Volcano monitoring applications of the Ozone Monitoring Instrument

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Abstract: The Ozone Monitoring Instrument (OMI) is a satellite-based ultraviolet (UV) spectrometer with unprecedented sensitivity to atmospheric sulphur dioxide (SO2) concentrations. Since late 2004, OMI has provided a high-quality SO2 dataset with near-continuous daily global coverage. In this review, we discuss the principal applications of this dataset to volcano monitoring: (1) the detection and tracking of large eruption clouds, primarily for aviation hazard mitigation; and (2) the use of OMI data for long-term monitoring of volcanic degassing. This latter application is relatively novel, and despite showing some promise, requires further study into a number of key uncertainties. We discuss these uncertainties, and illustrate their potential impact on volcano monitoring with OMI through four new case studies. We also discuss potential future avenues of research using OMI data, with a particular emphasis on the need for greater integration between various monitoring strategies, instruments and datasets.

The measurement of volcanic sulphur dioxide (SO2) degassing across various spatial and temporal scales of activity is one of the central components of volcano monitoring (Oppenheimer 2010; Sparks et al. 2012). Along with water and carbon dioxide, SO2 is among the most important volatile species emitted by volcanoes and, owing to relatively low ambient concentrations in the atmosphere and a distinctive spectral signature, it is particularly well-suited to remote sensing measurements (Stoiber et al. 1987; Galle et al. 2010; Oppenheimer et al. 2011). The SO2 output of a volcano can reveal much about magmatic processes beneath the volcano, including relationships between degassing and eruptive style (Oppenheimer et al. 2011). SO2 is of further interest owing to its impacts on Earth’s atmosphere and climate, as well as on the terrestrial biosphere and human society (Graf et al. 1997; Robock 2000; Delmelle 2003; Schmidt et al. 2012).

Volcanologists can currently exploit an unprecedented array of sensitive measurement techniques in the pursuit of accurate estimates of volcanic SO2 degassing (e.g. Mori & Burton 2006; McGonigle 2007; Galle et al. 2010; Oppenheimer et al. 2011). At an increasing number of volcanoes, SO2 emissions are continuously monitored, and may be integrated with measurements of seismicity, infrasound and deformation, as well as visual observations of activity (e.g. Edmonds et al. 2003; Arellano et al. 2008; Salerno et al. 2009; Galle et al. 2010). However, this level of monitoring is not typical, and owing to safety concerns, lack of funding or logistical problems resulting from their remote locations, gas emissions at the great majority of Earth’s active volcanoes are rarely or never measured (Sparks et al. 2012).

Recent technological advances in satellite-based spectrometers have led to unprecedented sensitivity to atmospheric SO2 as measured from space. This has resulted in a significant increase in the use of satellite datasets for the detection, monitoring and quantification of volcanic SO2 emissions (Bluth et al. 1993; Krueger et al. 1995; Carn et al. 2008; Rix et al. 2009; McCormick et al. 2012).
Although satellites have traditionally been used to study major explosive eruptions, the technological advances and improved sensitivity of modern satellite instruments may enable remote sensing of the full range of volcanic SO2 emissions, from passive degassing to mild intermittent Strombolian activity to large explosive paroxysms. Satelliteborne instruments therefore offer the potential for long-term regular volcano monitoring over global scales, avoid the inherent dangers of monitoring hazardous volcanic activity from close proximity on the ground and may also be more cost-effective than major ground-based monitoring networks. However, the use of satellites is not without uncertainty – a number of factors that may limit their application to studies of volcanic degassing remain poorly understood and in some cases completely unexplored.

In this article, we critically review the use of the Ozone Monitoring Instrument (OMI), a satellite-based UV–visible spectrometer for studies of volcanic degassing. We begin (in the section ‘Satellite monitoring of volcanic SO2 emissions’) with an overview of OMI and satellite remote sensing of SO2 emissions (focusing on UV instruments) and justify the use of satellites for the detection and tracking of large eruption clouds as well as for long-term monitoring of gas emissions. In the sections ‘OMI observations of major volcanic eruptions’ and ‘Long-term monitoring of degassing with OMI’ we provide a literature review of OMI’s use in these two major applications. In the section ‘Limitations of OMI’s application to volcano monitoring’, we present a discussion of the main uncertainties and limitations of OMI (and satellite instruments generally) to long-term studies of volcanic monitoring. These uncertainties are illustrated by new case studies in the section ‘Case studies of regional monitoring with OMI’, and lastly we provide an outlook for both OMI and future UV instruments.

Satellite monitoring of volcanic SO2 emissions

Satellite-based UV remote sensing of SO2 prior to OMI

The fortuitous detection of the large SO2 cloud released in the 1982 eruption of El Chichón by the Total Ozone Mapping Spectrometer (TOMS) was the first successful observation of volcanic SO2 emission from space (Krueger 1983; Schneider et al. 1999; Krueger et al. 2008). With relatively good spatial resolution and contiguous daily global coverage, the TOMS instrument family provided a robust and near-continuous record of volcanic SO2 releases by large eruptions for over two decades (Carn et al. 2003). Notable eruptions detected by TOMS include Nevado del Ruiz in 1985 (Krueger et al. 1990), Pinatubo in 1991 (Bluth et al. 1992) and Cerro Hudson in 1991 (Schoerberl et al. 1993), which all released very large quantities of SO2. The full TOMS eruption record, including that of the more sensitive Earth-Probe TOMS (operational from 1997), is discussed by Carn et al. (2003).

Subsequent UV instruments to TOMS included the European Global Ozone Monitoring Experiment (GOME) and Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) (Table 1). These instruments were hyperspectral imagers, where multiple wavelengths in the UV spectrum were used simultaneously in the retrieval of atmospheric trace gas concentrations. These instruments had poorer spatial resolution than TOMS, however, and did not achieve daily global coverage. Nonetheless, these instruments were demonstrated to be capable of detecting SO2 emissions from volcanic eruptions (e.g. Loyola et al. 2008). OMI combines the key advantageous traits of TOMS, GOME and SCIAMACHY (good spatial resolution, contiguous daily global coverage and hyperspectral imaging) in a single sensor.

The Ozone Monitoring Instrument

OMI was launched in 2004 as a component of NASA’s Earth Observation System (EOS) programme, carried aboard the Aura satellite in the ‘A-train’ constellation (Levett et al. 2006; NASA 2010). The A-train also contains a number of other UV instruments capable of detecting atmospheric SO2, including the Moderate Resolution Imaging Spectroradiometer (MODIS) and the Advanced Spaceborne Thermal Emission Spectrometer (ASTER), as well as the infrared (IR) Atmospheric Infrared Sounder (AIRS; Table 1). The advantage of the satellite constellation configuration lies in the potential inter-comparison of near-simultaneously gathered data from different instruments. The combined use of UV and IR instruments is complementary since IR sensors are not restricted to daylight operations and UV SO2 retrievals may be less affected by interference from water vapour.

OMI is a hyperspectral UV–vis spectrometer, designed to provide significant advances in SO2 detection over earlier UV instruments. OMI measures top-of-atmosphere radiances, resulting from solar backscatter and reflection from the Earth’s surface and atmosphere. This radiance spectrum is used to derive column concentrations of trace gases and other atmospheric parameters (Levett et al. 2006). The use of the full UV–vis spectrum (an approach inherited from GOME and SCIAMACHY) enables the simultaneous detection...
Table 1. Summary of recent satellite-based instruments used in the detection of volcanic SO$_2$. All except TOMS, GOME and SCIAMACHY are currently operational. All instruments except MSG-SEVIRI are carried by satellites flying a Sun-synchronous polar orbit; MSG-SEVIRI is in a geostationary orbit. Detection limits are calculated for a plume comprising five nadir pixels at 5σ, where σ is the standard deviation of scene noise, and refer to detection of SO$_2$ in the upper troposphere/lower stratosphere (Carn et al. 2003).

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Satellite</th>
<th>Data range</th>
<th>Equatorial overpass time</th>
<th>Nadir pixel dimensions</th>
<th>SO$_2$ detection limit (t)</th>
<th>Comments</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ultraviolet</td>
<td>TOMS</td>
<td>Earth-Probe</td>
<td>November 1997 to December 2005</td>
<td>11:16</td>
<td>$39 \times 39$ km</td>
<td>1440–3800</td>
<td>Global daily coverage</td>
</tr>
<tr>
<td>GOME</td>
<td>ERS-2</td>
<td>April 1995 to July 2011</td>
<td>10:30</td>
<td>$40 \times 320$ km</td>
<td>3600</td>
<td>Global coverage in 3 days</td>
<td></td>
</tr>
<tr>
<td>GOME-2</td>
<td>MetOp-A</td>
<td>October 2006 to present</td>
<td>09:30</td>
<td>$80 \times 40$ km</td>
<td>650</td>
<td>Global coverage in 1.5 days</td>
<td>Rix et al. (2009)</td>
</tr>
<tr>
<td>SCIAMACHY</td>
<td>ENVISAT</td>
<td>March 2002 to April 2012</td>
<td>10:00</td>
<td>$30 \times 60–120$ km</td>
<td>125</td>
<td>Global coverage in 6 days</td>
<td>Frankenberg et al. (2006)</td>
</tr>
<tr>
<td>OMI</td>
<td>Aura</td>
<td>July 2004 to present</td>
<td>13:45</td>
<td>$24 \times 13$ km</td>
<td>26</td>
<td>Global daily coverage</td>
<td>Levett et al. (2006)</td>
</tr>
<tr>
<td>Infrared</td>
<td>ASTER</td>
<td>Terra</td>
<td>December 1999 to present</td>
<td>10:30</td>
<td>$15/30/90$ m (circles)</td>
<td>0.05</td>
<td>Selected acquisitions only</td>
</tr>
<tr>
<td>MODIS</td>
<td>Terra</td>
<td>December 1999 to present, May 2002 to present</td>
<td>10:30</td>
<td>1 km</td>
<td>6</td>
<td>Global coverage in 1–2 days</td>
<td>Watson et al. (2004); Corradini et al. (2010)</td>
</tr>
<tr>
<td></td>
<td>Aqua</td>
<td></td>
<td>13:45</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>AIRS</td>
<td>Aqua</td>
<td>May 2002 to present</td>
<td>13:30</td>
<td>13.5 km (circle)</td>
<td>100</td>
<td>Global coverage in 2–3 days</td>
<td>Prata &amp; Bernardo (2007); Prata &amp; Kerkmann (2007)</td>
</tr>
<tr>
<td>MSG-SEVIRI</td>
<td>Meteosat-8</td>
<td>August 2008 to present</td>
<td>N/A</td>
<td>3 km</td>
<td>140</td>
<td>Measurements every 15 min</td>
<td>Karagulian et al. (2010); Walker et al. (2012); Carboni et al. (2012)</td>
</tr>
<tr>
<td>IASI</td>
<td>MetOp-A</td>
<td>June 2006 to present</td>
<td>09:30/21:30</td>
<td>12 km (circle)</td>
<td>7.5</td>
<td>Global coverage twice daily</td>
<td></td>
</tr>
</tbody>
</table>
of multiple trace gases, including SO$_2$, ozone, NO$_2$, BrO and OClO, as well aerosol loadings and cloud properties (Ahmad et al. 2003; Levelt et al. 2006). Like TOMS, OMI provides continuous daily global coverage, by means of a 2600 km-wide swath and Aura’s 14 daily polar sun-synchronous orbits, but has a nadir spatial resolution of 24 $\times$ 13 km$^2$, the best yet for a space-borne UV instrument (Fig. 1). Additionally, improvements in spectral resolution (c. 0.5 nm) and reduced noise levels have, together with the higher spatial resolution and use of multiple wavelengths, resulted in greatly decreased SO$_2$ detection limits over all prior UV sensors. Further increased sensitivity over TOMS results from the use of relatively more sophisticated retrieval algorithms; for discussion of these and a full overview of the OMI dataset, we refer the reader to a companion paper by Carn et al. (2013), as well as Yang et al. (2007, 2009a, b, 2010). OMI’s improved sensitivity over TOMS was demonstrated by their simultaneous retrievals of the SO$_2$ cloud released in the 27 January 2005 eruption of Manam Volcano, Papua New Guinea (Carn et al. 2009a). Both instruments reported similar SO$_2$ maxima in the core of the cloud, but OMI’s visualization of the diffuse cloud margins was noticeably better than that of TOMS. Similar results were demonstrated for the October 2005 eruption of Sierra Negra Volcano, Galápagos Islands (Thomas et al. 2009).

Satellite remote sensing for aviation hazard mitigation

Satellite-mounted instruments are critical for the detection and tracking of ash and SO$_2$ clouds from volcanic eruptions for aviation hazard mitigation (Prata 2009). While serious encounters are thankfully rare, aircraft that fly through volcanic clouds typically sustain considerable damage (e.g. Casadevall 1994a, b; Tupper et al. 2006; Rose et al. 2006). The recent Icelandic eruptions of Eyjafjallajökull (2010) and Grimsvötn (2011) demonstrated the potential economic impact of widely dispersed volcanic ash plumes on the aviation industry (Thomas & Prata 2011; Stohl et al. 2011, Marenco et al. 2011; Heard et al. 2012). These eruptions resulted in major overhauls of mitigation strategies and response infrastructure, as well as greater awareness of the need for better integration of various data sources (Sammonds et al. 2010; Dopagne 2011).

UV sensors offer certain major advantages to aviation hazard mitigation. IR ash retrievals can fail for optically very thick clouds shortly following eruption, if there is little thermal contrast between the volcanic cloud top and adjacent meteorological clouds, or if the ash particles are encased by ice owing to their high altitude (Carn et al. 2009a; Rose et al. 1995). UV retrievals of ash, or more frequently SO$_2$ as a proxy for ash cloud location, are less limited by these conditions. While the use of

**Fig. 1.** Schematic rendering of OMI’s measurement technique, summarizing key operational and orbital parameters. Inset shows the variation in OMI’s ground footprint spatial resolution, from nadir to the swath edge. After Levelt et al. (2006).
SO₂ as a proxy for ash has been utilized effectively in aviation hazard mitigation during recent eruptions (Thomas & Prata 2011), we note that, owing to vertical shearing, volcanic clouds can separate into distinct SO₂ and ash components (Prata & Kerkmann 2007).

Ash is arguably a more severe hazard than SO₂, causing abrasion of forward-facing airframe surfaces, interference with avionics and degradation of engine performance through melting and re-solidification within the engine interior (Tupper et al. 2004). Problems caused by SO₂ and its derivative sulphuric acid aerosol include crazing of cockpit and cabin windows, forward airframe damage and sulphate precipitation in turbines (Carn et al. 2009a). Detection and tracking of both these species rely heavily on satellite data, since high winds in the upper atmosphere have the potential to transport volcanic clouds over huge areas in relatively little time. This is particularly true if volcanic clouds penetrate jet-streams, as was observed following the eruptions of Nyamuragira (D.R. Congo) in 2006 and Jebel at Tair (Yemen) in 2007 (Carn et al. 2009a). Jet-streams are frequently sought by airliners to save time and fuel, potentially increasing the likelihood of encounters with volcanic clouds. The major volcanic hazard comes from explosive eruptions, since these often inject ash and SO₂ into the upper troposphere/lower stratosphere (UTLS), close to airliner cruising altitude. In the tropics and near-Equatorial regions, the air entrained in the plume is more humid, increasing its buoyancy and meaning that greater altitudes may be achieved some time after emission by SO₂ clouds which were initially not injected to hazardous levels of the atmosphere (Tupper et al. 2006, 2009). Bourassa et al. (2012) propose that the large SO₂ cloud from the 2011 Nabro eruption reached the UTLS via ascent in the anticyclonic Asian monsoon circulation.

While TOMS was a very effective instrument in aviation hazard mitigation, the improved sensitivity of OMI ought to enable observations of a greatly extended range of volcanic phenomena: the large SO₂ clouds injected into the UTLS may be trackable over greater distances and timescales after detection, and clouds from smaller eruptions (e.g. Volcanic Explosivity Index (VEI) c. 2–4) should be detectable and more accurately resolvable, as will diffuse cloud margins.

Long-term monitoring of volcanic emissions with satellites

A more complete assessment of persistent and continuous SO₂ degassing is sought for many applications. Long-term records of volcanic SO₂ are crucial for understanding patterns and trends in degassing at individual volcanoes and on regional scales, and ultimately may feed into volcanic hazard and risk assessment (Galle et al. 2010). Increasing activity at many volcanoes is preceded by elevated gas emissions, with SO₂ in particular escaping from decompressing magma (Symonds et al. 1994). Measurements of SO₂ emissions prior to the 1991 Pinatubo eruption provided crucial evidence for the successful forecast of a major imminent eruption, and the subsequent timely evacuation, responsible for saving c. 30 000 lives (Minnis et al. 1993; Daag et al. 1996; Oppenheimer et al. 2011). Pre-eruptive increases in SO₂, along with other monitored parameters, similarly provided advance warning of imminent eruption at Merapi (Indonesia) in 2010, facilitating evacuation and other response measures (Surono et al. 2012). Greater output of SO₂ has also been noted prior to eruptions at the Cook Inlet volcanoes, Alaska (Werner et al. 2011) and at Soufrière Hills, Montserrat (Edmonds et al. 2010). Understanding the climatic impact of tropospheric SO₂ in terms of local or regional pollution (Delmelle 2003) and sulphate-driven radiative forcing via the direct (Graf et al. 1997) and indirect aerosol (Schmidt et al. 2012) effects are also important, and dependent on accurate assessment of volcanic emission fluxes.

Deposition of SO₂ from the troposphere into the biosphere may also have a major impact on terrestrial vegetation, soil and groundwater pH and human health (Delmelle 2003). Extending the spatial and temporal coverage of volcanic SO₂ datasets would aid in producing global emission budgets of other volcanic volatiles, and in constraining the role played by volcanoes in global volatile cycling between major Earth reservoirs, such as the mantle and the atmosphere (Hilton et al. 2002; Fischer 2008; Pyle & Mather 2009; Wallace & Edmonds 2011).

Most global SO₂ budgets show that the largest fraction of average annual volcanic SO₂ release, at least on a decadal or longer timescale, is from persistent degassing into the troposphere (e.g. Berreheim & Jaeschke 1983; Pyle et al. 1996; Andres & Kasgnoc 1998; Halmer et al. 2002). Measurement of this degassing is largely by ground-based techniques such as correlation spectroscopy (COSPEC) and differential optical absorption spectroscopy (DOAS; e.g. Williams-Jones et al. 2008; Galle et al. 2010). A major limitation of existing global estimates of persistent tropospheric degassing, however, is a reliance on short measurement campaigns (which are logistically easier to implement than continuous monitoring) and a lack of data from many remote or inaccessible volcanoes. Although more volcanoes than ever before are now regularly monitored for a range of...
geochemical and geophysical parameters, the majority are not (Sparks et al. 2012). The potential impact of large, multi-year, global coverage satellite datasets in extending monitoring coverage is clear. However, the use of satellites in the monitoring of degassing remains a novel application since most lack sufficient sensitivity to SO$_2$, particularly in the lower atmosphere. Instruments such as TOMS detected major passive degassing events, for example at Miyake-jima and Popocatépetl (Carn et al. 2003), but generally there is little precedent for the studies we review below. It is only in the last decade that technological advances in satellite-based instruments have provided the capacity to attempt assessments of persistent volcanic degassing. Despite the sensitivity of instruments like OMI, there are a number of factors that limit the potential for satellites to resolve passive degassing fluxes into the troposphere, which will be described in the section ‘Long-term monitoring of degassing with OMI’.

**OMI observations of major volcanic eruptions**

*Database of detected eruptions*

Since the last published summary of large eruption clouds detected and tracked by OMI (Carn et al. 2009a), many further observations have been documented. We present here a representative selection of eruptions from throughout OMI’s lifetime in order to illustrate the great variety of eruption styles, intensities, durations and locations observed (Fig. 2; Table 2). We largely focus on those eruptions that have been discussed in the published OMI literature; for a more complete list, which necessarily includes some preliminary data, we refer the reader to the Global Sulphur Dioxide Monitoring home page (http://SO2.gsfc.nasa.gov/).

Since 2004, OMI has detected SO$_2$ clouds from eruptions of $>30$ volcanoes. The broad geographical spread of these volcanoes (Fig. 2) indicates that OMI’s detection is remarkably robust regardless of location. The only circumstance where OMI could fail to detect an eruption cloud owing to its location would be if a high-latitude plume were to be blown beyond the northern or southern terminators in the boreal or austral winter respectively, where it then remained. Although relatively few volcanoes lie in latitudes where this is likely, the SO$_2$ cloud released by the 2009 Sarychev Peak eruption did spend long periods above the Arctic Circle (Carn & Lopez 2011). The eruptions detected by OMI also cover a broad range of SO$_2$ burdens. OMI has observed drifting clouds from several major eruptions, such as the North Pacific eruptions at Okmok and Kasatochi (Aleutian Islands), Redoubt (Alaska) and Sarychev Peak (Kuril Islands) between 2008 and 2009. These eruptions provided many opportunities for researchers. The long-lived drifting clouds from Okmok and Sarychev Peak were exploited for validation studies comparing OMI data with estimates of SO$_2$ column using ground-based methodologies (Spinei et al. 2010; Carn & Lopez 2011). The continued

![Fig. 2. World map showing the location of selected volcanic eruptions with SO$_2$ release detected by OMI. Grey circles mark volcano locations, and area of the circles corresponds to maximum daily SO$_2$ mass burden measured by OMI.](http://sp.lyellcollection.org/)

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### Table 2. Major eruptions with SO₂ release detected by OMI, 2004–2011. Data source for SO₂ masses was OMI SO₂ group website; for eruption parameters and volcano locations it was the Smithsonian Institution’s Bulletin of the Global Volcanism Network

<table>
<thead>
<tr>
<th>Volcano</th>
<th>Location</th>
<th>Eruption start date</th>
<th>Maximum daily SO₂ burden ($\times 10^6$ kg)</th>
<th>Maximum plume height (km)</th>
<th>Number of days plume tracked by OMI</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sierra Negra</td>
<td>Galapagos Islands 0.83°S, 91.17°W</td>
<td>22 October 2005</td>
<td>416</td>
<td>15.2</td>
<td>10</td>
<td>Thomas et al. (2009)</td>
</tr>
<tr>
<td>Soufrière Hills</td>
<td>West Indies 16.72°N, 62.18°W Ecuador</td>
<td>20 May 2006</td>
<td>196</td>
<td>16.8</td>
<td>22</td>
<td>Carn et al. (2009a)</td>
</tr>
<tr>
<td>Tungurahua</td>
<td>1.467°S, 78.442°W</td>
<td>16 August 2006</td>
<td>35</td>
<td>15–16</td>
<td>3</td>
<td>Carn et al. (2008)</td>
</tr>
<tr>
<td>Rabaul</td>
<td>Papua New Guinea 4.271°S, 152.203°E</td>
<td>7 October 2006</td>
<td>282</td>
<td>18</td>
<td>12</td>
<td>Carn et al. (2009a); McCormick et al. (2012)</td>
</tr>
<tr>
<td>Nyamuragira</td>
<td>D.R. Congo 1.408°S, 29.20°E</td>
<td>27 November 2006</td>
<td>690</td>
<td>10</td>
<td>14</td>
<td>Carn et al. (2009a)</td>
</tr>
<tr>
<td>Manda Hararo</td>
<td>Ethiopia 12.17°N, 40.82°E</td>
<td>12 August 2007</td>
<td>5.3</td>
<td>–</td>
<td>2</td>
<td>Smithsonian Institution (2007c)</td>
</tr>
<tr>
<td>Jebel at Tair</td>
<td>Red Sea, Yemen 15.55°N, 41.83°E</td>
<td>1 October 2007</td>
<td>52</td>
<td>10</td>
<td>13</td>
<td>Carn et al. (2009a); Thomas et al. (2011)</td>
</tr>
<tr>
<td>Llaima</td>
<td>Southern Chile 38.692°S, 71.729°W</td>
<td>1 January 2008</td>
<td>30</td>
<td>12.5</td>
<td>6</td>
<td>Smithsonian Institution (2008a, b)</td>
</tr>
</tbody>
</table>

(Continued)
<table>
<thead>
<tr>
<th>Volcano</th>
<th>Location</th>
<th>Eruption start date</th>
<th>Maximum daily SO₂ burden ($\times 10^6$ kg)</th>
<th>Maximum plume height (km)</th>
<th>Number of days plume tracked by OMI</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chaitén</td>
<td>Southern Chile</td>
<td>2 May 2008</td>
<td>5.6</td>
<td>20+</td>
<td>7</td>
<td>Carn et al. (2009b)</td>
</tr>
<tr>
<td>Okmok</td>
<td>Aleutian Islands</td>
<td>12 July 2008</td>
<td>122</td>
<td>15+</td>
<td>9</td>
<td>Spinei et al. (2010); Thomas et al. (2011)</td>
</tr>
<tr>
<td>Kasatochi</td>
<td>Aleutian Islands</td>
<td>7 August 2008</td>
<td>1581</td>
<td>13.7</td>
<td>35</td>
<td>Krotkov et al. (2010); Thomas et al. (2011)</td>
</tr>
<tr>
<td>Redoubt</td>
<td>SW Alaska</td>
<td>22 March 2009</td>
<td>60</td>
<td>18.2</td>
<td>13</td>
<td>Lopez et al. (in press)</td>
</tr>
<tr>
<td>Sarychev Peak</td>
<td>Kuril Islands</td>
<td>12 June 2009</td>
<td>1200</td>
<td>13.7</td>
<td>50</td>
<td>Carn &amp; Lopez (2011)</td>
</tr>
<tr>
<td>Manda Hararo</td>
<td>Ethiopia</td>
<td>29 June 2009</td>
<td>3.9</td>
<td>–</td>
<td>2</td>
<td>Smithsonian Institution (2009)</td>
</tr>
<tr>
<td>Eyjafjallajökull</td>
<td>Southern Iceland</td>
<td>14 April 2010</td>
<td>27</td>
<td>9</td>
<td>15</td>
<td>Thomas &amp; Prata (2011)</td>
</tr>
<tr>
<td>Merapi</td>
<td>Java, Indonesia</td>
<td>29 October 2010</td>
<td>225</td>
<td>15.2</td>
<td>18</td>
<td>Surono et al. (2012)</td>
</tr>
<tr>
<td>Grimsvötn</td>
<td>NE Iceland</td>
<td>21 May 2011</td>
<td>220</td>
<td>20</td>
<td>20</td>
<td>Smithsonian Institution (2011)</td>
</tr>
<tr>
<td>Nabro</td>
<td>Eritrea</td>
<td>12 June 2011</td>
<td>$&gt;2000$</td>
<td>19</td>
<td>16</td>
<td>Carn et al. (in preparation); Clarisse et al. (2012)</td>
</tr>
</tbody>
</table>
elevated degassing at Redoubt after its eruption enabled comparison with airborne measurements and estimates of OMI’s detection limit at high latitudes to be made (Lopez et al. in press). The SO2 release from Kasatochi was the largest in a volcanic eruption since Cerro Hudson (1991), and the largest at high northerly latitudes in the satellite era (Krotkov et al. 2010). The largest low-latitude SO2 release since Mount Pinatubo (1991) was observed following the eruption of Nabro Volcano, in June 2011 (Carn et al. in preparation; Clarisse et al. 2012).

OMI has also demonstrated an ability to detect the SO2 clouds released by smaller eruptions (VEI < 4) which are important to characterize well owing to their more frequent occurrence. Eruptions where much of the SO2 burden was released into the troposphere rather than the stratosphere, such as Rabaul (Papua New Guinea) in 2006 and Piton de la Fournaise (Réunion) in 2007, were nonetheless well characterized (Carn et al. 2009a; McCormick et al. 2012; Thomas et al. 2011; Tulet & Villeneuve 2011). This is particularly encouraging since the higher levels of atmospheric water vapour at low altitudes can negatively impact IR retrievals of SO2 (Watson et al. 2004). Eruptions detected that have low to moderate SO2 release, shorter duration or lower altitude plumes include Merapi (2010) in Indonesia, Eyjafjallajökull (2010) and Grimsvötn (2011) in Iceland, Puyehue (2011) and Llaima (2008) in Chile, Tongurahua (2006) in Ecuador and Manda Hararo (2007 and 2009) in the Afar, Ethiopia. A particularly interesting example is the 2008 eruption of Chaitén (Chile), which despite the high intensity of the explosive eruption, the stratospheric penetration achieved by the cloud and the subsequent widespread ash fall-out, released strikingly low amounts of SO2 (Carn et al. 2009b). The rarity of rhyolitic eruptions like Chaitén makes collection of novel data such as this particularly important. OMI also detected small and short-lived eruptions from volcanoes with little prior evidence of activity, the Garbuna Group (Papua New Guinea) in 2005, Raoul Island (Kermadec Islands) in 2006 and Fourpeaked (Alaska) in 2006 (Carn et al. 2013).

OMI has also demonstrated consistently an ability to track drifting clouds for extended periods of time, an essential component of aviation hazard mitigation, since even dilute and aged volcanic clouds can be harmful to aircraft (Tupper et al. 2006; Rose et al. 2006; Carn et al. 2009a). The fast-moving clouds from eruptions at Nyamuragira and Jebel al Tair that entered the jet-stream were closely monitored for almost two weeks after initial emission (Carn et al. 2009a). The 2006 dome collapse at Soufrière Hills (Montserrat) released an SO2 cloud measured initially by OMI as only c. 0.2 × 10⁶ kg, but was nonetheless tracked for 23 days, until it finally dropped below detection limit 26 000 km west of the volcano (Carn et al. 2007). We note that this was partly attributable to the high injection altitude, c. 20 km (Carn & Prata 2010). The two month-long hemispheric transport of Kasatochi’s eruption cloud was observed consistently by OMI (Krotkov et al. 2010; Thomas et al. 2011). Similarly, the Sarychev Peak eruption cloud was tracked from 10 June to 31 July 2009, despite its frequent excursions into the Arctic Circle (Carn & Lopez 2011).

**Estimating total erupted SO2 mass**

Given that OMI has demonstrated its use as a tool for the detection and tracking of drifting SO2 clouds released by explosive eruptions (from around the world and on various scales), a key aim is to effectively determine the total SO2 release of an eruption. Large volcanic eruptions with major SO2 releases can have major impacts on the radiative balance of Earth’s atmosphere and hence climate (Schoerber et al. 1992; Robock 2000; Blake 2003). Linking improved measurements of eruptive SO2 release to subsequent perturbations in global temperatures will aid predictions of climate forcing and change, as well as informing potential controversial geoengineering strategies (Crutzen 2006; Robock 2008a, b; Robock et al. 2009; Jones et al. 2010).

Estimating the total release of SO2 from an eruption using OMI is complicated by the fact that it returns a daily snapshot of total SO2 loading. Between these snapshots SO2 may be added by further volcanic activity, may be lost via chemical reactions, may be diluted below the instrument’s detection limit or may persist in the atmosphere. One simple method of estimating total SO2 released in an eruption is to determine SO2 loss rate from a time series of daily OMI observations of SO2 mass burden in the evolving cloud (Carn et al. 2007). Estimates of loss rate may be used to extrapolate backwards from the time of the initial OMI observation of the SO2 cloud, to the time of the original SO2 emission. Chemical processing of SO2 (see the section ‘Persistence or transience of SO2 clouds’) is frequently slow in the UTLS, with e-folding times on the order of days rather than hours (Robock 2000; Krotkov et al. 2010), and this results in relatively long-lived clouds being tracked by OMI for several days. Much faster chemical processing in the lower atmosphere generally precludes the use of this method for tropospheric eruption clouds. This approach may have some validity in certain instances, for example, some volcanic eruptions involve a single, near-instantaneous release of SO2, smooth transport by
wind, and a straightforward evolution of gradually decreasing SO$_2$ cloud mass, such as the May 2006 dome collapse at Soufrière Hills (Carn & Prata 2010; Fig. 3a). The cloud’s mass decrease was matched by smoothly increasing distance between the OMI pixel with the maximum detected SO$_2$ column and the volcano, reflecting the fact that there was no significant further SO$_2$ emission at source.

However, many eruptions are more complex. Sustained SO$_2$ release, for example from the week-long fissure eruption of Sierra Negra in October 2005 (Geist et al. 2008) or the repeated explosive eruptions at Sarychev Peak in June 2009 (Rybin et al. 2011), precludes a simple mass decay curve in the OMI time series. In the hot and humid equatorial troposphere, mass decrease by rapid SO$_2$ processing and dilution competes with continued output from Sierra Negra to give a saw-tooth profile of SO$_2$ burden (Fig. 3b). Slower loss in the higher-latitude UTLS cloud from Sarychev Peak, however, results in stable SO$_2$ mass throughout the duration of the eruption (Fig. 3c). Sustained degassing in both cases is testified by the maximum SO$_2$ column location remaining close to the vent until the eruption has ceased, at which point dispersal of the cloud becomes dominant.

Further complexity in syn-eruption time series such as these may be introduced by secondary SO$_2$ produced from the post-emission oxidation of hydrogen sulphide (H$_2$S), another S-bearing volcanic gas species which, owing to measurement issues, is frequently not accounted for as part of sulphur emission budgets (e.g. Aiuppa et al. 2005). To date, eruptive H$_2$S release has only been measured once from space, by the Infrared Atmospheric Sounding Interferometer (IASI) during the 2008 eruption of Kasatochi (Clarisse et al. 2011). Conversion of H$_2$S to SO$_2$ via in-plume oxidation occurs on timescales on the order of a few days (Textor et al. 2003) and this may also cause increased mass burdens of SO$_2$ to be measured on days following an eruption, even if the eruption had ceased, and large SO$_2$ output had apparently stopped.

Resolving these different contributions to measured SO$_2$ mass burden from OMI observations alone may not be possible for many eruptions. Major changes in mass burden can occur between daily observations of the cloud and are therefore not easily resolvable. In an eruption’s early phases, intervals of increased SO$_2$ processing owing to scavenging from the plume by ice or ash particles may not be noticed. The gaps between OMI over-passes may simply be too great for accurate assessment of an SO$_2$ cloud’s evolution. A synergistic approach integrating data from multiple satellites, however, could reduce these data gaps (Table 1).

Several studies have compared different instruments in a range of volcanic scenarios (Thomas et al. 2009, 2011; Rix et al. 2009; Carn & Prata 2010; Corradini et al. 2010). A greater degree of integration between satellite datasets also provides useful input as well as validation for both trajectory and inverse modelling techniques, which in turn provide a more sophisticated means of estimating SO$_2$ lifetime and hence calculating total emitted mass (e.g. Kristiansen et al. 2010; Krotkov et al. 2010).

**Long-term monitoring of degassing with OMI**

**Published surveys of degassing**

Long-term high-temporal-resolution OMI surveys of tropospheric SO$_2$ degassing have been published for Ecuador and southern Colombia (Carn et al. 2008), Vanuatu (Bani et al. 2012) and Papua New Guinea (McCormick et al. 2012; Fig. 4), with new case studies presented in Carn et al. (2013). In each case, prior observations of degassing from these regions were sparse. These OMI surveys were able to identify and distinguish the actively degassing volcanoes, a clear demonstration of OMI’s spatial resolution, and to assess the relative strength and persistence of each volcano’s SO$_2$ emissions (Table 3). Over each region, OMI was shown to be able to detect SO$_2$ degassing consistently, and to observe changing levels of emission, with a high frequency of detection compared with complementary methods of continuous satellite-based monitoring (e.g. MODIS/ASTER thermal anomaly alerts, or Volcanic Ash Advisories). Multi-year time series of OMI-measured SO$_2$ burdens over the regions were produced, and these showed generally strong correlations with independent observations of volcanic activity.

OMI’s sensitivity to changes in volcanic degassing intensity and style can be illustrated by an increasing number of examples. Changes in SO$_2$ mass burden linked to cycles of conduit sealing and degassing were observed in daily OMI data.
over Reventador and Galeras, along with elevated SO2 emissions in the months prior to major explosive eruptions at Tungurahua (Carn et al. 2008). At Ambrym, a large peak in the OMI SO2 record coincides with ground observations and measurements of an exceptional surge in degassing during late 2004 and early 2005 (Bani et al. 2009, 2012). In Papua New Guinea, major eruptions at Manam (January 2005) and Rabaul (October 2006) were detected by OMI as well as observed from the ground, and the onset of an eruptive cycle at Langila (beginning April 2005) and elevated degassing from Rabaul (December 2007, February 2008 onwards) were also well correlated (McCormick et al. 2012). OMI also detected a major SO2 release from Bagana in June 2005, which was undetected from the ground (McCormick et al. 2012). Variations in SO2 detection by OMI between

Table 3. Active sources of volcanic SO2 in three regional-scale OMI surveys made by OMI. The volcano with greatest SO2 output in each region is in bold text. The total mass burden detected by OMI over the specified survey duration is shown too. A comparison between mean annual SO2 mass burdens calculated from OMI data and those extrapolated from ground-based data available in the literature is shown, with the consistent OMI underestimate emphasized.

<table>
<thead>
<tr>
<th>Region</th>
<th>Active volcanoes</th>
<th>Total SO2 mass (× 10^9 kg)</th>
<th>Duration of OMI survey</th>
<th>Mean annual SO2 mass (× 10^6 kg)</th>
<th>OMI</th>
<th>DOAS</th>
<th>OMI/DOAS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ecuador and southern</td>
<td>Galeras, Reventador, Tungurahua</td>
<td>c. 1.2</td>
<td>September 2004 to</td>
<td>234* 532–1272 † 0.44–0.18</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Colombia</td>
<td></td>
<td></td>
<td>September 2006</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Vanuatu</td>
<td>Vanuavala, Gaua, Ambae, Ambrnym, Lopevi, Yasur</td>
<td>c. 4.3</td>
<td>September 2004 to August 2011</td>
<td>614‡ 3100‡ 0.20</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Papua New Guinea</td>
<td>Manam, Langila, Ulawun, Rabaul, Bagana</td>
<td>c. 1.8</td>
<td>January 2005 to December 2008</td>
<td>349‡ 1160§ 0.30</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

References for data:
eruptive and quiescent phases at Soufrière Hills Volcano, Montserrat, were described by Hayer et al. (2010), and ascribed to variations in plume height rather than changing magma supply. OMI detected increased SO2 emissions two months prior to the opening of the Halema’uma’u vent on Kilauea in March 2008 (Carn et al. 2013). Variations in OMI-measured SO2 burdens over Nyiragongo were used to infer seasonal changes in tropospheric plume dispersion, as well as the volcano’s output (Carn et al. 2013).

**Comparison of OMI with ground-based data**

Despite these successes, there is a major limitation facing the application of OMI to long-term volcanic monitoring, namely the lack of robust validation of its tropospheric SO2 dataset. In general, good agreement has not been achieved between OMI estimates of SO2 degassing and those produced by ground-based DOAS surveys, the most widely used technique in SO2 emission monitoring. A comparison of annual budgets calculated by summing OMI daily mass burdens versus those extrapolated from DOAS average fluxes produced very different annual estimates for Papua New Guinea, Vanuatu and Ecuador (McCormick et al. 2012), with OMI estimates around 20–40% of those derived from DOAS measurements (Table 3).

Comparison of satellite and ground-based retrievals of SO2 degassing is difficult for a number of reasons. Measurements are made over very different spatial scales with consequently different sampling of plume heterogeneity. The detection limits of different sensors may not be comparable (Table 1). Surveys are rarely completed simultaneously, with OMI tending to observe relatively mature SO2 plumes, and much of the published DOAS literature refers to short-lived campaigns, rather than continuous monitoring (although initiatives such as the Network for Observation of Volcanic and Atmospheric Change may provide longer-duration datasets; Galle et al. 2010). A recent OMI validation attempt by Carn et al. (2011) demonstrated some correlation between OMI SO2 column concentrations and those measured during an aircraft-based campaign over Tungurahua Volcano, Ecuador, but the daily mass burdens measured were close to OMI’s detection limit throughout the survey.

More effective comparisons between ground- and satellite-based datasets may be achievable if the satellite data is used to derive SO2 emission fluxes. Fluxes may be calculated by (1) dividing total measured SO2 mass burdens by an estimate of SO2 lifetime, (2) integrating SO2 column densities along a perpendicular transect of the plume and multiplying by wind speed, or (3) multiplying wind speed and measured mass burden for a single pixel, and dividing by the pixel dimension parallel to wind direction (see Carn et al. 2013 for further discussion). Derived fluxes from OMI data and airborne COSPEC measured fluxes of SO2 emissions during the effusive phase of the 2009 Redoubt eruption (Alaska) were compared by Lopez et al. (in press), who found that the best correlation was achieved when fluxes were calculated by multiplying the total measured plume SO2 burden by wind speed and dividing by plume length. Lopez et al. (in press) also demonstrated that good linear agreement between near-simultaneous OMI and COSPEC column densities could be achieved, provided a correction was made for the different spatial resolution of the datasets. Key sources of error in the plume transect flux method (chiefly wind speed and plume height) and preliminary flux detection limits for good observation conditions were found by Campion et al. (2012). Good correlation was also demonstrated between OMI- and ASTER-derived fluxes and those measured by ground-based UV camera at Turrialba Volcano, Costa Rica (Campion et al. 2012). We note that satellite-derived estimates of SO2 emission fluxes have also been demonstrated from MODIS over Bezymianny (Kearney et al. 2008) and Etna (Merucci et al. 2011), and ASTER over Etna (Campion et al. 2010).

Significant progress in the use of OMI data for monitoring volcanic SO2 emissions hinges on studies like these, which combine multiple techniques and datasets. While multi-sensor approaches are increasingly commonplace in the tracking of drifting SO2 clouds in the UTLS, (e.g. Carn et al. 2009a; Thomas et al. 2009; Carn & Prata 2010), very few such comparisons of tropospheric or continuous degassing have been published, despite their potential. Both GOME-2 and IASI can detect passive degassing under certain conditions (Rix et al. 2009; Carboni et al. 2012). Concerted efforts to combine multiple satellite datasets, and where available ground-based DOAS, COSPEC and UV camera datasets, will provide much more insight into degassing than any single technique in isolation. While better correlation may be possible between satellite-derived and measured ground-based flux datasets, however, calculating flux from OMI requires highly specific conditions: plumes largely un-obscured by cloud, of a certain geometry that enables the plotting of multiple transects, and good constraint on local wind speed or SO2 loss rates. Comparatively small uncertainties in wind speed may lead to significant errors in calculated flux (although this is true of ground-based flux datasets too). Loss rates vary widely over orders of magnitude and are difficult to constrain accurately (see the section ‘Persistence or transience of SO2 clouds’). The comparison of satellite mass burdens...
and ground-based fluxes may produce different estimates of annual \( \text{SO}_2 \) budget, and this certainly requires further investigation, but their comparison over longer time periods generally shows correlation in degassing trends. This latter method is more robust, being useable (with acknowledgement of certain sources of interference) over larger durations and involving largely automatic, non-interactive processing.

**Limitations of OMI’s application to volcano monitoring**

We have demonstrated above that, while the use of OMI to track large drifting \( \text{SO}_2 \) clouds in the UTLS is well established, with clear future strategy and objectives in place, the applicability of OMI to long-term monitoring of volcanic degassing is somewhat more complex. While significant progress has been achieved, there remain a number of uncertainties and limitations, which we evaluate here with the aim of road-mapping future research in this field. Successful use of OMI for monitoring purposes requires three conditions to be satisfied:

1. The volcanoes under observation must emit enough \( \text{SO}_2 \) to exceed OMI’s detection limit, which, as we shall discuss, shows significant variability.
2. The \( \text{SO}_2 \) emissions must be reasonably persistent in the atmosphere over the volcano, that is, not rapidly removed by atmospheric chemical reactions or dispersion processes prior to satellite overpass.
3. Local factors which can interfere with the OMI retrievals, such as meteorological cloud cover, high plume ash content, high total ozone column amounts and the row anomaly must be acknowledged and accounted for (Table 4).

**Detectable levels of \( \text{SO}_2 \)**

OMI’s detection limit is the minimum mass of an \( \text{SO}_2 \) cloud which can be distinguished robustly from background noise. Our working definition is a cluster of five nadir pixels, each with \( \text{SO}_2 \) mass greater than \( 5\sigma \), where \( \sigma \) is the standard deviation of total scene noise (see Carn et al. 2013, for further discussion). The detection limit is not fixed, since noise levels vary with \( \text{SO}_2 \) cloud altitude, latitude and season (Fig. 5). Detection limits increase with decreasing \( \text{SO}_2 \) plume altitude, owing to the increased path length travelled by solar photons; along such paths the potential for scattering and absorption by non-\( \text{SO}_2 \) atmospheric constituents is increased. With increasing latitude, lower levels of solar insolation as well as the higher solar zenith angles both reduce signal-to-noise and increase detection limit. Seasonal sunlight variations are also significant: detection limits are much higher in winter than in summer. Therefore certain volcanic regions are likely to be better potential targets for OMI surveys than others. For

<table>
<thead>
<tr>
<th>Source of interference</th>
<th>Impact on OMI data</th>
<th>Solutions/mitigation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Meteorological cloud cover</td>
<td>Thick cloud overlying volcanic ( \text{SO}_2 ) plumes will shield them from incoming UV</td>
<td>Use OMI reflectivity data for broad overview of cloud cover</td>
</tr>
<tr>
<td></td>
<td>Highly reflective clouds beneath the plume may enhance signal by reflecting back more UV</td>
<td>Use higher spatial resolution satellite data (e.g. MODIS) to produce cloud maps and investigate coverage of OMI pixels</td>
</tr>
<tr>
<td>High plume ash content</td>
<td>Increased scattering/absorption of UV radiation by ash particles</td>
<td>Assess co-location of ash and ( \text{SO}_2 ) using OMI aerosol index dataset (qualitative use only)</td>
</tr>
<tr>
<td></td>
<td>Increased scavenging and chemical processing, using particles as reaction surfaces</td>
<td>More detailed ash analysis with other remote sensing data, for example IASI, MSG-SEVIRI, CALIPSO</td>
</tr>
<tr>
<td>Row anomaly</td>
<td>Data degradation from several detector rows in OMI CCD</td>
<td>Interpolate across missing rows from data either side</td>
</tr>
<tr>
<td></td>
<td>Dynamically evolving, has both worsened and improved with time during different periods</td>
<td>Fill missing values using other A-Train ( \text{SO}_2 ) data (e.g. AIRS for large UTLS eruptions)</td>
</tr>
<tr>
<td>High total ozone column</td>
<td>Large ozone signal saturates retrieval blocking tropospheric ( \text{SO}_2 ) signal</td>
<td>Sophisticated retrieval algorithms needed. Impact will be greatest at high latitudes, and on tropospheric ( \text{SO}_2 ) retrievals</td>
</tr>
</tbody>
</table>
example, low-latitude regions such as Papua New Guinea, Indonesia and the Caribbean have consistently lower detection limits than mid- or high-latitude regions such as Alaska, Kamchatka and Iceland; these latter regions suffer significant degradation of detection limit in winter, while low-latitude regions are expected to show consistent detection limits year-round. High latitude regions are also affected by higher total ozone column concentrations (see the section ‘Ozone’).

Equally important to a changing instrument detection limit is the issue that not all volcanoes release enough SO2 to exceed it, and therefore may not be good targets for satellite-based remote sensing. Volcanoes exhibit a wide range of degassing behaviours, with variations in the manner and amount of SO2 release frequently occurring on rapid timescales. While certain volcanoes such as Ambrym, Bagana and Etna are persistently strong emitters of SO2 (Bani et al. 2012; McCormick et al. 2012; Aiuppa et al. 2008) and hence are readily observed by OMI, many other volcanoes may not be suitable targets for satellite-based monitoring simply owing to their low SO2 output. In some cases, these low SO2 emission fluxes may be attributable to low magmatic sulphur content, or a low volume of degassing magma (Wallace & Edmonds 2011). Alternatively, quiescent or mildly active volcanoes may have very low SO2 fluxes owing to their low intensity of activity, which is controlled again by low volumes of magma at shallow depths in the crust. For example, during the OMI survey of Ecuador and southern Colombia by Carn et al. (2008), no emissions were detected from the mildly active volcano Sangay. In other cases, hydrothermal scrubbing of emitted volcanic gases may reduce the amount of SO2 released into the atmosphere (Symonds et al. 2001). Passively degassing volcanoes may also be poor targets if their plume is small and typically trapped in the boundary layer. OMI detects SO2 at Soufrière Hills much more frequently during extrusive phases, when the volcanic plume is thermally lofted into the free troposphere, than during eruptive pauses (Hayer et al. 2010). In cases such as this, where a volcano’s behaviour operates in cycles, it is anticipated that OMI’s detection potential will vary accordingly. In Papua New Guinea, SO2 emissions from Langila and Ulawun were much more readily detected during eruptive events than during passive degassing phases (McCormick et al. 2012). In other cases of continuous degassing, magma composition and its control on degassing style may limit the potential for detection. For example, basaltic systems where open-conduit degassing predominates may release SO2 more continuously and at higher fluxes than volcanoes erupting more evolved compositions, where a summit lava dome may modulate or prohibit continuous degassing.

Volcanoes which have SO2 emission fluxes below OMI’s detection limit may still be detectable with the use of time-averaging. Monthly (or longer duration) mean SO2 burden maps (Fig. 4) are useful aids to assessing long-term variability in degassing over regional scales not least because signal-to-noise is increased and volcanic SO2 can be more readily distinguished from background (Carn et al. 2008, 2013; McCormick et al. 2012). Systematic biases in the dataset may also be amplified however, and the loss of daily temporal resolution will limit the effectiveness of OMI data in revealing the true nature of variations in degassing at individual volcanoes.
Persistence or transience of $SO_2$ clouds

The persistence or transience of volcanic $SO_2$ over the source volcano is controlled by a combination of chemical and cloud processing of the $SO_2$, and its physical dispersion by wind. Together these can act to reduce the mass of $SO_2$ available for detection by OMI at the time of overpass, and hence lead to underestimates of the volcanic source’s strength. Conversely, minimal removal and dispersion could lead to overestimates, if relict $SO_2$ from previous days persists over the volcano and is measured during subsequent OMI overpasses.

Chemical processing of $SO_2$ is dominated by in-plume oxidation to sulphate aerosol, which cannot be effectively detected by UV remote sensing (although OMI’s aerosol index data may be able to reveal the location of large aerosol loadings). Oxidation occurs via gas-phase homogeneous reactions, aqueous-phase reactions or heterogeneous reactions on the surface of solids, with the dominant process depending strongly on the surrounding atmospheric conditions (Eatough et al. 1994). Gas-phase homogeneous processes are thought to be dominated by the presence of the hydroxyl radical (Faloona 2009), and may range in speed from 1 to 10% $SO_2$ converted per hour, with high temperature and relative humidity promoting more rapid conversion and cold conditions leading to very slow conversion (Eatough et al. 1994). Volcanic plumes erupted in lower latitudes may therefore be expected to experience more extensive and rapid conversion of $SO_2$ than those erupted at higher latitudes. Aqueous-phase reactions can be considerably faster, up to 100% conversion per hour under optimum conditions, such as elevated levels of ozone and hydrogen peroxide (Eatough et al. 1994). Under cloudy conditions, aqueous processing by $H_2O_2$ (non-precipitating clouds) and $O_3$ (precipitating clouds) may dominate oxidation pathways over gas-phase oxidation (Seigneur & Saxena 1988). Heterogeneous reactions are thought to be generally less significant unless in the presence of high concentrations of particulate matter, such as ash, aerosol, sea salt or mineral dust, many of which are most concentrated in the boundary layer (Li et al. 2006; Harris et al. 2012). The sulphate aerosol produced is removed from the troposphere (in a matter of hours to days) by wet/dry deposition, processes which also affect $SO_2$ mass and vary widely with factors such as seasonal weather variability, the availability and properties of deposition surfaces and rainfall or fog properties (Delmelle 2003; Langmann et al. 2009).

Assessing plume transience therefore requires knowledge of these processes and the resulting $SO_2$ lifetime in a given plume environment. Several studies have investigated $SO_2$ loss rate in tropospheric volcanic plumes, and their estimates span several orders of magnitude, from $10^{-3}$ to $10^{-6}$ s$^{-1}$ (Oppenheimer et al. 1998; McGonigle et al. 2004b; Rodríguez et al. 2008; Nadeau & Williams-Jones 2009; Carn et al. 2011). Although certain basic principles are maintained (loss appears as a rule to be faster in warm and humid low latitude conditions), $SO_2$ lifetime apparently varies widely, based on local atmospheric and meteorological conditions, as well as the measurement strategy used. The loss rates calculated following any single campaign may only be representative of the extant volcanic and atmospheric conditions.

While long-term field campaigns sampling a range of volcanic and atmospheric conditions could provide a more rigorous study, this is logistically difficult and the measurements required are complex. An alternative method is the use of atmospheric chemistry/transport models that simulate changing meteorological conditions and assess the resulting impact on $SO_2$ processing and removal. Recent and continuing work simulating volcanic plume dispersal and the fate of $SO_2$ on both global (e.g. Stevenson et al. 2003) and regional scales, in Nicaragua (Langmann et al. 2009), Indonesia (Pfeffer et al. 2006) and Ecuador (McCormick et al., in preparation) shows considerable promise. Good agreement, for example, has been achieved between model output and field estimates of sulphate deposition at Masaya (Langmann et al. 2009). However, owing to their computational requirements, these studies are not suitable for real-time continuous monitoring applications. In these instances, a useful first-order assessment of whether a volcano’s plumes are likely to be mainly transient or persistent can be made by calculating the maximum age attained by the plume before its $SO_2$ mass falls below OMI’s detection limit. By comparing measured plume length in OMI data with estimates of wind velocity, this age can be calculated, and provides crude constraint on $SO_2$ loss rates. This method was used successfully by McCormick et al. (2012) to explain why OMI was likely to underestimate $SO_2$ emissions from the volcanoes of Papua New Guinea. The availability of accurate wind data at plume altitude is a potential limiting factor, however.

Wind dispersal of plumes can also pose significant problems for volcano monitoring with OMI. High-velocity winds may blow $SO_2$ clouds considerable distances, either downwind of their source volcano, which may complicate interpretation of data in regions with multiple sources, or towards the edge of OMI’s swath, where spatial resolution is much coarser, affecting accurate mapping of the plume’s location and precluding detection (Carn et al. 2013). Horizontal wind velocity at vent altitude at the time of emission also exerts a
strong control on plume transport, which was demonstrated by Kinoshita (1996) at Sakurajima Volcano, Japan. Low-velocity winds (<5 m s\(^{-1}\)) resulted in plumes rising vertically above the vent to an equilibrium altitude before drifting horizontally. Medium velocity winds (5–10 m s\(^{-1}\)) led to plumes rising at an angle towards the same altitude. Under high velocity winds (>10 m s\(^{-1}\)), plumes barely rose, but were instead blown laterally away from the vent, or even as a mountain lee wave down the volcanic edifice. Lateral dispersion was also linked to wind velocity, particularly in the plume’s range of vertical extension. High-velocity winds were more likely to result in linear advection of the plume, in the direction of the wind and with little sideways spread. Fan- or belt-type dispersal patterns were more common under low-velocity winds. The impact of wind velocity on plume dispersion therefore seems to depend on the vertical profile of wind velocity, and specifically the relative horizontal wind speed of the plume centre of mass and ambient air (Freitas et al. 2010). At vent altitude, a high wind velocity could effectively trap a plume in the boundary layer, greatly reducing the potential for OMI’s accurate detection of it. At higher altitudes, however, once a plume has risen to equilibrium altitude, higher winds may be more favourable since the extensive spread and lateral dispersion of a fan-type pattern would shear and thin the plume, reducing the effective thickness and SO\(_2\) column amounts, again limiting OMI detection.

Sub-optimal observation conditions/general interference with OMI

Although detectable levels of SO\(_2\) which persist over the source volcano should be observable by OMI, certain local factors may still reduce or impede successful retrieval. The most significant of these are meteorological cloud cover, volcanic ash in the plume, high ozone columns and the row anomaly (Table 4).

Meteorological clouds. The impact of clouds on the OMI SO\(_2\) retrieval depends strongly on cloud optical thickness, the extent and continuity of cloud cover, and the altitude of the clouds relative to volcanic SO\(_2\) clouds (Krotkov et al. 2006). Thick cumulus and cumulonimbus shields with highly reflective upper surfaces may greatly reduce the penetration of incoming solar radiation into the lower atmosphere. This is particularly significant in satellite observation of tropospheric and boundary layer SO\(_2\) clouds, since meteorological cloud may be widespread throughout the entire troposphere. However, if volcanic clouds overlie thick weather cloud shields, then OMI’s sensitivity to the SO\(_2\) present will be increased as more radiation is reflected back rather than passing further into the lower atmosphere. Thin, broken cirrus clouds at higher altitudes are not expected to interfere greatly with the OMI retrieval owing to their relatively thin optical depth and their generally discontinuous nature. Cloud cover and type, and therefore its impact on the OMI retrieval, are highly variable. Over much of the Earth, the amount and type of cloud cover has a strong seasonal cycle and is also highly dependent on latitude (ISCCP 2012). However, in some regions other processes dominate variability, such as the El Niño frequency in eastern India and the western-central Pacific (Tselioudis & Rossow 2011). Since OMI’s local equatorial crossing time is constant, there should not be any significant effect from the diurnal cycle near the equator, but at higher latitudes this effect may become more pronounced.

Mitigation for cloud cover is currently limited to first-order estimates of the extent of cloud cover and altitude, using OMI’s effective reflectivity and cloud pressure data. These parameters provide a good overview of the potential severity of interference. More detailed analysis of cloud interference in future could focus on the variation in the sub-pixel area coverage by cloud over an OMI scene and assess potential impact on the SO\(_2\) retrieval. The near-simultaneous overpasses of OMI on Aura and MODIS on Aqua (another A-train satellite) would facilitate such a study.

Interference from volcanic ash. Interference by volcanic ash is a well-known problem in UV remote sensing measurements of volcanic SO\(_2\) emissions (e.g. Williams-Jones et al. 2008; Galle et al. 2010; Kern et al. 2010a, b). Increased scattering of light outside the SO\(_2\) plume effectively dilutes the measured column density. However, high concentrations of ash and other scattering aerosols within the plume artificially elongate the effective path length travelled by photons prior to their return to the detector and thus enhance SO\(_2\) column densities (Kern et al. 2010a, b). Whether scattering or absorption dominates is a function of the UV optical properties (especially single scattering albedo) of the ash. Errors of c. 30% are expected in typical volcanic plumes owing to these radiative effects, with greater errors possible under very high SO\(_2\) or ash loadings (Kern et al. 2010b, 2012). Additionally, high ash content in the plume can result in increased scavenging of SO\(_2\), since ash particles act as reaction and adsorption surfaces, leading to reduced SO\(_2\) mass by the time of satellite overpass (e.g. Witham et al. 2005; Delmelle et al. 2007).

Certain volcanic scenarios will result in significantly more ash-rich plumes than others, and are hence more prone to potential interference. For
example, explosive eruptions release considerably more ash than passive degassing; this is one factor that has tended to limit the application of ground- or air-based UV remote sensing techniques to less explosive degassing (Williams-Jones et al. 2008; Lopez et al. in press). Predicting radiative impact also depends on knowing ash particle size distributions, composition and shape, which determine the optical properties of the ash (e.g. Witham et al. 2005). Explosive silicic eruptions tend to produce relatively more fine ash than basaltic ones, owing to more extensive fragmentation resulting from a higher pre-eruptive magma viscosity and volatile content (Alidibirov & Dingwell 1996; Rose & Durant 2009). Eruptions with significant associated pyroclastic flows also produce ash that is generally finer-grained owing to comminution (milling) of grains (Rose & Durant 2009). Magma interactions with water may also produce both greater quantities of ash and a larger proportion of fine grains (e.g. Wohletz 1983); this was an important feature of the 2010 Eyjafjallajökull eruptions (e.g. Ilyinskaya et al. 2011). Another limiting factor in the potential extent of ash interference is whether or not ash and SO₂ plumes are co-located. Shortly following eruption, this is likely to be true, but as the plume ages, vertical stratification and differential responses by ash and SO₂ to wind shear begin to take effect, and the two plumes may separate (e.g. Thomas & Prata 2011; Walker et al. 2012). Variation in eruptive style may also lead to the two plumes having different locations (Thomas & Prata 2011). Plume ageing will tend to result in diminished ash interference regardless, owing to relatively rapid sedimentation of even fine ash (<63 μm) and aggregation processes (Rose & Durant 2009, 2011; Taddeucci et al. 2011; Brown et al. 2012). Nonetheless, since OMI and other satellite-based sensors aim to report accurate SO₂ mass burdens from their first observation of a plume for aviation hazard applications, it is essential to know if ash is present, and in what quantity.

The location of ash relative to the volcanic SO₂ plume is accomplished using auxiliary information, either OMI’s Aerosol Index (or AI, which is sensitive to ash as well as to sulphate aerosol) or the use of combined satellite datasets (Thomas & Prata 2011; Bonadonna et al. 2011). OMI’s AI is presently not validated, and so is best utilized to locate the presence of ash rather than provide detailed information on ash content and size distributions. The use of other satellite datasets, however, is far more established and tested; the recent eruptions in Iceland of Eyjafjallajökull and Grimsvötn, for example, allowed demonstrations of ash detection and characterization by IASI (Tolodano et al. 2012), the Advanced Along Track Scanning Radiometer (Grainger et al. 2013) and MSG-SEVIRI (Bonadonna et al. 2011; Francis et al. 2012). Sampling of ashfall deposits where possible provides further useful insight into both particle size distribution and atmospheric transport (Dellino et al. 2012).

**Ozone.** High total ozone column amounts can interfere with OMI’s SO₂ retrieval in the lower and mid troposphere since both SO₂ and ozone have strong absorbance at similar regions in the UV spectrum (Krotkov et al. 2006). Total ozone column varies with latitude, altitude and season (Fig. 6; Krueger 1989; Fishman et al. 2003; Burrows et al. 1999; Ziemke et al. 2006). The impact of ozone will be greatest at high latitudes, and will essentially lead to a masking of the contribution of SO₂ in the top-of-atmosphere irradiance signal detected by OMI. For low and mid tropospheric cases, the detection limit will be increased, and only strong sources may be detected. Lopez et al. (in press) showed that OMI could detect degassing associated with effusive activity at Redoubt Volcano, Alaska, and proposed an SO₂ detection limit of 2–4 × 10⁶ kg in high-latitude spring conditions. Persistent SO₂ release from the Norilsk industrial region in northern Siberia has also been frequently detected by OMI (NASA 2008) and GOME (Kholkar et al. 2008). These encouraging results suggest that, while high ozone columns may reduce OMI’s detection of SO₂, significant sources will still be observable.

**The row anomaly.** The so-called ‘row anomaly’ has been a serious source of interference in OMI data since 2008, and comprises bands of anomalously elevated noise in the along-track direction of OMI scenes. Thought to be a consequence of a blockage in the instrument’s field of view, the row anomaly is often of similar or greater magnitude to volcanic SO₂ in the scene, and hence can impede the detection of real plumes (http://www.kmi.nl/omi/research/product/rowanomaly-background.php; Carn et al. 2013).

**Case studies of regional monitoring with OMI.**

The preceding discussion indicated that good conditions for long-term monitoring with OMI might comprise a reasonably high flux SO₂ source (>10³–10⁴ kg day⁻¹), ideally in low to mid latitudes where detection limits are lower. Volcanoes at higher latitudes have to be bigger SO₂ sources in order to be detected above noise, although their emissions are less likely to be affected by rapid loss relative to those of low-latitude volcanoes. Local meteorology affects all volcanoes via the potential for chemical processing, plume dispersion and the extent of cloud cover. Volcanic activity also
plays a role, not just in the amount of emitted SO₂, but also in the style of release, injection altitude and the presence of ash.

Here we illustrate the interplay of these various factors with short case studies of degassing from four regions with multiple active volcanoes, exhibiting a range of activity types and levels, and lying across a range of latitudes.

**Mexico**

Two volcanoes are currently active in Mexico, Popocatépetl (5426 m above sea level, a.s.l.) and Colima (3850 m a.s.l.), with annual SO₂ emissions of 1871 and 735 × 10⁶ kg, respectively, reported for the year 1999 (INE-SEMARNAT 2006). More recently measured SO₂ fluxes were 2.45 ± 1.39 × 10⁶ kg day⁻¹ at Popocatépetl during passive degassing in March 2006 (Grutter et al. 2008), and a peak value of 2.7 × 10⁶ kg day⁻¹ at Colima during the 2004 extrusive lava eruption (Zobin et al. 2008). Outside of eruptions, Colima’s SO₂ flux is considerably lower.

We examined OMI monthly mean data for six months of 2007, and noted persistent strong SO₂ emissions from Popocatépetl and only weak sporadic emissions from Colima (Fig. 7). A new episode of lava dome growth began at Colima in February 2007 (Smithsonian Institution 2008b; Hutchison et al. 2013). No SO₂ was detected by OMI in January, but small amounts were detected in March and May (Fig. 7). Effusion continued with a low mean rate (0.01–0.03 m³ s⁻¹) until October, before increasing to 0.033 m³ s⁻¹ in November (Smithsonian Institution 2008b). A possible SO₂ plume appears in the OMI scene for November, but it is weak, incoherent and hard to unequivocally distinguish from background noise. We contend that the sporadic detection of Colima’s emissions by OMI is principally a result of the low activity during 2007.

The extensive SO₂ plumes close to Popocatépetl are complemented by persistent emissions from the Tula industrial complex (Fig. 7). De Foy et al. (2009) combined ground and airborne COSPEC and DOAS measurements with numerical simulations to propose that, during March 2006, SO₂ emitted from Popocatépetl accounted for just c. 10% of the total local burden, with the remainder emitted from urban sources in Mexico City and the Tula complex 70 km NW. OMI data, however, clearly show much stronger plumes over the volcano than for the industrial complex; this probably reflects the contrast in SO₂ injection altitude. Popocatépetl’s vent is c. 5400 m, while the Tula emissions are likely to be trapped in the boundary layer (De Foy et al. 2009). Inspection of daily images from 2007 supports this, with Tula’s
Fig. 7. SO$_2$ monthly mean plots for Mexico, from 2007. Active volcanoes are marked with an open triangle; the Tula industrial plant with an open diamond.
emissions being detected less frequently than those from the volcano. Popocatepetl’s plumes are more laterally extensive than those from Tula, perhaps owing to increased SO2 lifetime and stronger winds aloft in the free troposphere relative to the boundary layer. The steady-state passive degassing behaviour of Popocatépetl is reflected by the plume appearance (Fig. 7) with maximum SO2 concentrations over the vent and decreasing downwind. Mean daily burdens over Popocatepetl, with the contribution of Tula’s emissions removed, range from $170 \times 10^3$ to $620 \times 10^3$ kg; this is an order of magnitude lower than the daily flux reported by Grutter et al. (2008) and may reflect the relatively modest activity at the volcano during 2007, short SO2 lifetimes in the troposphere or other factors causing the underestimation of tropospheric SO2 emissions by OMI (discussed in the sections ‘Long-term monitoring of degassing with OMI’ and ‘Limitations of OMI’s application to volcano monitoring’).

**Italy**

Italy hosts three volcanoes with active, well-monitored, SO2 degassing. Etna (3330 m a.s.l.) is a persistently degassing, frequently erupting basaltic stratovolcano. A total volatile flux of $21 \times 10^6$ kg day$^{-1}$ is reported during both quiescent and eruptive phases; SO2 contributes c. 8%
Stromboli’s (924 m a.s.l.) degassing comprises continuous quiescent emission from a main open conduit punctuated by explosive degassing of deeper-sourced gas slugs, together with shallow hydrothermal emissions (Burton et al. 2007). Most SO2 degassing is via open conduit emissions totaling \( c. 200 \times 10^3 \) kg day\(^{-1}\), with fluxes of \( c. 620 \times 10^3 \) kg day\(^{-1}\) reported during the 2007 effusive eruption (Burton et al. 2007, 2009). Vulcano (500 m a.s.l.) is a closed conduit system, with fumarolic SO2 degassing. Fluxes of \( 12 \times 10^3 \) kg day\(^{-1}\) are typical, although \( 100 \times 10^3 \) kg day\(^{-1}\) was recorded during an unusual event in late 2009 (Vita et al. 2012).

A time series of daily SO2 mass burden measured by OMI over Italy during 2006–2007 is dominated by volcanic emissions (Fig. 8). During this period, Vulcano maintained a quiescent degassing state, Stromboli experienced a \( c. 40 \) day period of effusive activity in spring 2007 and Etna experienced two lengthy phases of effusive and Strombolian activity during the latter half of 2006, a few short-lived eruptive events in early 2007 and otherwise passive degassing (Burton et al. 2009; Aiuppa et al. 2008). OMI monthly mean SO2 burden maps from this interval only record plumes from Etna. Vulcano’s emissions are below detection limit, and although Stromboli’s persistent SO2 flux may just exceed detection limits in the lower troposphere, the plume may often be confined to the boundary layer (the volcano is \( <1000 \) m in height). During Stromboli’s eruption in early 2007, plumes are detected from the volcano in some daily OMI scenes, but they are comparable in mass to background noise.

The trends in SO2 burden shown in Figure 8 are therefore dominated almost entirely by Etna, which reflects the fact that it comprises \( c. 90\% \) of Italy’s total volcanic SO2 budget (Aiuppa et al. 2008; Burton et al. 2007, 2009; Vita et al. 2012). Comparing the OMI record to activity reports from the volcano, we see strong correlation. The duration of the July 2006 effusive eruption at Etna is a clear peak above the very low background. Overall increased SO2 mass burdens are reported through the second phase of summit eruptions, between September and December, with some peaks in the OMI record matching reported incidences of higher activity, such as the explosive pyroclastic
flow-forming eruption on 16 November (Behncke et al. 2008) and the intense fire fountaining and lava effusion in late December (Smithsonian Institution 2007a). The decrease in SO2 output from Etna during the passive degassing that followed the eruptions was clear, as were the peaks in SO2 release associated with several short-lived eruptions that occurred throughout spring 2007. Also at this time, small peaks above the background that coincided with the effusive eruption at Stromboli were noted. The mass burdens measured by OMI outside of eruptive phases throughout Figure 8 are very low (≪100 × 10^3 kg on most days), significantly lower than Etna’s non-eruptive/passive degassing flux (Aiuppa et al. 2008). This apparent underestimate is surprising, since Etna is one of the largest continuous SO2 point sources on Earth. A more detailed study of OMI observations of degassing from Etna is required to investigate this further.

Overall, OMI works as expected over Italy: while it is unable to resolve the relatively low fluxes of the two small sources of Vulcano and Stromboli, consistent detection of the plume from Etna is achieved. For passive degassing, we note that time-averaging is required to clearly resolve the plume, and also that OMI underestimates the SO2 emissions of Etna during these phases. However, this short study clearly demonstrates that OMI is sensitive to changes in activity and hence degassing regime at Etna and that the volcano represents a suitable target for future investigation with OMI data. This is particularly encouraging owing to the availability of high-quality ground-based records of degassing and also the increasingly successful application of other satellite instruments in studying SO2 emissions from Etna, such as MSG-SEVIRI (Corradini et al. 2009), ASTER (Campion et al. 2010) and MODIS (Merucci et al. 2011).

Kamchatka

The Kamchatka peninsula in Siberia has many active volcanoes, but owing to the difficulty of regular access to the region, measurements of SO2 emissions are scarce, existing only from Karymsky (1536 m a.s.l.), Mutnovsky (2322 m a.s.l.) and Bezymianny (2882 m a.s.l.) volcanoes (Fischer et al. 2002; Taran 2009). Estimates of SO2 release at other volcanoes have been made from comparison of relative plume strengths, as well as total heat flux, and an arc-scale total SO2 budget of c. 2000 × 10^6 kg day⁻¹ has been proposed (Taran 2009). The dome-building volcanoes of the northern group (Klyuchevskoy (4835 m a.s.l.), Bezymianny (2882 m a.s.l.) and Shiveluch (3283 m a.s.l.)) are thought to contribute most to this, a suggestion supported by OMI data presented by Taran (2009). Between May and October 2007, OMI observed mean daily burdens of c. 1200 ± 1000 t SO2 over Kamchatka. Plumes from passively degassing volcanoes such as Mutnovsky and Karymsky to the south were not observed, which was ascribed to OMI’s relatively coarse spatial resolution (Taran 2009).

We studied OMI monthly mean data from 2006 and generally found considerable inconsistency in the dataset. Winter months are characterized by high background noise, which makes unequivocal distinction of volcanic SO2 plumes very difficult. In January, February, November and December, the low sunlight conditions typical of high-latitude winters result in particularly high noise and render identification of volcanic emissions all but impossible (Fig. 9a). This is compounded by widespread cloud cover over the peninsula as indicated in the OMI reflectivity data (Fig. 9a); note that this signal will also contain a contribution from winter snow cover. This result does confirm the estimated impact of high-latitude winter conditions on OMI’s detection limit, discussed in the section ‘Published surveys of degassing’ and by Carn et al. (2013). However, in March, May and particularly August, there is a clearer sense of SO2 degassing from Kamchatka’s volcanoes (Fig. 9b). We base this assertion on pixels close to the volcanoes with notably higher SO2 mass than the surrounding noise. In common with Taran (2009), we report OMI detection of emissions from the northern group of volcanoes. The very close proximity of Bezymianny and Klyuchevskoy makes it difficult to distinguish the source volcano for the plumes seen in these four months; based on activity reports, however, we propose Bezymianny as a more likely source since Klyuchevskoy experienced an eruptive pause throughout 2006 (Smithsonian Institution 2007b). With the possible exception of June, no clear emission is observed from Shiveluch, despite reports of continued dome growth throughout 2006 (Smithsonian Institution 2006a, c). In contrast with Taran (2009), we do see SO2 emissions from the dominantly passive degassing volcanoes further south, Karymsky, Mutnovsky and possibly Avachinsky (2741 m a.s.l.). Karymsky experienced regular Strombolian activity in 2006, with regular reports of ash venting and thermal anomalies (Smithsonian Institution 2006a—c). No reports of activity for the other two are available, however, beyond isolated comments regarding fumarolic activity at Avachinsky (Smithsonian Institution 2010). We attribute our detection of emissions from these volcanoes to our month-long time-averaging approach. Maximum daily SO2 burdens summed across the entire region do not exceed c. 600 × 10^6 kg.

These results show a mixed forecast for OMI monitoring of Kamchatka’s volcanic activity.
While plumes are detected in several months, we acknowledge that these plumes, where observed, are rather weak, incoherent and generally of lower mass than expected from the arc-scale estimate of SO$_2$ degassing made by Taran (2009). Under the high noise conditions and extensive cloud cover of the winter months, we cannot distinguish any plumes clearly from the background noise. Any estimation of scene SO$_2$ burden is likely to contain significant erroneous contribution from this noise, and any genuine SO$_2$ signal is likely to be heavily suppressed, with the high ozone column concentration in the region (Fig. 6) also having an impact. These are clearly major limiting factors to continuous monitoring of volcanoes in Kamchatka, with OMI data realistically being of use during only the summer months when observation conditions are better. Nonetheless the remoteness of the region and general lack of gas emission monitoring of its volcanoes does make any data provided by OMI very valuable. We also note, encouragingly, that the volcanism reported during 2006 was relatively mild, yet OMI could still detect some SO$_2$ emissions: we propose that, during periods of elevated flux, OMI could prove more successful over Kamchatka.

Central American Volcanic Arc

The Central American Volcanic Arc (CAVA) is among the best studied in terms of SO$_2$ emissions, with flux estimates published for 13 persistent volcanic SO$_2$ sources (Mather et al. 2006). We studied OMI data over the CAVA from 2007, and noted apparent SO$_2$ emissions from several volcanoes. Masaya (635 m a.s.l.) in Nicaragua appeared to be the CAVA’s major SO$_2$ source with persistent strong plumes detected during 2007 (Fig. 10a–d). This may reflect the increase in SO$_2$ flux observed at Masaya (Witt et al. 2008) since the field campaign of Mather et al. (2006). Arenal (1657 m a.s.l.), Poás (2708 m a.s.l.) and Turrialba (3340 m a.s.l.) appear to also be significant emitters with strong SO$_2$ plumes detected over Costa Rica from April 2007 onwards (Fig. 10b–d). It is not always clear which of these volcanoes the SO$_2$ plumes observed by OMI originate from, although based on contemporary activity reports, we suggest that Arenal and especially Turrialba dominated. During 2007, Arenal was erupting lava with an increase from September (Smithsonian Institution 2007e), Poás experienced weak, low-temperature fumarolic degassing (Smithsonian Institution 2007e) and Turrialba experienced a significant increase in fumarolic degassing evidenced by the opening of two new fractures, temperature increases and significant chemical damage to surrounding vegetation (Smithsonian Institution 2007d, 2008a). OMI also observed SO$_2$ emissions from Santa María (3772 m a.s.l.; Fig. 10a), which we suggest is related to ongoing activity at the Santiaguito lava dome (Smithsonian Institution 2007f), while potential SO$_2$ emissions from Tacaná (4060 m a.s.l.; Fig. 10c) are harder to explain, owing to the absence of available activity reports. Possible plumes from Fuego (Fig. 10a–c) would agree with its ongoing open vent degassing during 2007 (Lyons et al. 2010), but these are hard to distinguish clearly from background noise.

Generally, inspection of Figure 10 reveals very weak plumes as typical. Mather et al. (2006) suggest a mean daily arc flux of 4.36 × 10$^6$ kg day$^{-1}$, for all volcanoes (excepting Turrialba, which has only recently started to degas significantly, e.g. Campion et al. 2012). The maximum daily burden for the region observed by OMI is only 1.1 × 10$^6$ kg day$^{-1}$ (in March 2007), and this total almost certainly includes a contribution from the relatively high background noise over the CAVA throughout 2007 (Fig. 10a–c); the noise is frequently of similar magnitude to the supposed volcanic SO$_2$ mass burdens. Either this represents OMI underestimating the arc-scale SO$_2$ budget by c. 75%, or a decrease in SO$_2$ flux from the CAVA since the measurements summarized by Mather et al. (2006).

The CAVA lies in similar latitudes (albeit in the Northern Hemisphere rather than the Southern) to Papua New Guinea, Vanuatu and Ecuador, three regions where, although OMI has shown considerable capability for detecting clear patterns of degassing both on a daily basis and in time-averaged monthly scenes, there has also been significant underestimate of total SO$_2$ output relative to budgets calculated from ground-based measurements (see the section ‘Long-term monitoring of degassing with OMI’). An underestimate for the CAVA has a number of potential explanations. Firstly, we note that our assumption of a mid troposphere plume altitude (see Carn et al. 2013) for the SO$_2$ retrievals plotted in Figure 10 will contribute to an underestimate since the real plume altitude is likely to be much lower, based on the volcano summit heights. Our use of the mid troposphere retrieval is justified, however, by the even higher noise in lower troposphere retrievals over the CAVA, where volcanic emissions cannot be distinguished from background. Secondly, OMI reflectivity data over the CAVA suggests that reasonably high cloud cover is typical, particularly what we interpret as orographic cloud over the more elevated parts of the region (Guatemalan and Costa Rican highlands). This cloud cover may be linked to the high background noise, although spatial correlation between the two is poor. While these data are not well-suited to a rigorous quantitative
comparison between two regions, we note that the average scene reflectivity over the CAVA is not significantly greater than in Papua New Guinea, or Ecuador, for example. We might also anticipate reasonably rapid chemical processing of SO\textsubscript{2} in the hot humid atmosphere over the CAVA.

Without more detailed investigation, the reasons for the discrepancy between our estimates of total daily SO\textsubscript{2} output from the CAVA and that of Mather \textit{et al.} (2006) remain uncertain. The scale of the OMI underestimate is comparable to that observed in Papua New Guinea, Vanuatu and Ecuador in previous studies (see McCormick \textit{et al.} 2012, and the section ‘Comparison of OMI with ground-based’). Significant uncertainty arises because the compilation of degassing fluxes in Mather \textit{et al.} (2006) covered the period 1997–2004 and so does not cover the period or our OMI observations. Diminished activity at several of the CAVA’s volcanoes since this compilation would clearly contribute to overall reductions in SO\textsubscript{2} degassing budget. However, SO\textsubscript{2} output at Masaya has increased (Witt \textit{et al.} 2008), as indicated by its status as the strongest source seen by OMI, despite a typical plume height of \textless1 km, as have emissions from Turrialba during 2007 (Smithsonian Institution 2007\textit{d}, 2008\textit{a}). The paucity of information relating to the majority of the other volcanoes in the arc cannot be taken as firm evidence for decreased activity. Awareness of variation in local meteorological effects is also important. A detailed study of cloud cover over the CAVA, together with an investigation of the high background noise evident in Figure 10, would make more robust comparisons with ground-based datasets possible. Extending the duration of the OMI survey should

\textbf{Fig. 10.} SO\textsubscript{2} three-monthly mean OMI plots for CAVA. Volcanoes are abbreviated as follows: Ta, Tacana; SMa, Santa Maria; F, Fuego; Pa, Pacaya (Guatemala); SA, Santa Ana; SMI, San Miguel (El Salvador); SC, San Cristobal; Te, Telica; M, Masaya; C, Concepcion (Nicaragua); A, Arenal; Po, Poas; Tu, Turrialba (Costa Rica).
enable comparison of periods where both high and low levels of volcanic activity are reported, and this could test whether activity is a primary control on successful use of the satellite dataset. A reliable source of independent information to verify the OMI data is essential.

Conclusions
The use of satellite-based instruments in volcano monitoring has progressed significantly in recent years, culminating today in an unprecedented range of instruments and techniques, which are being currently deployed with increasing success and high quality of data. OMI is among the most sensitive satellite-based instruments to atmospheric SO2, and hence presents and has presented unique opportunities for volcanologists, specifically in the study of degassing. This review has discussed two principal applications of OMI data in volcanology: the detection and tracking of large drifting SO2 clouds for aviation hazard mitigation, and the long-term monitoring of SO2 emissions and degassing patterns at individual volcanoes and on regional scales. The former is well established, and OMI is likely to continue as a reliable tool in this regard. The means to extend capability, towards estimating total SO2 release of volcanic eruptions, was discussed, with the principal message being a call for greater integration between OMI and other satellite datasets.

The use of OMI for volcano monitoring and quantification of persistent tropospheric degassing shows promise, yet still faces a number of uncertainties, not least in how to most effectively integrate OMI data with other high-resolution data streams, both satellite- and ground-based. We have defined several criteria that must be fulfilled for OMI to successfully assess volcanic degassing. The SO2 emission of a volcano under observation must exceed OMI’s detection limit, which varies with latitude, altitude and season. Furthermore, differences in style and intensity of activity can render certain volcanoes detectable only at certain times. OMI’s detection of SO2 emissions is greatly helped if tropospheric chemistry and plume dispersal are not too extensive; both factors may also contribute to underestimates of SO2 budgets in the satellite dataset, relative to ground-based measurements. Various sources of interference, namely meteorological cloud, volcanic ash, high total ozone columns and the row anomaly, must be also accounted for. Four case studies illustrate the varying impact of these factors at different locations and for different volcanic activity scenarios. In each case, major limitations were apparent. Emissions from Colima (Mexico) and Vulcano and Stromboli (Italy) were not convincingly detected at all. At Popocatépetl (Mexico) and Etna (Italy), while patterns in emissions fluxes were consistent with activity reports, OMI’s quantification of the emissions at both were significantly underestimated relative to more robust ground-based datasets. Challenging observation conditions over Kamchatka limit monitoring to the summer months, and even during these, an underestimate relative to available independent estimates of degassing was apparent. A large underestimate of total SO2 emissions by OMI relative to ground-based budgets was also observed in the CAVA.

These case studies and the preceding discussion show that it remains unclear in many cases whether volcanic activity or local atmospheric conditions are the primary control on the amount of SO2 detected.
by OMI. The inability to determine whether the variation in SO₂ mass burden in the atmosphere over a volcano is caused by volcanism or interference could pose an intractable problem for the successful use of OMI data in the monitoring of continuous volcanic SO₂ emissions into the lower atmosphere. Future investigation into OMI’s application to monitoring is strongly recommended in order to further probe the uncertainties outlined in this review. A full understanding of these factors would open up the widespread exploitation of the large volumes of data on tropospheric degassing already returned by the instrument since its launch in 2004. While the row anomaly renders the data currently being collected by the instrument of more limited use, a multi-year and freely available dataset does exist online in the NASA Goddard Earth Sciences Data and Information Services Centre. Computer software developed for the analysis and visualization of this dataset is also freely available (Carn 2011).

We stress particularly the need for a synergistic approach to volcano monitoring. While multi-satellite comparisons are underway, as well as studies combining satellite- with ground-based approaches, we strongly urge further integration of approaches and techniques. Continued work on understanding how best to use and where possible to improve satellite data is also imperative. Looking beyond OMI, two subsequent generation satellite-borne instruments will provide advanced SO₂ detection capability and therefore present great potential for continued volcano monitoring efforts after the end of the OMI mission lifetime (Fig. 11). This is especially important if degradation of the OMI dataset via the row anomaly continues. NASA’s Ozone Mapping and Profile Suite (OMPS) was launched in October 2011 aboard the Suomi National Polar-orbiting Partnership satellite, and will provide complementary global SO₂ column concentrations, although owing to a coarser spatial resolution, capability may be more limited than that of OMI for detection of tropospheric SO₂ and passive degassing.

The Dutch-designed Tropospheric Ozone Monitoring instrument (TROPOMI), scheduled to be launched aboard ESA’s Sentinel-5 Precursor in 2015, is an even more promising future instrument. TROPOMI has been designed as a replacement for OMI and SCIAMACHY and will offer both the global daily coverage of the former with the large spectral range of the latter. TROPOMI will measure in the UV–vis–near-infrared–short-wavelength infrared ranges, with high spectral resolution and better nadir spatial resolution than either earlier satellite. This instrument is an exciting prospect, as it should be expected to show even greater sensitivity to SO₂ than OMI. The focus on tropospheric measurements, chiefly owing to the mission focus on air quality, is greatly advantageous to volcano-monitoring applications too. Clearly, in the time prior to TROPOMI’s launch, further investigation of limitations and uncertainties in the OMI dataset could be of great future value to the subsequent use of TROPOMI data.

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