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# Extended observations of volcanic $SO_2$ and sulfate aerosol in the stratosphere

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## Abstract

Sulfate aerosol produced after injection of sulfur dioxide ($SO_2$) into the stratosphere by volcanic eruptions can trigger climate change. We present new satellite data from the Ozone Monitoring Instrument (OMI) and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) missions that reveal the composition, structure and longevity of a stratospheric $SO_2$ cloud and derived sulfate layer following a modest eruption (0.2 Tg total $SO_2$) of Soufriere Hills volcano, Montserrat on 20 May 2006. The $SO_2$ cloud alone was tracked for over 3 weeks and a distance of over 20 000 km; unprecedented for an eruption of this size. Derived sulfate aerosol at an altitude of ~20 km had circled the globe by 22 June and remained visible in CALIPSO data until at least 6 July. These synergistic NASA A-Train observations permit a new appreciation of the potential effects of frequent, small-to-moderate volcanic eruptions on stratospheric composition and climate.

## 1 Introduction

Over the past three decades, satellite remote sensing of volcanic clouds following major explosive eruptions has delivered new insights into the magnitude of volcanic sulfur dioxide ($SO_2$) emissions (e.g., Carn et al., 2003), and thus into their effects on the atmosphere and climate (Robock, 2000). However, significant uncertainty surrounds the $SO_2$ yield and, consequently, the atmospheric impacts of more frequent explosive eruptions of intermediate magnitude, defined here as those events with a Volcanic Explosivity Index (VEI) (Newhall and Self, 1982) of 2 to 4. Volcanic plumes generated by eruptions of this size have the potential to reach altitudes of up to 25 km (Newhall and Self, 1982), well within the stratosphere at all latitudes, and may occur several times a year, compared to roughly once per decade for events of VEI 5 or above (Simkin and Siebert, 1994). A more quantitative understanding of $SO_2$ release by this common class of volcanic activity would therefore be of significant benefit to numerous endeav-



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ors, including modeling of atmospheric chemistry and climate, and petrological studies of volcanic degassing and volatile recycling (e.g., Wallace et al., 2003).

Sulfur emissions by intermediate-scale eruptions have been inadequately sampled by remote sensing due to the relatively low sensitivity of satellite sensors such as the Total Ozone Mapping Spectrometer (TOMS), used to measure $SO_2$ in volcanic eruption clouds since 1978 (Carn et al., 2003; http://toms.umbc.edu). The six ultraviolet (UV) TOMS wavelengths were not optimal for $SO_2$ retrievals, although the situation was ameliorated with the most recently deployed iteration of the instrument, Earth Probe (EP) TOMS (Gurevich and Krueger, 1997). However, the low $SO_2$ sensitivity coupled with the coarse spatial resolution of TOMS (39–50 km at nadir) meant that its ability to detect volcanic clouds of small geographic extent and/or containing low $SO_2$ amounts was compromised.

We reveal here the significant improvements in $SO_2$ sensitivity and volcanic cloud tracking ability offered by a new UV satellite sensor, the Ozone Monitoring Instrument (OMI), in orbit on NASA's Aura satellite since July 2004. OMI is one of a new generation of charge-coupled device (CCD)-based hyperspectral UV/Visible spectrometers, building on the heritage of TOMS, and more recently the Global Ozone Monitoring Experiment (GOME) (Burrows et al., 1999) and Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) (Bovensmann et al., 1999). GOME and SCIAMACHY first demonstrated the advantages of high spectral resolution and full spectral coverage for space-based mapping of $SO_2$ and other trace gases (e.g., Eisinger and Burrows, 1998; Afe et al., 2004). OMI offers several improvements over these sensors, including better spatial resolution and contiguous global coverage. The capabilities of OMI were demonstrated following a lava dome collapse at Soufriere Hills volcano (SHV), Montserrat (West Indies), on 20 May 2006, which triggered the release of a volcanic plume that entered the stratosphere.



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## 2 The Soufriere Hills lava dome collapse, 20 May 2006

A lava dome-forming eruption has been in progress at SHV (16.7° N, 62.2° W, alt. ∼1000 m) since July 1995. The eruption has featured several major dome collapses involving >10 million $m^3$ of lava, with episodes of explosive activity accompanying and/or following most of these events. Until 2006, reported maximum ash cloud heights during this explosive activity, derived from IR GOES data, had never exceeded ∼16 km (Rose and Mayberry, 2000; Ellrod and Schreiner, 2004), although this does not exclude the possibility that related $SO_2$ clouds were located at higher altitudes. A notable feature of many large dome collapses at SHV is that they have occurred during or shortly after intense rainfall, suggesting a possible trigger mechanism (Matthews et al., 2002; Carn et al., 2004).

Prior to May 2006, the previous major dome collapse at SHV (and the largest of the eruption to date) occurred on 12-13 July 2003 (Montserrat Volcano Observatory (MVO) reports available at: http://www.mvo.ms). After an extended period of quiescence beginning in late July 2003, dome growth resumed in early August 2005. High lava extrusion rates (8–10 $m^3\,s^{-1}$) were reported at SHV in April–May 2006. The 20 May 2006 dome collapse also coincided with heavy rain on the island, peaking at ∼11:00–12:00 UT with explosive venting of depressurized magma in the conduit, at which time an ash cloud was reported at ∼17 km altitude by the Washington VAAC (see http://www.ssd.noaa.gov/VAAC/archive.html). This is the highest altitude reached by an ash cloud in the 11-year SHV eruption to date, although the collapse, which involved ∼100 million $m^3$ of lava dome, was not the largest by volume. We point out that this event was not an explosive volcanic eruption in the typical sense, driven directly by decompression and fragmentation of ascending volatile-rich magma. Rather, the most likely scenario is that rapid removal of overburden (the lava dome) from the conduit caused magma that would otherwise have ascended passively beneath the dome to explosively decompress and release a powerful jet of gas and ash that reached stratospheric altitudes. Local atmospheric conditions were very calm on the 20 May, and this



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may have favored the high altitude reached by the plume (Bursik, 2001).

## 3 Satellite data

OMI is a pushbroom sensor designed for daily, contiguous global mapping of ozone, $SO_2$ and several other trace gases with a nadir spatial resolution of 13×24 km (Levelt et al., 2006). For $SO_2$ measurements, the smaller footprint, higher spectral resolution (0.45 nm in the 306–380 nm UV2 channel), and full UV spectral coverage of OMI results in a two order of magnitude increase in sensitivity relative to TOMS, allowing detection of tropospheric volcanic plumes and small eruptions (Krotkov et al., 2006).

The $SO_2$ algorithm described by Krotkov et al. (2006) has been supplanted by an enhanced algorithm in the publicly released OMI $SO_2$ dataset (see http://so2.umbc.edu/omi for further information), to improve the accuracy of retrievals for high $SO_2$ loadings. The new algorithm uses sets of discrete OMI wavelengths (up to 10) to simultaneously retrieve ozone, $SO_2$ and the effective surface reflectivity (Yang et al., 2007[1]). Six of the bands correspond to EP-TOMS wavelengths and four are centered at extrema of the $SO_2$ absorption cross-section in the 310.8–314.4 nm wavelength range. Longer wavelengths are used for large $SO_2$ loadings to avoid underestimation of $SO_2$ due to saturation at shorter wavelengths. The retrieval derives the three geophysical parameters by adjusting them until the differences between the measured radiances and forward model calculations at these discrete bands are minimized. Our forward model employs accurate radiative transfer that accounts for multiple Rayleigh scattering, rotational Raman scattering (Ring effect), ozone and $SO_2$ absorption, and surface reflectivity. The minimization procedure requires a weighting function for $SO_2$, which is calculated based on assumed $SO_2$ profiles centered at a prescribed altitude. The default altitude for $SO_2$ clouds in the upper troposphere/lower stratosphere (UTLS) is

[1]Yang, K., Krotkov, N. A., Krueger, A. J., Carn, S. A., Bhartia, P. K., and Levelt, P. F.: Linear Fit Algorithm for $SO_2$ vertical column retrieval from the Ozone Monitoring Instrument (OMI), J. Geophys. Res., in preparation, 2007.

15 km, which we adopt for the SHV $SO_2$ cloud; the altitude dependence of $SO_2$ sensitivity is small in the UTLS. The algorithm yields adjustments to the initial estimates of ozone and reflectivity, and the $SO_2$ vertical column density (VCD).

Aura is in NASA's A-Train constellation of five satellites (which also includes the Aqua and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) spacecraft), which carry sensors that observe the same air mass within a ~15 minute window. The Atmospheric Infrared Sounder (AIRS) and Moderate Resolution Infrared Spectroradiometer (MODIS) on Aqua, and the Microwave Limb Sounder (MLS) on Aura, provided IR and microwave observations of the SHV volcanic cloud; these data are discussed in a separate paper (Prata et al., 2007[2]). The CALIPSO satellite, carrying the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), was launched into the A-Train in April 2006. CALIOP provides high-resolution vertical profiles of aerosol and clouds, and allows discrimination of cloud phase and the identification of the presence of non-spherical aerosols (Winker et al., 2003).

## 4 Data analysis

OMI first detected the $SO_2$ cloud emitted from SHV at 17:00 UT on 20 May, ~6 h after emission. Under the influence of the easterly phase of the Quasi-Biennial Oscillation (QBO), the cloud then proceeded to move westwards across the Caribbean Sea and then the Pacific Ocean, at an average velocity of ~13 m/s (Fig. 1). OMI continued to track the $SO_2$ cloud until 13 June, when it became dispersed over a broad region from the Indian Ocean to Africa, at least 26 000 km from Montserrat (an animation showing OMI $SO_2$ retrievals for 20 May–11 June 2006 is available; see supplementary material http://www.atmos-chem-phys-discuss.net/7/2857/

[2]Prata, A. J., Carn, S. A., Stohl, A., and Kerkmann, J.: Long range transport and fate of a stratospheric volcanic cloud from Soufriere Hills volcano, Montserrat, Atmos. Chem. Phys. Discuss., submitted, 2007.



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2007/acpd-7-2857-2007-supplement.zip). No ash was detected in the $SO_2$ cloud using the OMI UV Aerosol Index, which is sensitive to aerosol absorption. Coincident IR data also detected no significant amounts of ash in the SHV emissions (Prata et al., 2007[2]), possibly due to icing of ash particles, although rapid fallout of dense ash is considered the most likely reason for the low ash content. This fact, combined with the cloud's high altitude (see below) above jet cruising levels, probably minimized the cloud's impact on aviation.

$SO_2$ burdens in the cloud were calculated by summing all OMI pixels with retrieved $SO_2$ VCDs above 0.6 DU (the approximate $3\sigma$ noise level in OMI $SO_2$ retrievals for clouds at 15 km) (Krotkov et al., 2006) in daily images (Fig. 2). This procedure involves some error associated with calculation of $SO_2$ burdens in a cloud that at times straddled several adjacent OMI orbits and that was in motion between consecutive OMI orbits. An exponential fit of the $SO_2$ burdens provides an estimate of stratospheric $SO_2$ dispersion rates and the initial eruption mass (Fig. 2). We exclude the first OMI observation (20 May) from the fit as the cloud was probably too optically thick for accurate $SO_2$ retrievals at that time, and IR retrievals also indicate significant ice burdens in the cloud until ~30 h after emission (Prata et al., 2007[2]), which may have sequestered some $SO_2$. After 9 June the cloud became dispersed over a wide area, rendering mass calculations less accurate, hence these burdens are also excluded. Fitting the remaining data gives an initial eruption $SO_2$ mass of 0.22 Tg and a $SO_2$ dispersion rate of $\sim 9 \times 10^{-7}\ s^{-1}$, equivalent to an e-folding time of ~13 days. Extrapolation of the exponential decay curve yields a remnant $SO_2$ burden of 10 kilotons (kt) 38 days after the eruption, and ~1 kt after 67 days.

Derived sulfate aerosol was observed in backscatter data from the CALIOP lidar aboard CALIPSO. Fortuitously, CALIOP detected a sulfate aerosol layer located at ~20 km altitude over the Philippines in its 'first-light' image collected on 7 June 2006 (see: http://www.nasa.gov/mission_pages/calipso/news/First_Light.html). Inspection of the coincident OMI image of the SHV $SO_2$ cloud confirmed that the aerosol layer comprised sulfate derived from the volcanic $SO_2$, and also provided an accurate altitude

for the volcanic cloud. CALIOP was still able to clearly detect the stratospheric sulfate anomaly on 6 July (Fig. 3).

## 5 Discussion

The $SO_2$ mass in the cloud emitted by SHV on 20 May 2006 was not exceptional, being 2 orders of magnitude lower than the 1991 Pinatubo emission. The ability of OMI, CALIPSO, and other A-Train sensors to track such clouds for extended periods promises a significant advance in our understanding of the atmospheric impacts of moderate-scale volcanic eruptions. During 25 years of TOMS $SO_2$ measurements over 30 events that produced 0.1–0.3 Tg of $SO_2$ were detected, including the 1992 Mt. Spurr eruptions and the 1994 Rabaul eruption, none of which were tracked for more than 7 days by TOMS. OMI's high sensitivity to $SO_2$ therefore provides a new tool to investigate long-range transport of $SO_2$ clouds, validate trajectory models, and study the conversion of $SO_2$ to sulfate. Our observations suggest that moderate volcanic eruptions may play more of a role in sustaining the "nonvolcanic" background stratospheric sulfate layer, observed in periods between major volcanic inputs into the stratosphere (e.g., Hofmann and Rosen, 1981), than is widely recognized.

The presence of other gases, particularly halogen species, in the SHV stratospheric cloud is worth considering due to their potential effects on stratospheric chemistry and ozone abundance. We note that the volcanic cloud emitted by Hekla (Iceland) in February 2000, which contained ~0.2 Tg of $SO_2$ and hence was similar in magnitude to the SHV cloud, had caused dramatic in-cloud ozone destruction after only 33–34 h of atmospheric residence (Rose et al., 2006). Considering that the SHV aerosol layer was still clearly visible at an altitude of 19–20 km in CALIPSO lidar data on 6 July 2006 (Fig. 3), 47 days after the dome collapse, it is probable that the cloud had local impacts on stratospheric chemistry.

The tropospheric plume emitted by SHV during typical dome-forming activity contains significant concentrations of HCl (Edmonds et al., 2002), with $HCl/SO_2$ ratios



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often above 3 and up to ~6 measured in the months prior to the 20 May collapse by ground-based FTIR spectroscopy (MVO reports available at: http://www.mvo.ms). HCl exsolves from rising magma at low pressures within the lava dome, and MVO seismicity and field data suggest that the 20 May explosions probably occurred within the dome and upper conduit. The absence of erupted pumice and the resumption of lava dome growth on the same day also imply that only the shallow conduit was involved. Models of explosive eruption columns that account for scavenging processes in volcanic plumes imply that over 25% of emitted HCl can potentially reach the stratosphere (Textor et al., 2003). Stratospheric injection of HCl was hence very likely in this case, and has been confirmed using data from MLS on Aura (Prata et al., 2007[2]).

BrO has also been detected in SHV emissions (Bobrowski et al., 2003), although recent work invokes a secondary origin for volcanic BrO with primary magmatic HBr as the precursor (e.g., Oppenheimer et al., 2006). Gas phase oxidation of HBr by OH radicals and heterogeneous oxidation of HBr involving sulfate aerosol are two possible routes for BrO production in volcanic clouds (Oppenheimer et al., 2006), and both would have been possible during the long residence time of the SHV $SO_2$ cloud and the derived sulfate aerosol layer observed by CALIPSO.

## 6 Conclusions

The data presented here are the first example of coupled satellite observations of $SO_2$ and derived sulfate aerosol following a relatively modest volcanic eruption. Such measurements promise to greatly improve our knowledge of the atmospheric impacts of volcanic activity, and will probably eliminate any uncertainty as to the origin of future volcanically-induced stratospheric sulfate enhancements. This will enable a more rigorous definition of genuinely "nonvolcanic" stratospheric aerosol conditions, and permit assessments of potential effects of the proposed geoengineering strategy of injecting sulfate precursors such as $SO_2$ into the stratosphere to mitigate anthropogenic climate change (e.g., Wigley, 2006).



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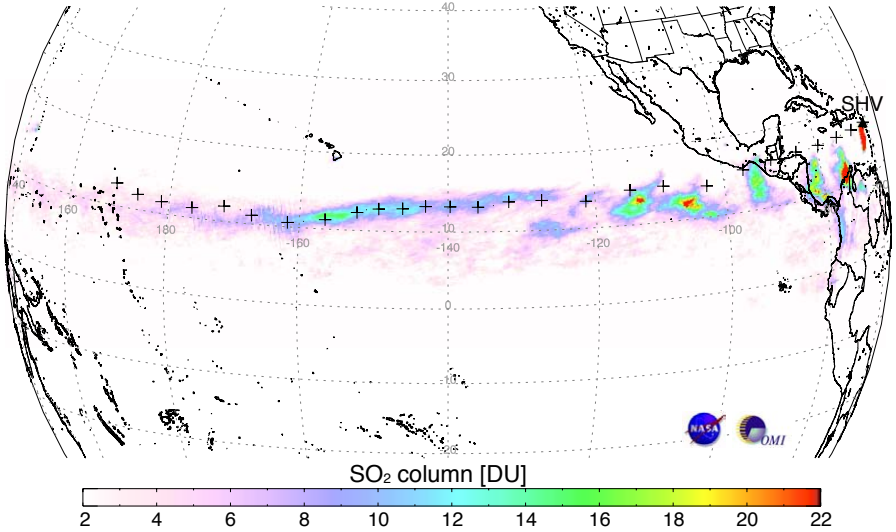


**Fig. 1.** Cumulative $SO_2$ VCDs measured by OMI in the SHV volcanic cloud from 20 May–6 June 2006 as the cloud crossed the Pacific Ocean. The dotted line is a HYSPLIT (Draxler and Rolph, 2003; Rolph, 2003) forward trajectory for a cloud at 20 km altitude, initialized at 11:00 UT on 20 May at SHV, with crosses plotted every 12 h. The trajectory covers 315 h (~13 days) of cloud transport.



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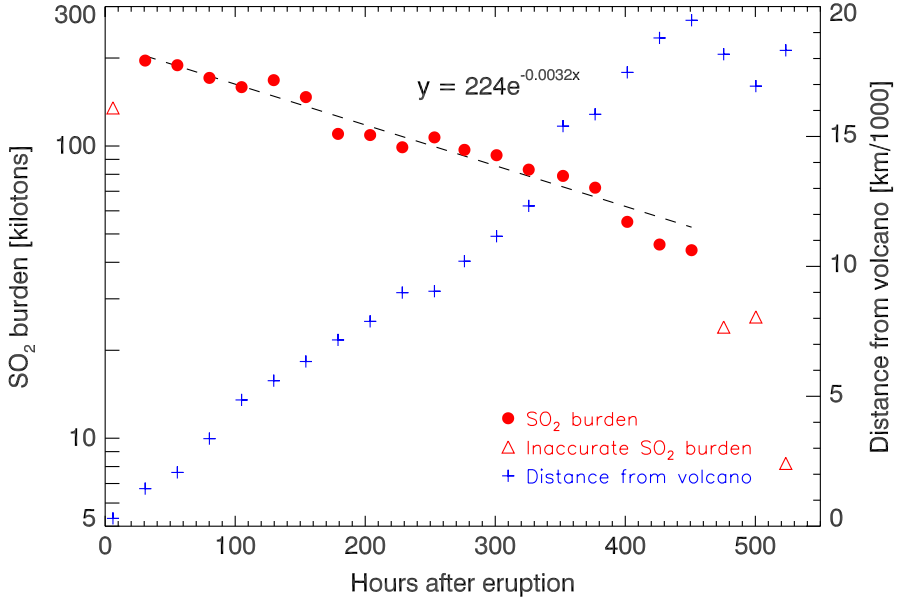


**Fig. 2.** Decay of the $SO_2$ burden measured by OMI in the SHV volcanic cloud and distance traveled by the cloud from 20 May–11 June 2006. The equation is an exponential fit (dashed line) to the $SO_2$ burdens denoted by circular symbols. The triangular symbols were excluded from the fit, for reasons discussed in the text. The location of the maximum $SO_2$ VCD on each day was used as a reference point to calculate the distance from SHV.



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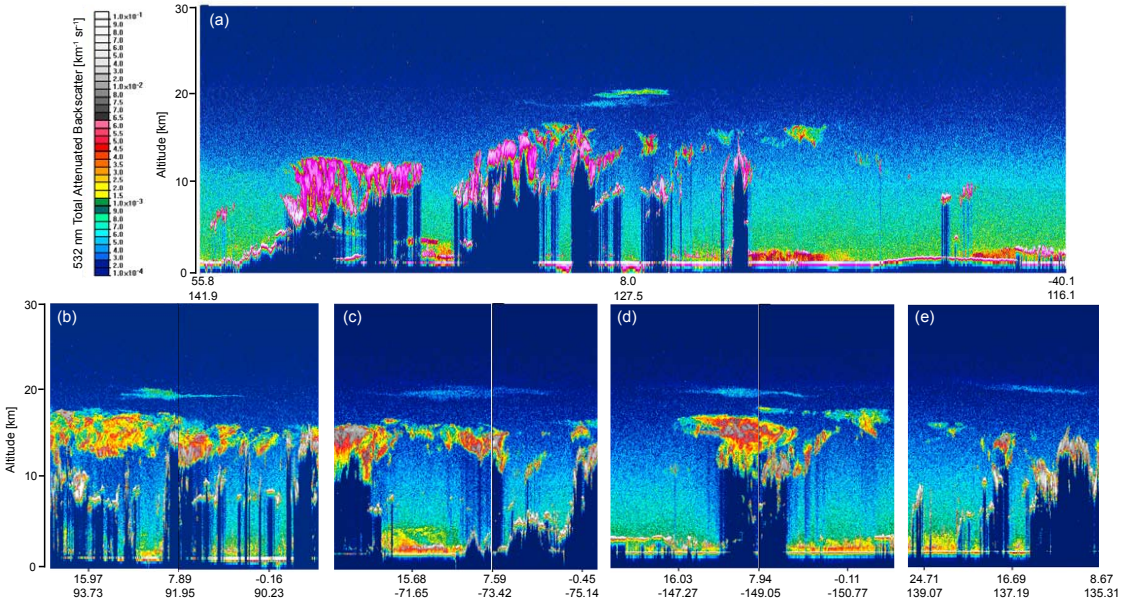


**Fig. 3.** CALIOP lidar curtains (532 nm attenuated backscatter) from the CALIPSO satellite, 7 June–6 July, 2006. Latitudes and longitudes of locations along the CALIPSO ground-track are given below each image. **(a)** "First-light" image on 7 June 2006 at 17:04 UT. The sulfate aerosol layer derived from the SHV $SO_2$ cloud can be clearly seen as a scattering layer in the tropics at an altitude of ~20 km; **(b)** 8 June at 19:39 UT; **(c)** 22 June at 06:40 UT; **(d)** 29 June at 06:23 UT; **(e)** 6 July at 16:33 UT. All data were taken under nighttime conditions.



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