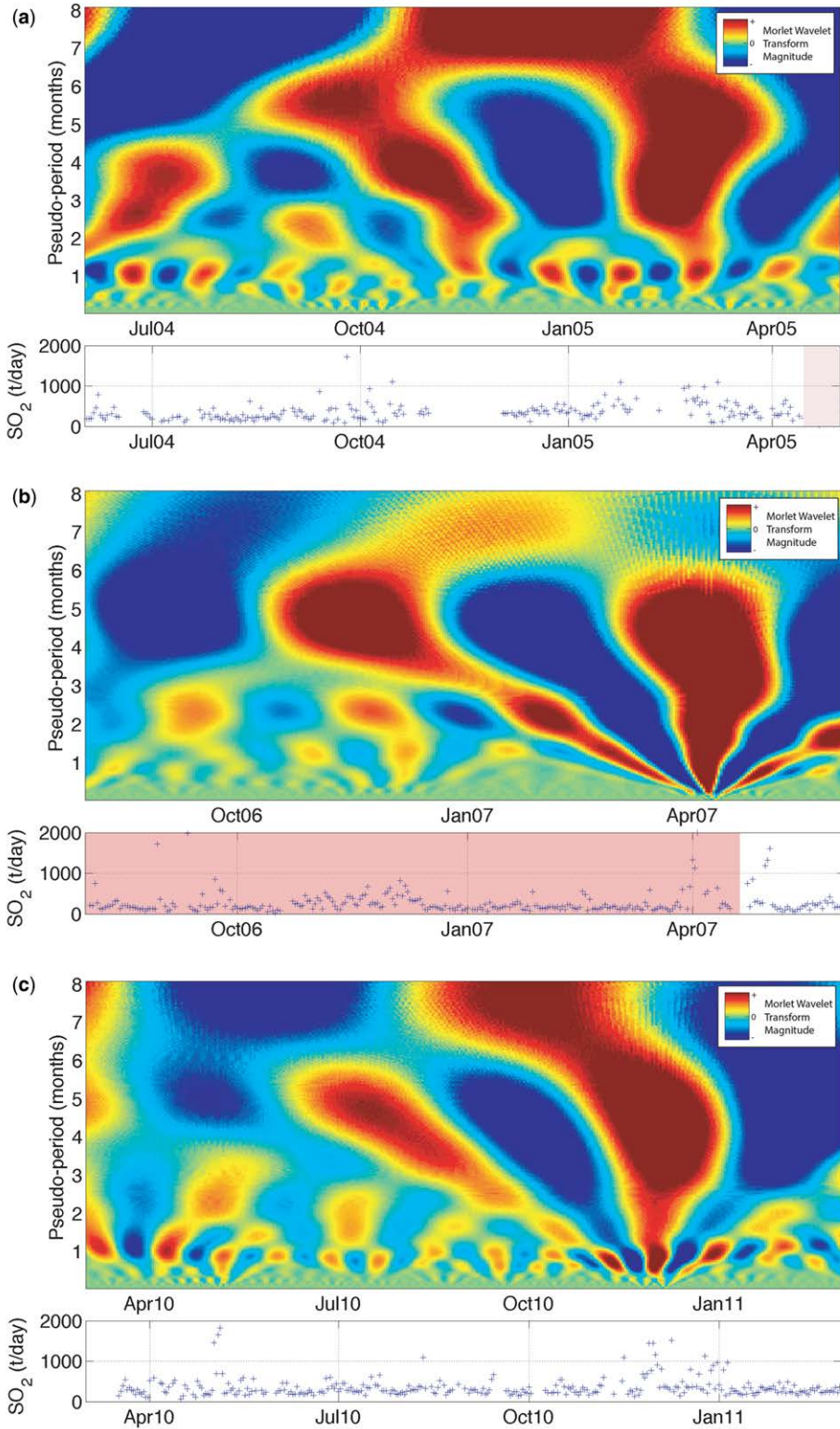
bear no obvious relation to the extrusion periods, three of which occurred while the network was present (Fig. 1). The mean daily SO₂ flux during each extrusion and pause episode is given in Table 1. It can be clearly seen that there is no correlation between lava extrusion rate and daily SO₂ flux. In fact, up until the end of phase V, the mean daily SO₂ flux was actually generally higher for the pause periods, when no lava extrusion occurred (Table 1). The three cycles captured by the network are all characterized by peak SO₂ fluxes above the mean daily average for the whole eruption (*c.* 500 t/day) separated by periods of lower SO₂ output (*c.* 300–400 t/day) lasting approximately 12 months (Fig. 1, Table 2). The first of the 1.5–2.3 year cycles recorded by the spectrometer network began in 2002 (Table 2). The peak of the cycle occurred in late 2003 and the highest recorded mean daily flux was 3592 t/day on 18 December 2003 (Fig. 1). The second cycle began in 2005 and had a maximum daily flux of 3218 t/day on 2 December 2005. The third cycle occurred during 2007–2009. It produced a maximum flux of 4598 t/day SO₂ on 9 September 2008.

During the troughs separating the peaks in these cycles, SO₂ fluxes were in general below the long-term eruption average. The average daily flux during the first trough in 2004–2005 was 404 t/day. During the second trough, in 2006–2007, the daily average flux was 304 t/day. Although the sustained average daily flux was low during these troughs, there were sporadic daily flux values that well exceeded the eruption daily average (Fig. 1). High values of 13 000 and 4000 t/day were recorded in the first and second troughs, respectively, with the former value being the highest daily SO₂ flux value recorded for the whole eruption. Removal of this value from the first trough gives a daily average of 311 t/day, which is indistinguishable from the average flux measured during the second trough in 2006–2007. Another feature of note is the higher mean daily SO₂ flux and longer duration of the third cycle relative to the previous two (Table 2).

Table 2. Summary of the length and mean SO₂ output for each cycle, where *n* is the number of SO₂ flux data points for each cycle, *f* is the fraction of days in each cycle that SO₂ flux data is available and *d* is the duration of the cycle in days

Cycle number and dates	Mean SO ₂ flux, t/day	Max flux date	Max flux	<i>n</i>	<i>f</i>	<i>d</i>
1 – June 2002 to April 2005	517	18 December 2003	3592	875	0.80	1092
2 – April 2005 to July 2007	464	2 December 2005	3218	752	0.82	919
3 – July 2007 to January 2010	664	9 September 2008	4599	732	0.87	842
From February 2010 to December 2013	394	26 March 2012	4594	1126	0.81	1386

The fourth row shows similar data for the period February 2010 to the end of December 2013 for comparison, which is not part of a cycle.



Periodicity in the SO₂ flux signal

Wavelet analysis confirms the presence of both a long-period (*c.* 2 year) cycle and a shorter-period (4–5 months) cycle (Figs 3 & 4). The *c.* 2 year cycle represents the long wavelength cycles that we can ‘see’ in the time series, described above and in Table 2. The 4–5 month cycle cannot be discerned with the naked eye; however one important feature to note is that it persists during periods of no lava extrusion (Fig. 4). Thus both the 4–5 month cycle and the *c.* 2 year cycle are decoupled from lava extrusion. The plots also show an apparent 5-year cycle that is an artefact owing to edge effects and will be ignored. The wavelet analysis also highlights the smaller scale periodicity on the order of *c.* 50 days (Fig. 4), which has been documented by Nicholson *et al.* (2013).

Discussion

We first explore possible external forcing that may be responsible for generating the 4–5 month and the *c.* 1.5–2.3 year periodicity in the SO₂ time series, such as the effect of humidity variations on the depletion rate of SO₂ from the plume as well as changes in the degree of SO₂ scrubbing that can be controlled by changes in the groundwater level and hence rainfall. We also consider changes in the shallow permeability of the conduit and lava dome; for example, open vent conditions might promote high gas fluxes whereas the presence of a lava dome might subdue degassing. We then consider deeper processes that may modulate SO₂ solubility in the deeper magmatic system, based on published models of magma reservoir depths and connectivity (Melnik & Costa 2014).

Meteoric and groundwater SO₂ scrubbing

It has been shown that SO₂ may be removed from the volcanic plume during the entrainment of warm moist air into the plume (e.g. Oppenheimer *et al.* 1998; Rodriguez *et al.* 2008). SO₂ might also be removed prior to degassing, by the subsurface hydrothermal system via a process referred to as scrubbing (e.g. Doukas & Gerlach 1992; Sutton *et al.* 2001; Symonds *et al.* 2001; Gerlach *et al.* 2002; Duffell *et al.* 2003; Werner *et al.* 2006; Werner *et al.* 2012). These two processes involve the chemical interaction of volcanic SO₂ with water and oxygen, whereby the interaction of SO₂

with H₂O leads to a reduction of SO₂ and the formation of hydrosulphuric acid (H₂SO₃), a weak acid (Doukas & Gerlach 1992). This acid may be further slowly oxidized to form mild sulphuric acid (H₂SO₄).

Atmospheric water (humidity) may account for the extensive removal of SO₂ from the Soufrière Hills volcanic plume, (10^{-3} s^{-1}) at plume ages >10 min (e.g. Oppenheimer *et al.* 1998). A later study showed that removal rates can be lower (10^{-4} s^{-1} ; Rodriguez *et al.* 2008), but was carried out during the dry season. The fixed UV spectrometers typically measure the plume within 5 min of the gas being emitted from the vent and Rodriguez *et al.* (2008) propose that the current network may underestimate the quantity of SO₂ in the plume by up to 70% on some days. These studies combined suggest that a systematic variation in humidity throughout the year might influence the mass of SO₂ present in the plume in a systematic way. The relative humidity is heavily dependent on the rainy season. Such a control on the SO₂ loss would be modulated by the wet and dry seasonal changes that occur on the island, with the wet season coinciding with the annual Atlantic hurricane season. Increased rainfall would act to increase the moisture content of the atmosphere and would be expected to enhance the rate at which SO₂ is removed from the plume. The rainy season may also act to increase groundwater levels and hence promote scrubbing in the subsurface hydrothermal system. Geochemical studies of spring waters have revealed the existence of a large hydrothermal/geothermal system in the southern portion of the island, beneath the Soufrière Hills Volcano (e.g. Chiodini *et al.* 1996). The hot springs or ‘Soufrières’ (fumaroles) on the flanks of the volcano prior to the onset of the current eruption contained sulphate and chloride species, indicative of dissolution of magmatic gases into the water, or ‘scrubbing’ (Chiodini *et al.* 1996).

These processes are subannual and cannot account for the 1.5–2.3 year cycles in the SO₂ time series. They are, however, candidates for the 4–5 month cycle shown in Fig. 4. We performed wavelet analysis on two meteorological datasets from time periods overlapping with the SO₂ data. The first is the daily mean relative humidity data (Fig. 5), which is a proxy for removal of SO₂ from the plume and the second is daily rainfall (Fig. 6), which might be a proxy for groundwater levels and thus a proxy for the subsurface scrubbing of

Fig. 4. Continuous wavelet analysis of SO₂ data shown at higher resolution for three subsections of Figure 3. (a) shows data from June 2004 to April 2005, (b) from August 2006 to May 2007 and (c) from March 2010 to February 2011. Ornament otherwise as in Figure 3. Note that the transforms plotted here do not show an edge-effect mask, as in the other figures, since data are available outside the region shown in the three panels.

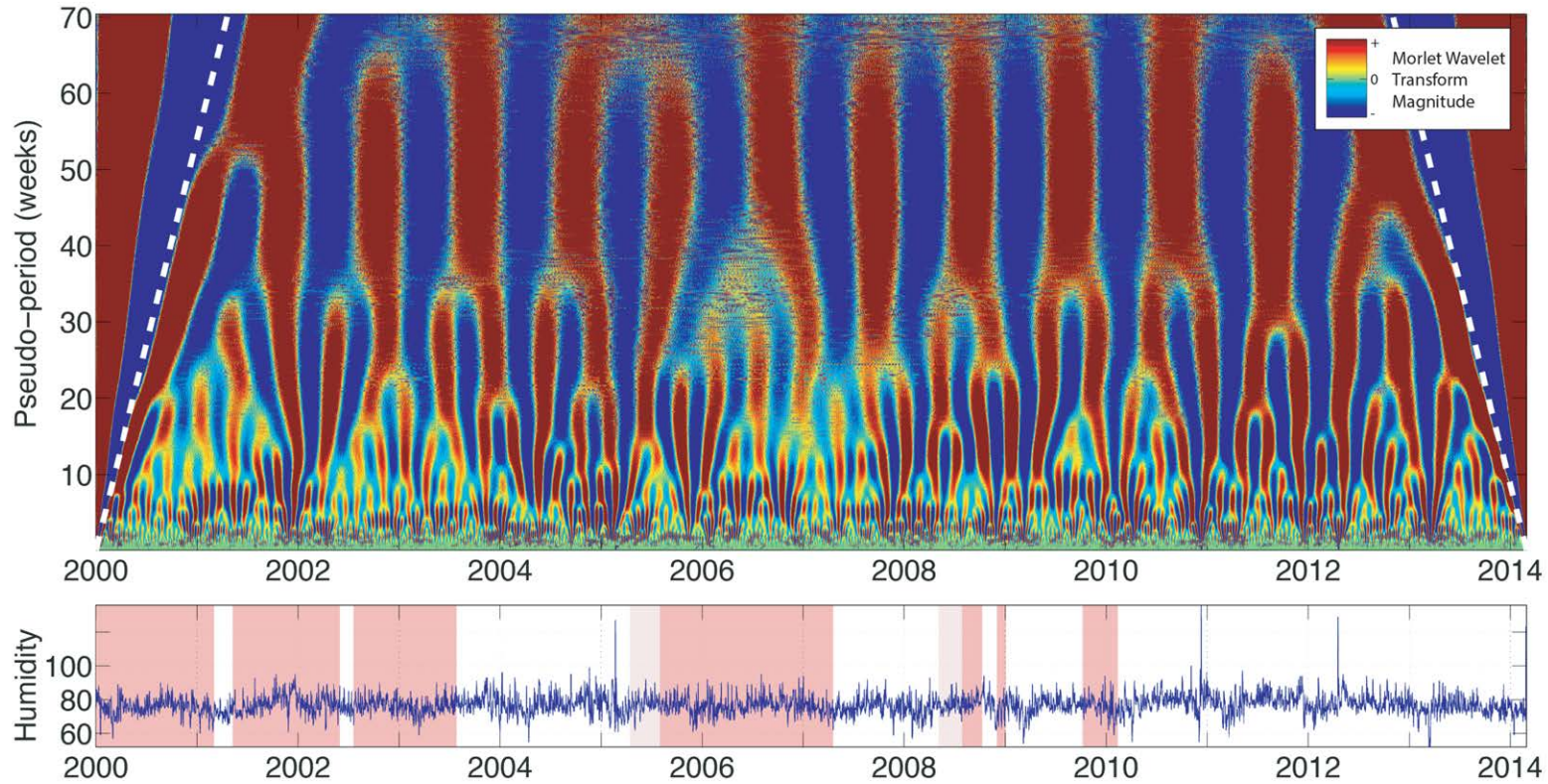


Fig. 5. Continuous wavelet analysis of the relative humidity data from 2000 to 2013. Ornament as in Figure 3.

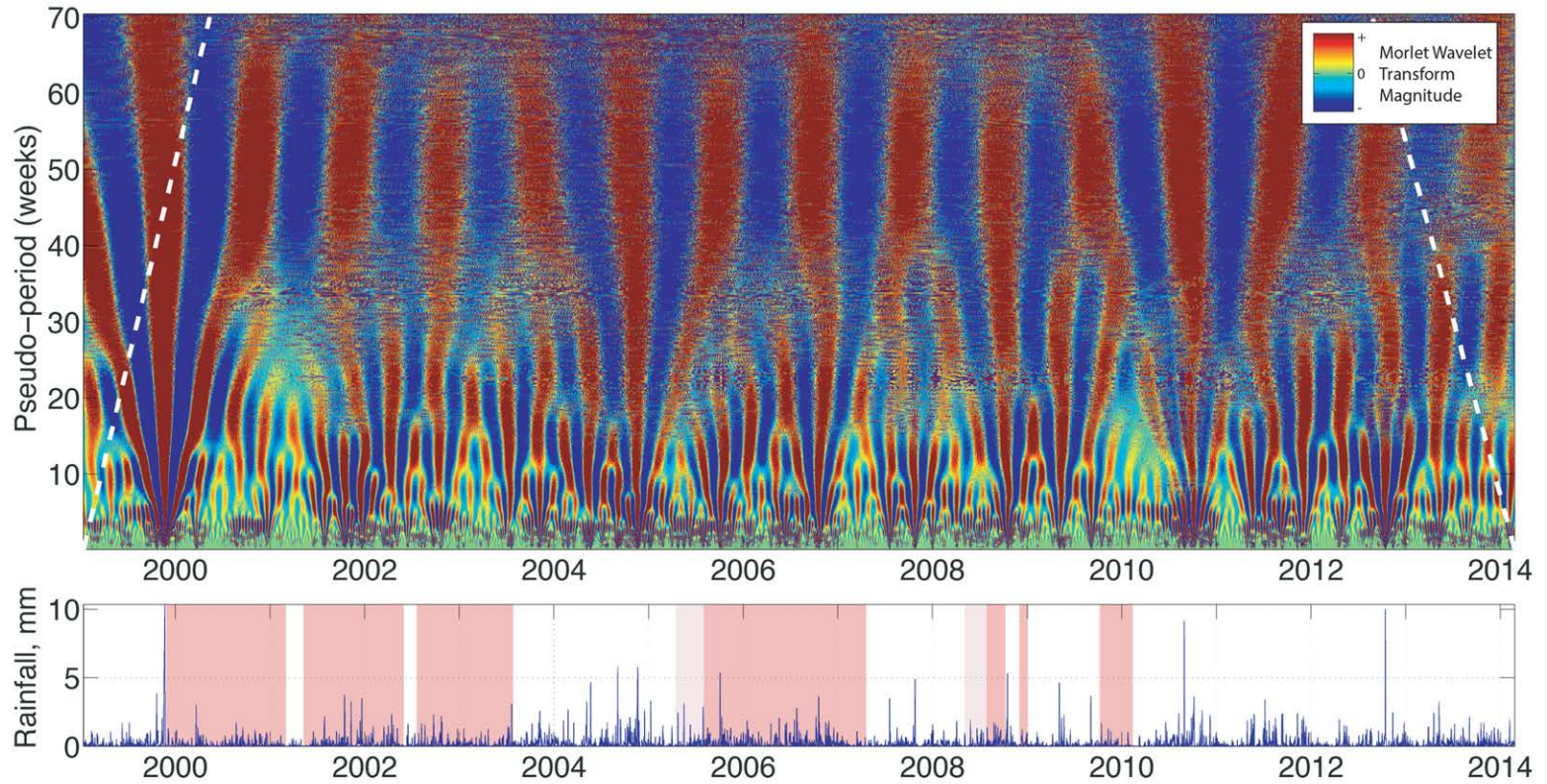


Fig. 6. Continuous wavelet analysis of the daily rainfall from 2000 to 2013 measured at Hope. Missing data were filled using a linear interpolation scheme. Ornament as in Figure 3.

SO₂ from magmatic fluids. Both datasets have a strong annual signal as expected, the relative humidity wavelet plot (Fig. 5) displays a periodicity at around 25–30 weeks (*c.* 6 months) and a much weaker signal at around 17–20 weeks (4–5 months); however, the strength of the periodicity varies throughout the time series. The daily rainfall wavelet plot (Fig. 6) shows a possible weak periodicity at 10–15 weeks. The relative strength and longevity of the 4–5 month cycle in the SO₂ data suggest that it is not controlled by relative humidity, whose 4–5 month cycle is weak and variable in strength. If the humidity and/or rainfall were the dominant control on SO₂ flux we would expect to see evidence for 6 month cyclicity in the SO₂ time series, which is not observed. We therefore reject the possibility of the 4–5 month SO₂ cycle being controlled by meteorological or groundwater conditions and therefore suggest that the 4–5 month cycle is being generated by the volcanic system.

Modulation of degassing by the lava dome and lava flux rate

Dome collapses and explosions at the Soufrière Hills Volcano are frequently accompanied by releases of large quantities of SO₂ (e.g. Herd *et al.* 2005; Carn & Prata 2010; Komorowski *et al.* 2010). Lava domes trap volatiles in their porosity structure (Taisne & Jaupart 2008). The mass of the dome generates a downward pressure that can close fractures around the volcanic conduit, inhibiting volatile leakage. The effectiveness of this trapping is dependent on the dome height and area of the base (Taisne & Jaupart 2008), thus a wider and higher dome would be more efficient at trapping SO₂.

There are no detailed data regarding the width of the base of the lava dome throughout the eruption; however the 1 km diameter crater puts constraints on the dome base area and height. There have been a number of occasions when the base of the dome has filled the entire crater, for example during extrusion phase II (late May 2002 till July 2003); also, in phase III, from February 2007 to April 2007, the dome had a volume of *c.* $200 \times 10^6 \text{ m}^3$, which is the largest to date (Ryan *et al.* 2010; Wadge *et al.* 2010). The large dome persisted through the next pause period and through extrusive phases IV and V, with a net volume of $38 \times 10^6 \text{ m}^3$ of lava being added to the dome during each of these extrusion episodes (Stinton *et al.* 2014).

The dome height was >1000 m above sea-level on 4 April 2007 (Ryan *et al.* 2010; Fig. 7a) and has been consistently greater than this since, particularly during phase V (Stinton *et al.* 2014). The low SO₂ flux during late 2006 and early 2007 appears consistent with a dome-modulated SO₂ signal. However the largest degassing cycle occurred from

June 2007 until January 2010, when one of the largest domes of the eruption was present in the crater (Wadge *et al.* 2010; Fig. 7a). In mid-2003 the SO₂ flux was increasing contemporaneously with an increasing dome volume and height (Fig. 7a). Thus the size of the lava dome has not exerted a first-order control on the SO₂ flux cycles we observe.

The highest extrusion rates during the eruption occurred in early 2006 leading up to the 20 May 2006 collapse (Wadge *et al.* 2010) and also during phase V (Stinton *et al.* 2014). Both of these periods coincide with a waning SO₂ flux (Fig. 7b), contrary to what is expected if the magma extrusion rate controls SO₂ degassing. Conversely the general trend of increasing SO₂ fluxes observed between phases III and IV occurred during a period of no lava extrusion. Thus, as in the case of dome volume, there is no correlation between the SO₂ flux and lava extrusion rate (Fig. 7b). We therefore propose that the 1.5–2.5 year and the 4–5 month cycles in the SO₂ signal are modulated neither by the lava dome nor by the lava extrusion rate, but by deeper process within the volcanic plumbing system.

Deformation-induced pressure changes causing degassing

Models of ground deformation (e.g. Elsworth *et al.* 2008; Foroozan *et al.* 2010; Paulatto *et al.* 2010; Foroozan *et al.* 2011) suggest that the Soufrière Hills plumbing system comprises two magma reservoirs linked by a dyke, one reservoir at *c.* 6 km and the other at *c.* 13 km depth (Fig. 8). Other geophysical studies (e.g. Foroozan *et al.* 2010, Melnik & Costa 2014) have shown that the GPS deformation signal that correlates with extrusion (Fig. 1) is influenced by the deformation of both reservoirs. In this section, we explore the feasibility of deformation-induced localized pressure changes that might promote degassing.

In their model, Melnik & Costa (2014) assumed a steady magma influx into the lower reservoir with an elastic elliptical dyke connecting both reservoirs. The degree of connectivity between the two reservoirs is heavily dependent on the dyke width and determines how much influence the deformation of either reservoir has on the deformation measured at the surface by the GPS network at any time during the deformation cycle in Figure 1. Pressure in the lower reservoir is a function of the magma influx rate into the lower reservoir and the degree of connectivity between the two reservoirs, which modulates the ease with which the magma can ascend into the upper reservoir. For the case of strong connectivity between the two, the individual reservoir pressures vary synchronously. However for weak connectivity, the shallow magma chamber pressure may show large periodic fluctuations,

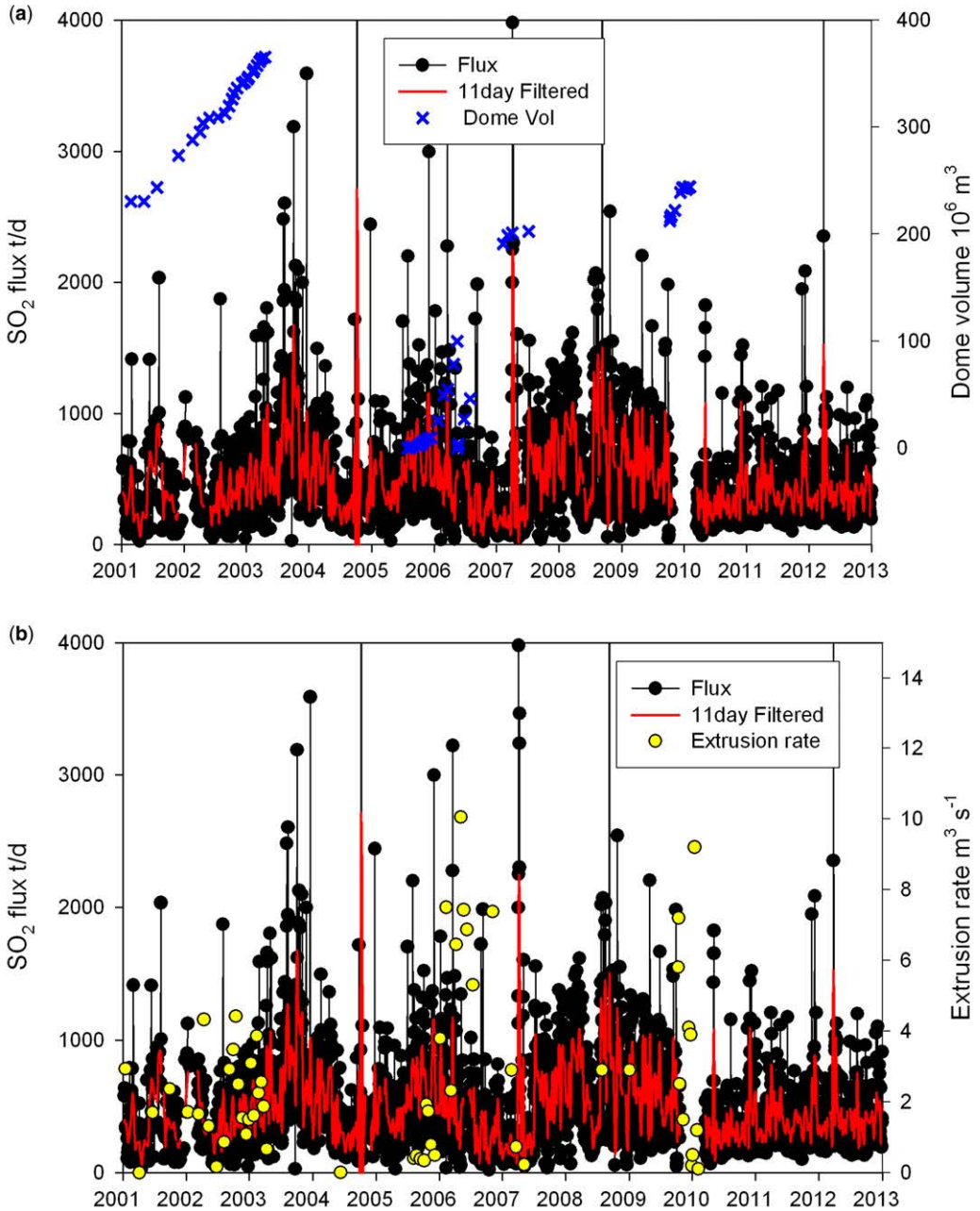


Fig. 7. Plots to show SO₂ flux with time, from 2001 to 2013. Also shown (in red) are the data after the application of a 11-day filter. Co-plotted with the SO₂ flux are (a) dome volume with time and (b) andesite extrusion rate with time.

whereas the pressure in the lower reservoir may increase steadily. The continuous increase in pressure will act to open the base of the dyke, forcing magma into it and causing it to swell after the stress and strain thresholds are overcome.

Melnik & Costa (2014) demonstrated that the dual-chamber-conduit model for Soufrière Hills Volcano is highly nonlinear and can exhibit fluctuations with a double period behaviour, with periods for small-amplitude oscillations of the order of *c.* 5

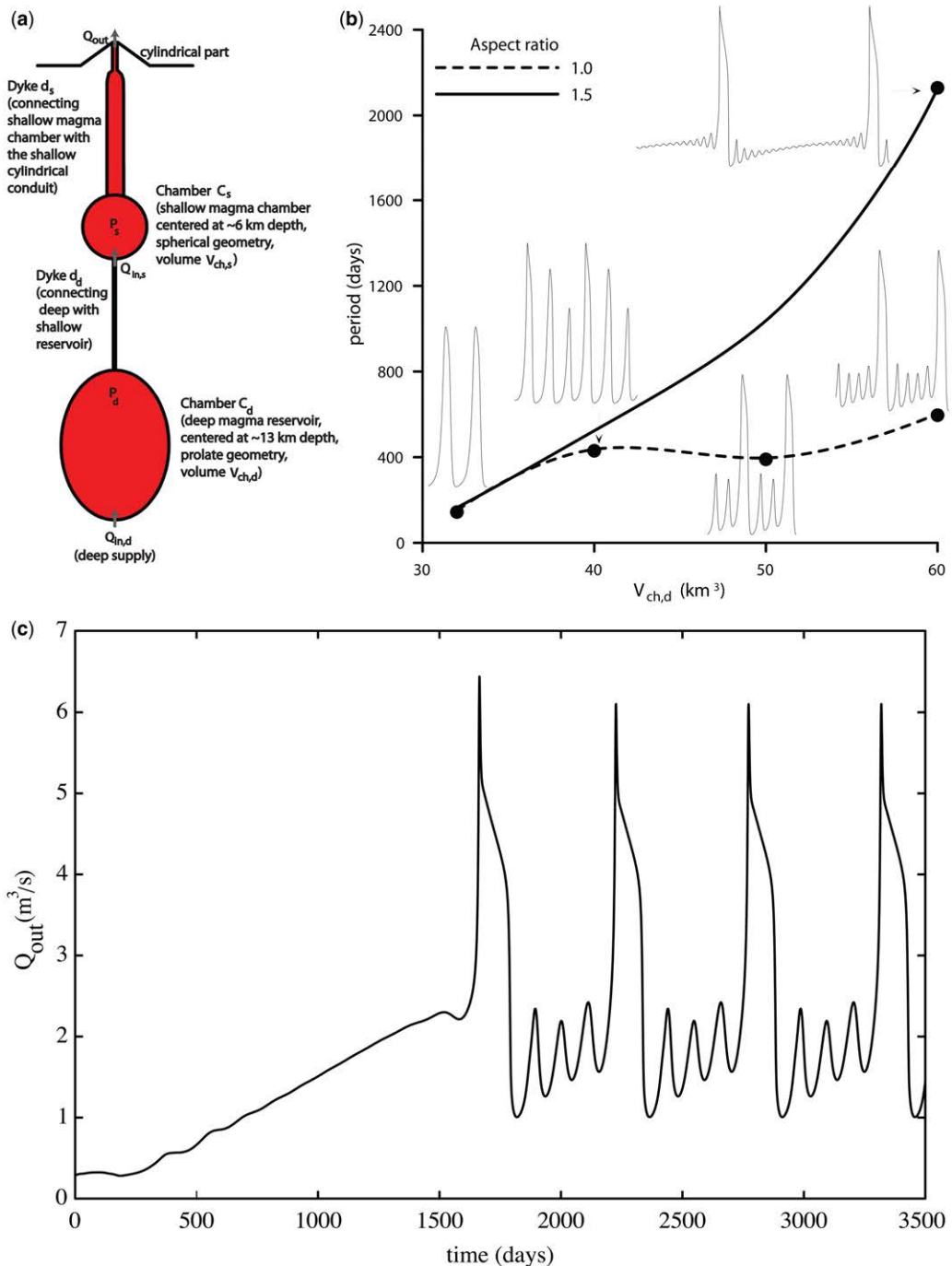


Fig. 8. Dual magma reservoirs and the origin of double periodic behaviour at Soufrière Hills Volcano, Montserrat: (a) schematic diagram to show the Soufrière Hills plumbing system constrained by measurements of ground deformation (after Melnik & Costa 2014); and (b) relationship between the deep reservoir volume ($V_{ch,d}$) and the periodicity of magma delivery to the surface (y-axis). As the volume of the deep reservoir is increased, the model predicts an increasingly divergent dual periodicity. (c) A plot of magma flux, Q_{out} , with time, illustrating the case for which the dual periods are 2 years and 4.5 months.

months, whereas the large peak of increase in discharge rate occurs every *c.* 2 years (Fig. 8). Such double periodic behaviour, characterized by two distinct periods of pulsations, was initially described in Costa *et al.* (2007a). Melnik & Costa (2014) showed that the duration of the low-frequency fluctuations and the presence of the higher-frequency fluctuations are strongly controlled by the geometry of the system such as shallow dyke extent and magma chamber size and depth (Fig. 8). Alternations between phases of high and low magma discharge rate from the model of Melnik & Costa (2014) are not in agreement with observations of magma fluxes for the Soufrière Hills Volcano. However, there are reasonable combinations of the dual-chamber–conduit system parameters that give the proper periodicity observed instead for SO₂ degassing (Fig. 8b).

Considering the sensitivity of the system to small changes in the geometry and the fact that, for a magma rheology of Bingham-type, discharge rate between the two major pulses is zero until a critical chamber overpressure is reached (Melnik & Sparks 2005), we can hypothesize that, by accounting for a more realistic description of the system geometry and the effective viscosity of magma, the system can still show a double periodicity behaviour with the higher-amplitude fluctuations characterizing the magma extrusion, whereas the lower-amplitude oscillations survive only for the SO₂ degassing. In particular, it may be envisaged that the magma's high yield strength might inhibit magma outflow in the shallowest part of the system (the dyke-conduit part), but magma exchange between the two reservoirs may still exist and gas may rise independently throughout the system. Such a mechanism could explain the modulation of degassing whereby, during periods of magma extrusion, SO₂ is released together with the magma (yielding the *c.* 2 year period), albeit with some delay and/or offset caused by gases rising quasi-independently of magma. In contrast, when magma cannot flow because of its yield strength, SO₂ can be released independently from lava (yielding the *c.* 5 month period).

The rate of SO₂ degassing will be modulated by the pressure variations within the lower reservoir and rate of magma flux into the dyke. Christopher *et al.* (2014) used geochemical evidence to suggest a pulsed supply of basalt from the lower reservoir to the shallower parts of the plumbing system. Future development of numerical models may allow the testing of this hypothesis of long-term SO₂ modulation through the geometry and dynamics of the deep magmatic system at Soufrière Hills Volcano (e.g. Melnik & Costa 2014), accounting for more realistic descriptions of the geometry of the system (which is currently not well constrained) and

magma properties, such as the increase in the yield strength owing to crystal content variations, magma permeability variations, lateral gas escape and the coupling with energy loss, viscous dissipation and 2D and stick-slip effects (e.g. Costa & Macedonio 2005; Costa *et al.* 2007b, 2012).

Prolonged SO₂ flux after phase V

The sustained flux of SO₂ from Soufrière Hills Volcano has extended for more than four years beyond the last magma extrusion episode. These observations have implications for the magma supply into the deep reservoir thought to be driving the eruption and hence for eruption forecasting and hazard assessment. Since February 2010, SO₂ fluxes have remained high but are in general below the eruption-long average rate of around 500 t/day. The wavelet analysis shows that this part of the time series does not show quite such strong periodicity as earlier in the eruption, but more time and data are required to confirm this. The maintenance of high SO₂ fluxes alone might not be an accurate indicator of sustained magma supply at depth; high degassing rates might after all be sourced from merely a cooling magma. It seems reasonable instead, from the analysis we have presented here, that a criterion for recognizing the cessation of deep magma supply and perhaps the 'end of the eruption' might be that the SO₂ time series no longer shows periodicities of the type documented here that might be related directly to deep magma supply into a dual reservoir system.

Conclusions

From a detailed time series of volcanic SO₂ fluxes derived from two fixed scanning UV spectrometers at Soufrière Hills Volcano, Montserrat, we have shown that:

- High SO₂ fluxes have been sustained through eruptive pauses and over four years after the last extrusion episode and that the emission rate of volcanic SO₂ is clearly strongly decoupled from lava extrusion rates.
- Lava extrusion rates have exhibited strong 1–2 year cyclicity, resulting in five eruptive phases. Wavelet analysis demonstrates that there are periodicities on the order of 4–5 months and 1.5–2.3 years within the SO₂ time series. The latter period is similar to the wavelength of the cyclicity observed in lava extrusion but is non-systematically offset.
- We demonstrate that the two periods observed in the SO₂ flux time series are not driven by external forcing (e.g. SO₂ scrubbing, modulation of degassing by the lava dome).

- The observed periodicities (*c.* 2 years and *c.* 5 months) are consistent with pressure changes accompanying deformation in a coupled magma reservoir system whereby double periodic behaviour may arise under circumstances of limited connectivity between the two reservoirs. Such a mechanism could explain the modulation of degassing whereby, during periods of magma extrusion, SO₂ is released together with the magma (yielding the *c.* 2 year period), albeit with some delay and/or offset caused by gases rising quasi-independently of magma. In contrast, when magma cannot flow because of its yield strength, SO₂ can be released independently from lava (yielding the *c.* 5 month period).

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