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## Sensitivity of OMI SO<sub>2</sub> measurements to variable eruptive behaviour at Soufrière Hills Volcano, Montserrat

C. S. Hayer<sup>1†</sup>, G. Wadge<sup>1</sup>, M. Edmonds<sup>2</sup> and T. Christopher<sup>3,4</sup>

<sup>1</sup> ESSC, Dept. of Meteorology, University of Reading, Reading, RG6 6AL, UK
 <sup>2</sup> COMET+, Department of Earth Sciences, University of Cambridge, Cambridge, CB2 3EQ, UK
 <sup>3</sup> Montserrat Volcano Observatory, Montserrat, West Indies
 <sup>4</sup> Seismic Research Centre, University of West Indies, St. Augustine, Trinidad

Corresponding author: C. Hayer, <u>catherine.hayer@physics.ox.ac.uk</u>

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#### Abstract

Since 2004, the satellite-borne Ozone Mapping Instrument (OMI) has observed sulphur dioxide (SO<sub>2</sub>) plumes during both quiescence and effusive eruptive activity at Soufrière Hills Volcano, Montserrat. On average, OMI detected a SO<sub>2</sub> plume 4-6 times more frequently during effusive periods than during quiescence in the 2008-2010 period. The increased ability of OMI to detect SO<sub>2</sub> during eruptive periods is mainly due to an increase in plume altitude rather than a higher SO<sub>2</sub> emission rate. Three styles of eruptive activity cause thermal lofting of gases (Vulcanian explosions; pyroclastic flows; a hot lava dome) and the resultant plume altitudes are estimated from observations and models. Most lofting plumes from Soufrière Hills are derived from hot domes and pyroclastic flows. Although Vulcanian explosions produced the largest plumes, some produced only negligible SO<sub>2</sub> signals detected by OMI. OMI is most valuable for monitoring purposes at this volcano during periods of lava dome growth and during explosive activity.

#### 1. Introduction

Sulphur dioxide (SO<sub>2</sub>) has a strong absorption signature in the UV and a low background abundance in the atmosphere, both of which facilitate its measurement in volcanic plumes and make SO<sub>2</sub>

<sup>&</sup>lt;sup>†</sup> Now at AOPP, Dept. of Physics, University of Oxford.

emission rate a valuable volcano monitoring tool (e.g. Oppenheimer et al., 2011; Shinohara, 2008; Edmonds et al., 2003a). Satellite observations of volcanic SO<sub>2</sub> emissions, although of lower resolution, complement ground-based spectrometer systems, which may not be effective during ash-producing eruptive events (Christopher et al., 2010) or for measuring vertically-rising plumes.

There have been many observations of volcanic SO<sub>2</sub> plumes from space-borne instruments. However, there have been few opportunities to compare these observations with ground-based instruments, and even fewer that allow for the assessment of their complementarity. Soufrière Hills volcano has a well-established ground-based instrument array along with other monitoring data (seismometers, GPS, tiltmeters, infrasound monitors). This provides an ideal test case to investigate the ground- and satellite-based measurement of SO<sub>2</sub> emission at a volcano with a highly variable, but well constrained, activity level.

In this paper, we conduct an analysis of OMI observations during different types of volcanic activity in order to assess the utility of OMI for volcano monitoring. We estimate plume heights for the different styles of activity observed and assess whether this can explain the differences in the ability of OMI to detect SO<sub>2</sub> at this volcano. We provide evidence from the 2008 – 2010 degassing record that shows that OMI detects SO<sub>2</sub> more frequently during eruptive activity than during periods of quiescence and we explore the mechanisms that might cause this.

#### 2. Soufrière Hills Volcano and observations of SO<sub>2</sub> emissions

Soufrière Hills Volcano, Monserrat is a dome-forming andesitic volcano located within the Lesser Antilles island arc in the Caribbean (16.7° N, 62.2° W). The current eruption began in 1995, undergoing periods of quiescence with passive degassing interspersed with 5 phases of active lava extrusion. During the periods of active extrusion, the volcano displayed cycles of dome-forming and large explosive collapses. The last period of active extrusion ended in a large dome collapse on 11 February 2010. Since that time, the volcano has continued to passively degas.

The SO<sub>2</sub> emissions from Soufrière Hills are monitored using a ground-based array of Ocean Optics UV spectrometers, with the data processed using Differential Optical Absorption Spectrometry (DOAS) (Edmonds et al., 2003a). The long-term SO<sub>2</sub> flux has been approximately constant (over months-years timescales) since the start of the eruption, during periods of both quiescence and during lava dome growth, with an average flux of 574 t/d (Christopher et al., 2010) to the end of Phase 5. Since the end of Phase 5, the average from been 375 t/d. However, the DOAS record does display a second-order variability with a multi-year periodicity (Christopher et al., 2014).

When the plume is located higher in the atmosphere, there are some challenges with the DOAS retrieval. This is because of the geometry of the retrieval and the longer pathlength from the instrument. Due to the viewing geometry of the ground-based spectrometer systems, it is impossible to determine if a plume is small with a lower altitude or is larger but higher in the atmosphere when using a single instrument (Edmonds et al., 2003a). When the plume is higher in the atmosphere, the photons travel a longer pathlength. This can lead to a light dilution effect, as photons are scattered away from the instrument (Kern et al., 2010) which in turn can lead to an underestimate of the SO<sub>2</sub> loading. The presence of ash in the plume, a common occurrence at a dome-forming volcano like Soufrière Hills, can also cause errors in retrievals (Edmonds et al., 2003a). The optical thickness of the ash reduces or completely blocks sunlight from penetrating the plume, artificially reducing the retrieved SO<sub>2</sub> mass or rendering the retrieval impossible. If a plume is emplaced at a high altitude, it may be influenced by a different weather pattern to that at lower altitudes. Since the locations of the DOAS instruments are fixed, if the plume does not overpass them, a retrieval is impossible. All of these effects are more important for high altitude plumes, and so are likely to be associated with explosive events.

Satellite instruments have been observing volcanic SO<sub>2</sub> emissions since the 1982 eruption of El Chichón when the plume was observed by the Total Ozone Mapping Spectrometer (TOMS) (Mayberry et al., 2002). Since then, a number of different instruments have been used to monitor volcanoes and observe volcanic eruptions. These have included UV and IR instruments, such as the Ozone Monitoring Instrument (OMI), the Global Ozone Monitoring Experiment (GOME), the Atmospheric InfraRed Sounder (AIRS) and the Infrared Atmospheric Sounding Instrument (IASI) (Carn & Prata, 2010; Theys et al., 2013). The low ambient concentrations of SO<sub>2</sub> make it ideal for monitoring purposes, compared to the more abundant volcanic gases of CO<sub>2</sub> and H<sub>2</sub>O.

OMI is an instrument on board the polar orbiting NASA Aura platform, overpassing Montserrat twice daily, at 01:45 and 13:45 local time (05:45 and 17:45 UTC). Since OMI is an UV instrument, it is only able to make measurements on the day side of the orbit (13:45 LT; 17:45 UTC). The orbital geometry of the satellite means that it takes a single daily snapshot of the SO<sub>2</sub> atmospheric loading rather than making a continuous flux measurement. Data are not returned by OMI on some days, for example when the swath of the instrument does not cover Montserrat or all the pixels are below the noise threshold of 0.4 DU. OMI's Charge Coupled Device (CCD) has been affected by a degeneration known as the OMI Row Anomaly (ORA), meaning various pixels across the CCD cannot be used. The ORA is thought to have been caused by movement of Aura's protective shield into OMI's field of view, partially blocking it. The degeneration began in August 2008 and has fluctuated over time, with more pixels becoming affected, but also with the regeneration of a few affected pixels. The ORA

has been stable since July 2011 (Flower & Carn, 2015). The reduced spatial coverage caused by the ORA reduced the probability of observing the plume by 2% in Phase 4 and 12% in Phase 5.

SO<sub>2</sub> plumes have been observed by satellite instruments from Soufrière Hills since 1997. TOMS and OMI observations together have measured 0.5 Mt of SO<sub>2</sub> released largely during major explosive activity, a contribution which represents around 13% of the total documented sulphur dioxide emission during the eruption up to the end of 2011 (Carn & Prata, 2010; Edmonds et al., 2014).

To date, there have been five phases of extrusive activity at SHV since 1995 (1) (Wadge et al., 2014). Since the new generation of satellites were not in orbit for the first two phases, and some not for the third phase, only the last two phases are considered in this paper. The fourth phase (Phase 4) began on 29 July 2008 and ended on 3 January 2009. On 6 October 2009, the volcano entered its fifth phase (Phase 5) of extrusive activity, involving lava dome growth and collapse, pyroclastic flows and explosions, which concluded on 11 February 2010 with a collapse of ~50 million m³ of lava dome material and an explosion with a large tephra and gas plume (Cole et al., 2010; 2014). During most of Phase 5, the Soufrière Hills DOAS array was inoperative and the OMI observations were the most frequent and sometimes the only method of measuring SO<sub>2</sub> emission.

*Table 1: Frequency of SO<sub>2</sub> signals observed by OMI before, during and after Phases 4 and 5.* 

| Period          | Dates                   | No. of<br>days | Days with data available | Days with signals | % signals |
|-----------------|-------------------------|----------------|--------------------------|-------------------|-----------|
| Before Phase 4  | 21/02/2008 - 28/07/2008 | 159            | 153                      | 20                | 13        |
| Phase 4         | 29/07/2008 - 03/01/2009 | 159            | 147                      | 49                | 33        |
| After Phase 4   | 04/01/2009 - 11/06/2008 | 159            | 130                      | 7                 | 5         |
| Before Phase 4b | 27/06/2008 – 28/07/2008 | 32             | 31                       | 7                 | 23        |
| Phase 4b        | 03/12/2008 - 03/01/2009 | 32             | 31                       | 24                | 77        |
| After Phase 4b  | 04/01/2009 - 04/02/2009 | 32             | 28                       | 1                 | 4         |
| Before Phase 5  | 30/05/2009 - 05/10/2009 | 129            | 97                       | 8                 | 8         |
| Phase 5         | 06/10/2009 - 11/02/2010 | 129            | 110                      | 63                | 57        |
| After Phase 5   | 24/02/2010 - 02/07/2010 | 129            | 96                       | 18                | 19        |

#### 3. OMI measurements, retrievals and uncertainties

The OMI dataset used was the SO<sub>2</sub> retrieval, OMSO2 (OMI Team, 2012), obtained from the NASA Mirador data store (<a href="http://mirador.gsfc.nasa.gov/">http://mirador.gsfc.nasa.gov/</a>). The accuracy of the SO<sub>2</sub> retrieval is highly dependent upon the correct assumption of the Centre of Mass Altitude (CMA) of the SO<sub>2</sub> plume (Yang et al., 2007), but there are a number of other factors which also affect the retrieval, discussed below.

#### 3.1 Radiative Cloud Fraction (RCF)

The fraction of the sky covered with meteorological cloud (reported as a fraction of 1) (RCF) is the biggest cause of error in the SO<sub>2</sub> retrieval, after those introduced by an incorrect CMA (section 3.4). Thin, high altitude cirrus clouds are not expected to cause significant errors in the retrieval; however, thicker cloud can cause error (Yang et al., 2007). If the SO<sub>2</sub> plume is located below an optically thick layer of meteorological cloud, the cloud masks the SO<sub>2</sub> and this leads to an underestimate of the true SO<sub>2</sub> loading. If the SO<sub>2</sub> plume is located above a thick layer of cloud, this can artificially enhance the loading as more UV radiation is reflected off the cloud than would be off the ground (McCormick et al., 2013). Therefore the smaller the average RCF, the more reliable the retrieval. The plumes which are observed by OMI (SO<sub>2</sub> plume altitudes determined from Washington VAAC Volcanic Ash Advisory reports) are generally higher than the surrounding meteorological cloud (meteorological cloud altitudes from metadata within the OMSO2 data set, generated using methods described by Joiner and Vasilkov (2006) and Joiner et al. (2010)); this was found to be true for 90% of days with a plume during Phase 4 and 84% of days during Phase 5 (Fig. 1 and Fig. 2). For these dates, the average RCF over the area covered by the plume is usually less than 0.25 (81% of dates for Phase 4 and 84%) for Phase 5). The averaged RCF over a 2° area centred on Montserrat is also found to be less than 0.25 for 77% of days during Phase 4 and 93% of days during Phase 5. Analysis of the effect of the RCF on the CMA TRL and TRM (section 3.4) retrievals indicates that when the RCF is below 0.2, the errors are dominated by noise from other sources (Carn et al., 2013). The errors are expected to increase linearly for RCFs greater than 0.2. The effect of cloud was also observed to be more pronounced on the TRL retrieval than on the TRM.

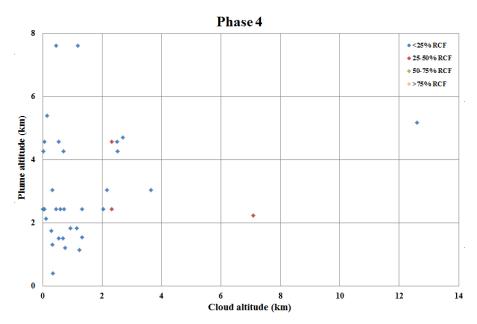


Fig. 1: Comparison of the altitude of the meteorological cloud (OMSO2 metadata) and the  $SO_2$  plume (Washington VAAC VAA archive) during Phase 4. The meteorological cloud altitude is an average over the region covered by the  $SO_2$  plume. The colour of the diamonds indicates the percentage radiative cloud fraction, as denoted in the legend.

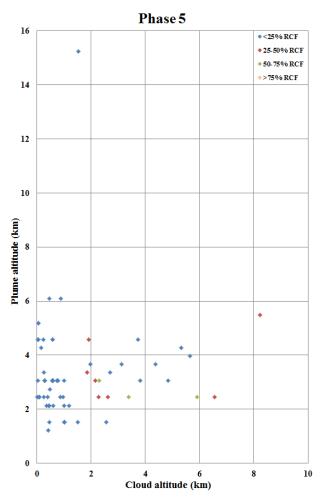


Fig. 2: Comparison of the altitude of the meteorological cloud (OMSO2 metadata) and the  $SO_2$  plume (Washington VAAC VAA archive) altitude during Phase 5. The colour of the diamonds indicates the percentage radiative cloud fractions, as denoted in the legend.

#### 3.2 Sensor Viewing Angle

The sensor viewing angle (or zenith angle) varies across an instrument's swath. For OMI, the viewing angle reaches  $\pm 70^{\circ}$  about the central viewing point (nadir). This change in the angle is caused by the curvature of the Earth and results in the pixels at the edge of the swath being significantly wider than those at the centre of the swath. At nadir, OMI's pixels are  $\sim 24$  km wide; the outer pixels are  $\sim 155$  km wide (Fig. 3). If a small plume is observed at the centre of the swath, it is likely to be covered by a number of pixels. However, at the outer edges, the plume may be smaller than one pixel, meaning the loading will be averaged over the whole pixel. If the SO<sub>2</sub> loading of the plume is very low, this could lead to the plume not being observed as the average loading value for the pixel may not exceed the detection threshold. The more oblique viewing angle at the edge of the swath will lead to a longer pathlength for the light passing through the plume, which will act to partially counteract the impact of this effect. There has, to date, been no extensive analysis of these 'edge effects'.

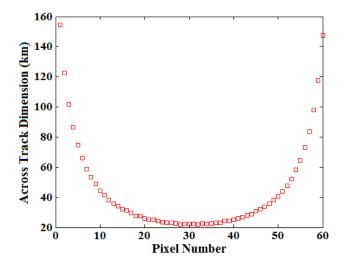


Fig. 3: Variation of the across-track width of OMI pixels with scan position (from OMI team, 2012, Figure 7). As you move away from the centre of the swath, the curvature of the Earth causes the across-track dimension of the pixel to increase from  $\sim$ 24 km at the nadir point (centre) to  $\sim$ 155 km at the swath edge.

#### 3.3 Solar Zenith Angle (SZA)

Since Aura is a sun-synchronous orbiter, the satellite equatorial overpass occurs at the same local time every day. Changes of the inclination of the Earth over the course of a year will cause only minor changes in the SZA at the Equator; however, since Montserrat is at 16 °N, there is slightly more variation in the SZA. Fig. 4a shows the variation of the SZA over Phase 4 and Fig. 4b shows the variation over Phase 5. Phase 4 occurred over a longer period and hence the variation is greater. The change in the SZA from one day to the next is small however. The seasonal variation of the SO<sub>2</sub> detection limit is primarily dependent on this change in SZA. As discussed by Carn et al. (2013, Fig.

12) and McCormick et al. (2013, Fig. 5), for mid (7.5 km) and lower (2.5 km) tropospheric plumes, the variation over the course of the year is small, of the order of 0.01 kilotons and <0.1 kilotons respectively.

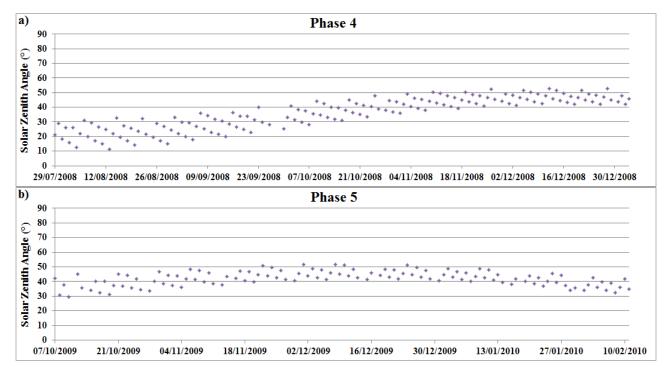


Fig. 4: Variation of the solar zenith angle over Phase 4 (a) and Phase 5 (b).

The impacts of each of these effects on the retrieval are small relative to the errors that may be introduced as a result of an incorrect assumption of the CMA (which can exceed 100%). They are therefore not considered further within this paper.

#### 3.4 Centre of Mass Altitude

The SO<sub>2</sub> mass loading retrieval was initially performed for four assumed altitudes of the centre of mass of gas within the following four atmospheric layers: Planetary Boundary Layer (PBL), 0.9 km above the ground surface; Lower Troposphere (TRL), 2.5 km; Middle Troposphere (TRM), 7.5 km; and Lower Stratosphere (STL), 17.5km. The mass loading was then interpolated linearly (Yang et al., 2007) between two of these CMA classes to the estimated altitude for the gas plume, usually obtained either from a Volcanic Ash Advisory (VAA) by the Washington Volcanic Ash Advisory Center (W-VAAC, http://www.ssd.noaa.gov/VAAC/), estimated as the top of the ash cloud from satellite imagery, radiosonde soundings, pilot reports, or from observations made by the Montserrat Volcano Observatory (MVO) (Fig. 5). The VAA reports include the ash cloud top height and the centre of mass altitude of the SO<sub>2</sub> plume, which is required for the retrieval. However, ash plumes are regularly found slightly lower in the atmosphere than the corresponding SO<sub>2</sub> plume, thus reducing the mismatch (Mayberry et al., 2002). As the plume ages and sinks within the atmosphere, it becomes more likely

that using the ash cloud top height will lead to an underestimate of the altitude of the CMA, which in turn will lead to an overestimate of the SO<sub>2</sub> loading in the plume (Yang et al., 2007).

During the retrieval, the altitude used for the CMA was derived from the VAA reports where possible. For dates when these were not issued, a DOAS-derived altitude was used, if available, or the plume was assumed to be at the level of the TRL class (2.5 km).

Each CMA is associated with an a priori assumption on the shape and vertical distribution of SO<sub>2</sub> within the atmosphere. The use of an incorrect SO<sub>2</sub> profile, and hence CMA, was shown by Carn et al. (2013) to cause a significant overestimate of the atmospheric SO<sub>2</sub> loading if the plume was located above the assumed altitude and an underestimate if it was located below the assumed height. If the 2.5 km CMA is used and the plume is actually located higher than 2.5 km, the retrieval can produce an overestimate of the true atmospheric loading of SO<sub>2</sub>, up to 175%. However, the impact of the plume being located lower on this kernel is less severe. For both the TRL (7.5 km) and STL (17 km) bands, the reverse is true. The impact of the plume being located higher than the assumed altitude is less severe, while the impact of the plume being located under the assumed altitude is greater, with up to a 60% underestimate of the atmospheric loading. The impact of meteorological cloud below the plume on the retrieval is limited if the plume is located at the assumed altitude or higher. However, if the plume is actually located lower than the assumed altitude, this leads to an overestimate in the atmospheric loading down to almost the level of the meteorological cloud. Below this, the retrieved loading drops off very quickly, due to the masking of the SO<sub>2</sub> plume beneath the meteorological cloud (Yang et al., 2007).

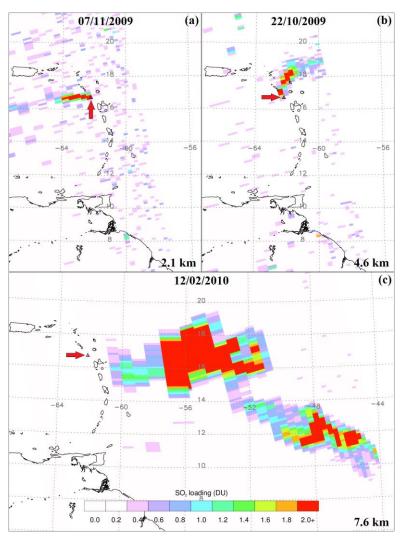


Fig. 5: Examples of SO<sub>2</sub> plumes, emitted from SHV (arrowed), as observed by OMI: (a) small (665 t); (b) medium (2147 t); (c) large (138,381 t). The altitude of each plume is shown. Note that the direction of the plume is controlled by local weather patterns, with the NE Trade Winds being dominant at lower tropospheric altitudes (a). Dates (dd/mm/yyyy) of the images and the latitude and longitude of the Eastern Caribbean are shown.

#### 4. Results

The DOAS record shows that the SO<sub>2</sub> emission rate was variable during Phase 4, ranging from <100 to >2000 t/day (Fig. 6). There is no correlation between DOAS SO<sub>2</sub> emission rate and the explosive volcanic events, though this could be impacted by the timing of the event (UV spectroscopy is not possible during the night) or due to the ash content of the plume increasing the opacity of the plume (Fig. 6). During the 10-11 April 2011 eruption of Mt. Etna, ground-based DOAS underestimated the SO<sub>2</sub> loading of ash rich plumes by up to an order of magnitude (Boichu et al., 2015). OMI detected SO<sub>2</sub> infrequently between 29 July 2008 and 2 December 2008 (on average once every 4.6 days), and then more frequently from 3 December 2008 until 3 January 2009 (every 1.3 days). During the period 3 December 2008 to 3 January 2009 (Phase 4b), the lava dome grew at a rate of ~ 13 m<sup>3</sup>s<sup>-1</sup> (Wadge et al., 2014), and enhanced rockfall activity reflected this high rate of lava extrusion (Fig. 6), a

behaviour previously seen during the eruption, as reported by Calder et al. (2005). During this period of lava extrusion, SO<sub>2</sub> emission rates measured by DOAS were mostly <1000 t/d and on average, lower than those for 29 July 2008 to 3 January 2009 (Fig. 6), during which time the lava extrusion rate was <1 m<sup>3</sup>s<sup>-1</sup> (Wadge et al., 2014).

During Phase 5, lava was extruded at an average rate of  $\sim 7 \text{ m}^3\text{s}^{-1}$  (Stinton et al., 2014), accompanied by rockfalls, larger collapses producing pyroclastic flows, and occasional Vulcanian explosions (Fig. 7; Komorowski et al., 2010). The activity level underwent strong cycles during this phase.

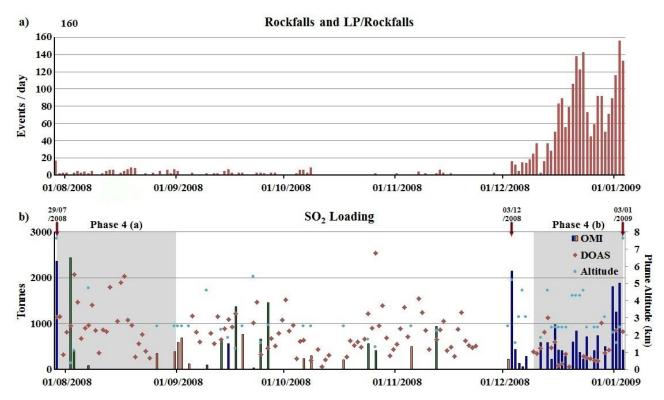


Fig. 6: Monitoring data for Phase 4: a) Rockfall seismicity; b)  $SO_2$  records from DOAS and OMI along with the plume altitude. The data source for the altitude of the plume is denoted by the colour of the bars: blue = VAA; green = DOAS; orange = assumed altitude of 2.5 km. Red arrows show the 3 main Vulcanian explosions; shaded areas denote lava extrusion.

There is a general correlation between the OMI data and the rockfall seismicity data (Fig. 7b), particularly for October 2009. Specific daily correlations can also be seen: for example, high seismicity and SO<sub>2</sub> loading, together with large pyroclastic flows, occurred on 11<sup>th</sup> December 2009 (Fig. 7), but not in all cases. A large SO<sub>2</sub> signal persisted for 12 days as a result of the large dome collapse and Vulcanian explosion at the end of Phase 5, on 11<sup>th</sup> February 2010 (Fig. 5c; Fig. 7c). Conversely, at 14.49 local time on 8<sup>th</sup> January 2010, there was a large Vulcanian explosion, producing an 8 km eruption column, which did not produce a signal detectable by OMI. This lack of signal may have been caused by two factors. First, the OMI overpass occurred 23 hours after this explosion, such that a significant fraction of the SO<sub>2</sub> could have been removed from the atmosphere through chemical

processing (Rodriguez et al., 2008), or made less detectable as the plume lost altitude. Second, the 8<sup>th</sup> January 2010 explosion is thought to have involved older, degassed dome material (Cole et al., 2010) and so perhaps there was only minimal contribution from the reservoirs of gas contained within the conduit or the magma chamber, both of which have been hypothesized as important sources of SO<sub>2</sub> in previous lava dome collapses (Edmonds et al, 2003b).

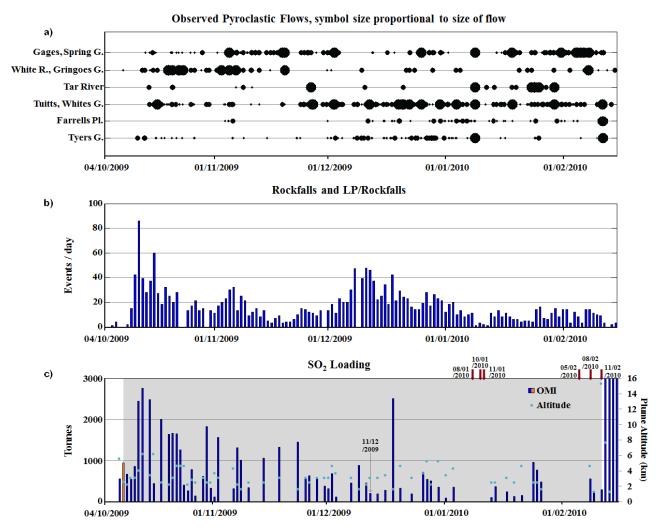


Fig. 7: Monitoring data for Phase 5: a) Pyroclastic flow location (y-axis) and size (circle diameter); b) Rockfall seismicity; c) SO<sub>2</sub> daily loading as measured by OMI and plume altitude. Coloured bars, red arrows and shaded areas as in Fig. 6 (a and b after Cole et al., 2010).

During Phase 5, OMI data were acquired over Montserrat on 110 of 129 days (Table 1), with a plume detected by OMI on 63 days (57%), but only on 13% of days from the non-extrusive periods before and after Phase 5. Over the whole of Phase 4, data were collected on 147 of 159 days, and a signal was observed on 49 days (33%). However, this includes the period of hiatus (October – November) when there was no lava extrusion. If we only consider the final part of the phase (03/12/08 – 03/01/09), then a signal was observed on 77% of days. For the quiescent periods before and after Phase 4, a signal was measured on 9% of days. For Phases 4 and 5 OMI detected 4-6 times as many

signals during the periods of lava extrusion as during non-extrusion.

#### 5. Discussion

We now evaluate the significance of the ability of OMI to detect  $SO_2$  clouds preferentially during lava extrusion. We propose two causes: (1) an increase in the emission rate of  $SO_2$  from the volcano or; (2) the increased lofting of gas to high altitudes. The first mechanism delivers more  $SO_2$  into the atmosphere; the second results in OMI being able to observe the same amount of  $SO_2$  at higher apparent loadings due to the plume being emplaced at higher altitudes. Higher altitude plumes mean a shorter path length with more backscattered photons crossing the plume, especially if the plume is located above the meteorological cloud, yielding a higher signal to noise ratio.  $SO_2$  may also be removed more slowly at higher altitudes. An  $SO_2$  removal rate of 30%/day has been proposed for Soufrière Hills, due to wet and dry deposition and conversion to sulphate (Rodriguez et al., 2008). Chemical processing in the plume is expected to be more rapid at lower altitudes due to clouds and rain (the former providing surfaces for heterogeneous chemistry and the latter removing sulphate aerosol from the atmosphere). Fig. 8 shows the distributions of observed plume altitudes. The mean altitude for Phase 4 was  $\sim 2.5$  km and  $\sim 3$  km for Phase 5.

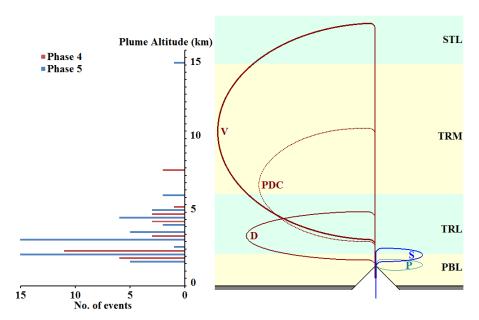


Fig. 8: Expected and observed plume altitudes at SHV. Left: Frequency of VAA plume heights for Phase 4 (red) and Phase 5 (blue). Right: OMI CMA atmospheric layers and the schematic  $SO_2$  emplacement altitude ranges (curves) expected for each of the physical mechanisms envisaged at SHV (with an assumed vent altitude of 1 km a.s.l.). P = passive degassing; S = increased deep degassing or increased conduit permeability; V = Vulcanian explosion; PDC = co-pyroclastic flow plumes; D = hot dome.

During extrusive periods, the volatile transport from depth may increase for short periods, leading to an increase in SO<sub>2</sub> emission rate and more frequent detection by OMI. There are three possible

mechanisms to achieve this: i) a greater rate of supply of sulphur from the deep plumbing system; ii) an increase in the andesite lava extrusion rate, increasing the amount of SO<sub>2</sub> advected as a vapour phase; and iii) an increase in the permeability of the conduit to SO<sub>2</sub> rise. The presence of a lava dome at the top of the conduit, as was the case for both Phases 4 and 5, will tend to increase lithostatic pressure and lower permeability within the system (Woods et al., 2002). A larger dome may also act as a high-level reservoir of SO<sub>2</sub> (Taisne & Jaupart, 2008). The higher overpressure from a larger dome may also increase the amount of gas that can be stored immediately below the dome. In Phase 5, there was a large explosion as the gas-rich core of the dome was partially exposed during the dome collapse on 11 February 2010, and which may have evacuated the magma within the conduit. The permeability of the whole system is at its highest after such events, leading to pulses of degassing (Edmonds et al., 2003a). However, the long-term DOAS-measured SO<sub>2</sub> degassing record does not show any significant difference in the rate of SO<sub>2</sub> degassing between extrusive and non-extrusive periods (Christopher et al., 2010), suggesting that variable SO<sub>2</sub> emission is not responsible for the OMI results. It is possible that the DOAS time series misses some of the higher altitude plumes, which are likely to have a higher SO<sub>2</sub> loading.

During extrusive periods, the plume attained higher mean altitudes than during non-extrusive periods. We consider three possible processes by which such lofting was achieved: (a) Vulcanian explosions; (b) buoyant plumes rising off pyroclastic flows following dome collapse ("co-pyroclastic flow clouds"); (c) the higher temperature and buoyancy of the atmosphere above the hot dome during lava extrusion.

#### 5.1. Vulcanian explosions

Vulcanian explosions inject gas and tephra plumes into the atmosphere with high initial momentum, and there are accurate observations of their altitude. Druitt et al. (2002) performed an extensive survey of the 88 Vulcanian explosions between August and October 1997. These events were associated with plume heights of 3-15 km (with a mean of  $\sim 9$  km) measured using NOAA satellite data, (heights around 20% lower were found using GOES satellite data), an average dense rock equivalent (DRE) volume of  $3.8 \times 10^5$  m<sup>3</sup> and a range of  $0.01 - 17.5 \times 10^5$  m<sup>3</sup>. The explosions observed during Phases 4 and 5 reached a similar altitude range to the 1997 sequence of explosions (5-15 km) (Komorowski et al., 2010; Cole et al., 2014) albeit with a slightly smaller range in DRE volumes ( $2-26 \times 10^5$  m<sup>3</sup>).

Table 2: Vulcanian explosions from 2008 - 2010. (<sup>1</sup>Cole et al. (2014); <sup>2</sup> Washington VAAC VAA data)

| Date       | Time (UTC) <sup>1</sup> | Hours to OMI overpass | Maximum Plume Height (km) <sup>2</sup> | Height at overpass (km) <sup>2</sup> | OMI loading (tonnes)   |
|------------|-------------------------|-----------------------|--|--------------------------------------|------------------------|
| 29/07/2008 | 03:30                   | 14                    | 12                                     | 8                                    | 2359                   |
| 03/12/2008 | 01:34                   | 16                    | 12                                     | 5                                    | 2146                   |
| 03/01/2009 | 11:04                   | 7                     | 11                                     | 8                                    | 419                    |
| 08/01/2010 | 19:49                   | 22                    | 8                                      | 2                                    | -                      |
| 10/01/2010 | 05:28                   | 12                    | 7                                      | 2                                    | -                      |
| 11/01/2010 | 00:27                   | 17                    | 6                                      | 2                                    | -                      |
| 05/02/2010 | 17:49                   | 24                    | 7                                      | 2                                    | -                      |
| 08/02/2010 | 23:57                   | 18                    | 5                                      | 1                                    | 242 (09/02/10)         |
| 11/02/2010 | 16:49                   | 1                     | 15                                     | 15                                   | 291 (45,000: 12/02/10) |

Of the nine Vulcanian explosions in Phases 4 and 5 (Table 2), only four produced a large OMI signal, one produced a small signal and the remaining four did not produce a detectable plume. The three that produced the largest OMI observed signals (29 Jul. 2008, 3 Jan. 2009 and 11 Feb. 2010) erupted the largest proportions of pumiceous tephra, interpreted as the evacuation of magma from the conduit (Cole et al., 2014). The lack of a detected plume and minor pumice production for the other explosions suggests they were driven by shallow pressurization of the dome itself (e.g. Gottsmann et al. (2011)). A large SO<sub>2</sub> signal was produced on 3 December 2008, but there was very little pumice associated with this event. This explosion had the greatest seismic energy of all nine events, and a large amplitude strain signal. Gottsmann et al. (2011) interpreted this event as the result of rapid pressurization of the whole volcanic system with a concentration of gas beneath the lava dome driving the ballistic-dominated explosion.

The factors that mitigate against effective ground-based measurement of the SO<sub>2</sub> loading produced by Vulcanian explosions are the vertical nature of the plume, the high altitude of the plume and the ash content. For OMI, it is largely the timing of the overpass. If the explosion occurs just after overpass, then much of the SO<sub>2</sub> can be lost from the plume due to chemical processing and wet and dry deposition, before the next observation opportunity. The altitude of the SO<sub>2</sub>-bearing plume may also decrease in the manner of ash-bearing plumes (Table 2), as observed in other eruptions (Dacre et al., 2011). This in turn will tend to reduce the strength of the signal.

#### 5.2. Co-pyroclastic flow plumes

The altitudes attained by buoyant plumes rising from pyroclastic flows are generally lower than those attained from Vulcanian explosions, as only thermal buoyancy forces are involved. We estimate

plausible altitudes by assuming that the plume source can be treated as an 'instantaneous' event, which requires the rise time of the plume to be less than the release time (Woods & Kienle, 1994). This is reasonable if the elutriation time of the hot tephra and gas out of the flow and into the plume is less than the plume rise time. There are very limited data on pyroclastic flow plume rise times, however the average rise time of pyroclastic flow plumes during the 1990 Redoubt eruption was 200 - 500 seconds (Woods & Kienle, 1994) and during the Mt. St. Helens 1981 eruption, 350 - 500 seconds (Woods & Wohletz, 1991) and we assume these rise times are typical. The emplacement time for individual pyroclastic flows at Soufrière Hills is typically 120 - 180 seconds, validating the assumption that the source is 'instantaneous'.

Larger collapse-derived pyroclastic flow plumes can attain heights of up to 10 km (e.g.  $25^{th}$  June 1997, Loughlin et al. (2002)) but there are few accurate observations. These flows typically have DRE volumes of  $0.1 - 10 \times 10^6 \, \text{m}^3$ ; a lava density of 2400 kg m<sup>-3</sup>; and a specific heat of 1100 J kg<sup>-1</sup> K<sup>-1</sup>. The temperature of typical flow deposits (as measured by Cole et al. (2002)) are up to  $\sim 900 \, \text{K}$ . Using an elutriation temperature of  $\sim 600 \, \text{K}$ , as suggested by Woods & Kienle (1994), produces a temperature change of  $\sim 300 \, \text{K}$ . The proportion of solids elutriated are in the range 4 - 16% (Bonadonna et al., 2002), and we assume that 10% of the total mass releases its heat content to the plume. The maximum plume height, H (m), of an instantaneous source with Q (Joules) available thermal energy, calculated from the above values, is given by:

$$H = 1.87 \times Q^{1/4}$$

and yields maximum plume heights of 3.2, 5.7 and 10.1 km for flow volumes of 0.1, 1.0 and  $10 \times 10^6$  m<sup>3</sup> respectively (Morton et al., 1956).

#### 5.3. Hot dome plumes

During periods of enhanced lava extrusion rate, hot lava extruded at the surface of the dome leads to an increase in the average surface temperature of the dome. Gas leaving the dome entrains the surrounding air that has been heated by the dome surface, making it more buoyant and increasing the altitude at which the gas-rich plume attains its level of neutral buoyancy.

The average volumetric lava flux in Phase 5 was about 6  $\text{m}^3\text{s}^{-1}$ . Wooster et al (1997) calculated that the thermal power emitted by the 1991 – 1993 lava flow on Mount Etna was 1 – 10 GW from a similar lava flux. The proportion of the total power emitted via convection ranged from 30 to 50%. The mechanism of convective heat transfer from a defined source area is also employed to study forest fires. Harrison & Hardy (2002) found an empirical relationship between the maximum observed

plume height, H (m) and the maximum power of the fire, P (GW):

$$H \approx 1403 \times P^{0.36}$$

For a fire with P in the range 1-10 GW, which is the range of convective power emission expected for Soufrière Hills,  $H \sim 1.4-3.2$  km. This estimate does not include the effect of wind on the plume height, which generally results in a lower H for a given size of fire. Freitas et al (2010) used a numerical model (3-D Active Tracer High resolution Atmospheric Model (ATHAM)) to investigate the effect of wind on the plume from a 0.1 km² fire. This is a similar surface area to that covered by the central core of the lava dome during Phase 5, which is estimated as 0.07 km² (the area covered by the core and talus combined is estimated as 0.8 km²). The heat flux from this size of fire was estimated at 80 kWm², which produces a total energy emission of 8 GW – the same order of magnitude as that expected from SHV. The maximum plume height for the 0.1 km² fire was 3.3 km in calm conditions and 1.7 km for the windy case, when the plume was bent over. We would therefore expect a plume within the range 1.5-4 km during active lava extrusion.

The estimated ranges in altitude of the  $SO_2$  plumes for the separate mechanisms described are shown in Fig. 8: Vulcanian explosions (3 – 15 km), co-pyroclastic flows (3 – 10 km), hot lava domes (1.5 – 4 km), enhanced supply of  $SO_2$  from depth (1.5 – 2 km). Most of the OMI signals recorded during Phases 4 and 5 were in the PBL and the lower troposphere (1.5 – 6 km) (Fig. 8). Those signals in the PBL are likely to have been formed by an enhanced rate of  $SO_2$  degassing or by lofting above a hot lava dome. The plumes in the lower troposphere are likely to have been formed by lofting due to copyroclastic flows or above hot domes.

Some of the VAA values of Fig. 8 are probably underestimates. For tephra-rich plumes, two distinct components tend to separate within 24 hours of emplacement, with a lighter SO<sub>2</sub>-rich plume at a greater altitude than that of the tephra plume (Mayberry et al., 2002).

#### 6 Conclusions

In conclusion, our study of the 2008 - 2010 SO<sub>2</sub> release from Soufrière Hills shows that:

- i) During "passive" degassing, when no lava is being extruded, SO<sub>2</sub> loads in the atmosphere are up to six times less likely to be measured by OMI than during periods of lava extrusion.
- ii) This variability in OMI detection capability is probably usually caused, not by real changes in the gas flux from the volcano, but by lofting of the gas plume during vigorous dome growth to 1.5 to 6 km above the volcano.

- iii) Lofting of  $SO_2$  is produced in three main ways: by Vulcanian explosions (3 15 km), by copyroclastic flow plumes (3 10 km) and by buoyant rise above the hot surface of the lava dome (1.5 4 km).
- iv) Some Vulcanian explosions, involving magma from the conduit, show strong SO<sub>2</sub> signals, others just involving explosions within the dome, do not. This corroborates the mechanisms proposed by Komorowski et al. (2010), Gottsmann et al. (2011) and Cole et al. (2010).
- v) OMI measurements will detect SO<sub>2</sub> loading events, due to Vulcanian explosions for example, that will be missed or underestimated by ground-based measurement systems, due to the increased path length caused by the altitude and the ash loading of the plume (Boichu et al, 2015).

OMI, and other satellite-based instruments, are biased towards measurements of emissions from eruptive volcanoes and so emissions into the troposphere by persistently degassing volcanoes, which make up the majority of SO<sub>2</sub> emissions, may be missed. Ground-based instruments are more likely to capture these emissions, but will miss plumes emplaced higher in the atmosphere. Since neither satellite- nor ground-based instruments favour retrieval of plumes emplaced in the middle atmosphere, it is expected that these plumes will be under-reported by an observation system involving both instrument types.

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