

Review

Multi-decadal satellite measurements of global volcanic degassing

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ABSTRACT

Satellite instruments have been providing measurements of global volcanic emissions of sulfur dioxide (SO₂) since 1978, based on observations in the ultraviolet (UV), infrared (IR) and microwave spectral bands. We review recent advances in satellite remote sensing of volcanic gases, focusing on increased instrument sensitivity to tropospheric SO₂ emissions and techniques to determine volcanic plume altitude. A synthesis of ~36 years of global UV, IR and microwave satellite measurements yields an updated assessment of the volcanic SO₂ flux to the upper troposphere and lower stratosphere (UTLS) between 1978 and 2014 (~1–2 Tg/yr). The present availability of multiple UV and IR satellite SO₂ products provides increased confidence in calculated SO₂ loadings for many eruptions. We examine the temporal and latitudinal distribution of volcanic SO₂ emissions and reassess the relationship between eruptive SO₂ discharge and eruption magnitude, finding a first-order correlation between SO₂ emission and volcanic explosivity index (VEI), but with significant scatter. Based on the observed SO₂-VEI relation, we estimate the fraction of eruptive SO₂ emissions released by the smallest eruptions (~0.48 Tg/yr), which is not recorded by satellite observations. A detailed breakdown of the sources of measured SO₂ emissions reveals intuitively expected correlations between eruption frequency, SO₂ loading and volcanic degassing style. We discuss new constraints on e-folding times for SO₂ removal in volcanic plumes, and highlight recent measurements of volcanic hydrogen chloride (HCl) injections into the UTLS. An analysis of passive volcanic emissions of SO₂ detected in Ozone Monitoring Instrument (OMI) SO₂ data since 2004 provides new insight into the location and stability of the dominant sources of volcanic SO₂ over the past decade. Since volcanic SO₂ emissions constitute a random, highly variable perturbation to the atmosphere-climate system, continued monitoring of volcanic SO₂ emissions from space by multiple UV and IR instruments to extend the current multi-decadal record is essential, and near-global, geostationary measurements of SO₂ may be available by the end of the current decade.

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Contents

1. Introduction	100
2. Satellite measurements of volcanic gases	101
3. Satellite instruments and data	105
3.1. TOMS (UV)	107
3.2. TOVS/HIRS (IR)	107
3.3. AIRS (IR)	107
3.4. OMI (UV)	108
3.5. MLS (microwave) and IR limb sounders	111
3.6. IASI (IR)	111
3.7. Volcanic activity and eruption altitude information	111
4. Results and discussion	112
4.1. Eruptive degassing	112
4.1.1. General trends in eruptive degassing	112
4.1.2. Eruptive SO ₂ flux	116

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4.1.3.	VEI-SO ₂ loading relationships	117
4.1.4.	Sources of eruptive volcanic SO ₂ emissions	121
4.1.5.	Lifetimes of SO ₂ in the UTLS	123
4.1.6.	Volcanic HCl emissions into the UTLS	123
4.2.	Passive volcanic SO ₂ degassing	125
5.	Discussion	127
6.	Conclusions	127
Acknowledgments		129
Appendix A. Supplementary data		129
References		129

1. Introduction

Since the first satellite observation of volcanic sulfur dioxide (SO₂) emissions, made after the major eruption of El Chichón (Mexico) in 1982 (Krueger, 1983; Krueger et al., 2008), satellites have provided unique and critical observations of global volcanic degassing. Prior to the satellite remote sensing era, which for SO₂ measurements began in 1978 with the launch of the first Total Ozone Mapping Spectrometer (TOMS; Krueger, 1983; Krueger et al., 1995) and TIROS Operational Vertical Sounder (TOVS) instruments (Prata et al., 2003), the variability of sulfur emissions from volcanic eruptions was very poorly understood, aside from what could be gleaned from early glaciochemical studies of ice cores (Hammer, 1977; Hammer et al., 1980). Satellite measurements provided unique constraints on the vast SO₂ clouds discharged by the major eruptions of El Chichón in 1982 and Pinatubo (Philippines) and Cerro Hudson (Chile) in 1991 (Krueger, 1983; Doiron et al., 1991; Bluth et al., 1992; Schneider et al., 1999; Constantine et al., 2000; Guo et al., 2004; Krueger et al., 2008), spawning a paradigm shift in our understanding of the petrological origins of ‘excess sulfur’ emissions (e.g., Luhr et al., 1984; Westrich and Gerlach, 1992; Wallace and Gerlach, 1994; Scaillet et al., 2003; Oppenheimer et al., 2011; Wallace and Edmonds, 2011). More recently, improvements in instrument sensitivity have extended the capabilities of satellite measurements to include smaller eruptions, tropospheric SO₂ emissions (e.g., passive volcanic degassing) and other minor volatile species present in volcanic emissions such as hydrogen sulfide (H₂S), hydrogen chloride (HCl), bromine monoxide (BrO) and chlorine dioxide (OCIO) (e.g., Eisinger and Burrows, 1998; Carn et al., 2008b; Theys et al., 2009; Clarisse et al., 2011a,b; Carn et al., 2013; Theys et al., 2014; Carn, 2015a).

Oppenheimer et al. (2011) review the significance of volcanic sulfur emissions for volcano monitoring and their wide-ranging impacts on several facets of the Earth system, including atmospheric chemistry and climate, terrestrial and aquatic environments, and human and animal health. In the context of satellite measurements, which are global in scope and provide unique observations of the largest volcanic SO₂ clouds, the primary motivation for monitoring volcanic SO₂ emissions is their potential impact on atmospheric chemistry and climate (e.g., Robock, 2000). Many of the relevant science questions that we can address using satellite measurements of volcanic degassing have been outlined by the Stratospheric Sulfur and its Role in Climate (SSIRC; <http://www.sparc-ssirc.org/>) working group, part of the World Climate Research Program’s (WCRP’s) Stratosphere-Troposphere Processes and their Role in Climate (SPARC) project. Current research foci include: the stratospheric sulfur burden and its partitioning between sulfur species including SO₂, aerosol, H₂SO₄, and OCS, etc.; trends in stratospheric aerosol optical depth (SAOD) and the major SAOD forcing mechanisms; the nature of ‘background’ stratospheric aerosol (e.g., Deshler et al., 2006; Nagai et al., 2010; Solomon et al., 2011); the role of minor volcanic eruptions in modulating the stratospheric sulfur burden, particularly since the 1991 Pinatubo eruption; trends in emissions of aerosol precursor gases (e.g., SO₂); and the time-constant for SO₂ to H₂SO₄ conversion in SO₂-rich volcanic plumes. In order to address these questions, we require measurements of aerosol precursor gases (including SO₂ and OCS) and analyses of their temporal variability. Monitoring stratospheric injections of sulfur by

volcanic eruptions is of prime importance, though quantification of tropospheric volcanic sulfur emissions is also needed since tropospheric sulfur can cross the tropopause via other mechanisms such as deep tropical convection (e.g., Sheng et al., 2015).

Continuous monitoring of volcanic injections of SO₂ into the upper troposphere and lower stratosphere (UTLS) is also required to assess the potential impacts of, and need for, geoengineering of Earth’s climate (specifically, the branch of geoengineering referred to as solar radiation management). Injection of sulfate aerosol precursors into the stratosphere, mimicking volcanic forcing of climate, has been proposed to mitigate the impacts of anthropogenic global warming (e.g., Crutzen, 2006; Wigley, 2006; Robock et al., 2009). Accurate characterization of the time-averaged volcanic SO₂ flux into the stratosphere is crucial in this regard, and analyses of volcanic eruptions of varying magnitude provide unique insights into the behavior of SO₂ in the UTLS after injection. Recently, volcanic influence on the post-Pinatubo stratosphere has been scrutinized in considerable detail in order to establish the cause of the reduced rate of global warming observed in global mean surface temperatures over the last decade (dubbed the global warming ‘hiatus’). A particular focus has been the role of ‘small’ volcanic eruptions, or those with a volcanic explosivity index (VEI; Newhall and Self, 1982) below 5. Although alternative causes and even the very existence of the global warming hiatus are subject to ongoing debate (e.g., Schmidt et al., 2014; Steinman et al., 2015; Karl et al., 2015), several studies have concluded that SAOD perturbations by small volcanic eruptions not included in pre-hiatus climate model simulations have likely played a role (e.g., Solomon et al., 2011; Vernier et al., 2011; Neely et al., 2013; Ridley et al., 2014; Santer et al., 2014, 2015). But regardless of the dominant forcing factors influencing recent trends in global warming, it remains crucial to monitor volcanic degassing and quantify SO₂ and other volcanic gas fluxes into the atmosphere.

In this contribution we review the current state of our knowledge of volcanic degassing (primarily focused on SO₂ emissions) based on multi-decadal satellite measurements since 1978, which have been collated in a new volcanic SO₂ emissions database (Carn, 2015c). This review discusses both the volcanological and atmospheric applications of the SO₂ emissions database. We do not offer a detailed overview of any specific volcanic eruptions, but instead synthesize ~36 years of satellite observations and augment previous compilations of satellite SO₂ data for volcanic eruptions (e.g., Bluth et al., 1993, 1997; Carn et al., 2003) and estimates of global volcanic SO₂ emissions (e.g., Kellogg et al., 1972; Cadle, 1975; Berresheim and Jaeschke, 1983; Le Guern, 1982; Stoiber et al., 1987; Spiro et al., 1992; Andres and Kasgnoc, 1998; Halmer et al., 2002; Diehl et al., 2012), incorporating a significant amount of new data on smaller eruptions and passive degassing collected since 2004. The paper is organized as follows. Firstly (Section 2), we briefly review recent developments in satellite remote sensing of volcanic gases, focusing on improvements in instrument sensitivity and in techniques to determine volcanic plume altitude. In Section 3 we describe the ultraviolet (UV), infrared (IR) and microwave satellite datasets used to generate the multi-decadal volcanic SO₂ emissions database. The main results are presented in Section 4, including the temporal and latitudinal distribution of volcanic SO₂ emissions, updated estimates of the annual volcanic SO₂ flux from volcanic eruptions (including the fraction not recorded by satellite

observations) and a reassessment of the relationship between eruptive SO₂ discharge and eruption magnitude (VEI). We also provide a detailed breakdown of the sources of the measured SO₂ emissions and attempt to classify them based on degassing style. New constraints on e-folding times for SO₂ removal in volcanic plumes are discussed, and we also highlight recent measurements of volcanic HCl injections into the UTLS. An analysis of the major sources of passive volcanic emissions of SO₂ detected in satellite data is presented in Section 4.2, followed by a discussion (Section 5) and conclusions.

2. Satellite measurements of volcanic gases

Although to date no satellite instrument has been deployed to specifically target volcanic emissions, passive UV, IR and microwave sensors have been used to measure seven volcanic gas species since 1978 (SO₂, H₂S, HCl, CO, BrO, OCIO and CH₃Cl; Table 1). However, SO₂ is by far the most frequently measured volcanic gas, and it is currently not possible to measure other volcanic gases from space with comparable accuracy or frequency to that of SO₂. It is the major target for remote

Table 1
Volcanic gases measured or potentially detectable from space.

Sensor ^a	Volatile species								Timespan	Reference(s) ^b	
	H ₂ O	CO ₂	CO	SO ₂	H ₂ S	HCl	BrO	OCIO	CH ₃ Cl		
TOMS*										1978–2005	1, 2
SBUV* (P)										1978–present	3, 4
HIRS*										1978–present	5
GOME										1995–2003	6, 7, 8, 9
MODIS*										1999–present	10, 11
ASTER										1999–present	12, 13, 14
MOPITT			Red							1999–present	15
SCIAMACHY (L)	Gray	Gray		Red						2002–2012	8, 16, 17, 18
MIPAS (L)										2002–2012	19
AIRS	Gray	Gray	Gray							2002–present	20, 21
ACE (L)	Gray	Gray	Gray		Gray					2003–present	22
SEVIRI										2004–present	23
OMI							Red	Red		2004–present	18, 24, 25, 26
MLS* (L)	Gray	Gray			Red	Gray		Red		1991–2001; 2004–present	27, 28, 29, 30
TES (P)				Red						2004–present	31
GOME-2*	Gray			Red			Red	Red		2006–present	18, 32, 33, 34
IASI*	Gray	Gray		Red	Red					2006–present	15, 35, 36
OMPS*				Red			Gray			2011–present	37
VIIRS				Red						2011–present	38
CrIS				Gray						2011–present	39
AHI				Gray						2015–present	40
GOSAT (P)	Gray	Gray	Gray							2009–present	41
OCO-2	Gray	Gray	Gray							2014–present	42

Red = detected in a volcanic cloud; Light gray = potentially detectable but not yet proven in a volcanic context and/or not viable for routine volcanic measurements (e.g., due to background interference).

a. Sensor acronyms: TOMS: Total Ozone Mapping Spectrometer; SBUV: Solar Backscatter Ultraviolet Instrument; HIRS: High-resolution Infrared Radiation Sounder; GOME: Global Ozone Monitoring Experiment; MODIS: Moderate resolution Imaging Spectroradiometer; ASTER: Advanced Spaceborne Thermal Emission and Reflection Radiometer; MOPITT: Measurements of Pollution in the Troposphere; SCIAMACHY: Scanning Imaging Absorption Spectrometer for Atmospheric Chartography; MIPAS: Michelson Interferometer for Passive Atmospheric Sounding; AIRS: Atmospheric Infrared Sounder; ACE: Atmospheric Chemistry Experiment; SEVIRI: Spinning Enhanced Visible and Infrared Imager; OMI: Ozone Monitoring Instrument; MLS: Microwave Limb Sounder; TES: Tropospheric Emission Spectrometer; IASI: Infrared Atmospheric Sounding Interferometer; OMPS: Ozone Mapping and Profiler Suite; VIIRS: Visible Infrared Imaging Radiometer Suite; CrIS: Cross-track Infrared Sounder; AHI: Advanced Himawari Imager; GOSAT: Greenhouse Gases Observing Satellite; OCO-2: Orbiting Carbon Observatory 2.

* = Sensor flown (or to be flown) on multiple satellites; P: nadir profiling and/or pointable instrument (limited mapping capability); L: limb instrument (vertical profiling); all other instruments provide some spatial mapping capability (SCIAMACHY provided nadir and limb observations).

b. 1. Krueger et al. (1995); 2. Krueger et al. (2000); 3. McPeters et al. (1984); 4. McPeters (1993); 5. Prata et al. (2003); 6. Eisinger and Burrows (1998); 7. Thomas et al. (2005); 8. Afe et al. (2004); 9. Noël et al. (1999); 10. Watson et al. (2004); 11. Corradini et al. (2009); 12. Urai (2004); 13. Pugnagh et al. (2006); 14. Campion et al. (2010); 15. Martínez-Alonso et al. (2012); 16. Noël et al. (2004); 17. Lee et al. (2008); 18. Theys et al. (2014); 19. Höpfner et al. (2013); 20. Carn et al. (2005); 21. Prata and Bernardo (2007); 22. Doeringer et al. (2012); 23. Prata and Kerkemann (2007); 24. Krotkov et al. (2006); 25. Yang et al. (2007); 26. Carn et al. (2013); 27. Read et al. (1993); 28. Prata et al. (2007); 29. Pumphrey et al. (2015); 30. W.G. Read, pers. Comm.; 31. Clerbaux et al. (2008); 32. Noël et al. (2008); 33. Rix et al. (2009); 34. Theys et al. (2009); 35. Clarisse et al. (2008); 36. Clarisse et al. (2011a,b); 37. Carn et al. (2015); 38. V.J. Realmutto, pers. Comm.; 39. Gambacorta and Barnet (2013); 40. Carn (2015a); 41. Schwandner and Carn (2011); 42. Schwandner et al. (2015).

sensing due to its ease of measurement relative to the dominant volcanic volatile species, which are typically water vapor (H_2O) and carbon dioxide (CO_2). The characteristics of SO_2 that enable space-based measurements over at least 5 orders of magnitude in atmospheric column amount (without spatial or temporal data averaging) are its relative abundance in volcanic emissions (typically 2nd or 3rd after H_2O and CO_2 ; e.g., Oppenheimer et al., 2011), low background SO_2 concentrations away from strong anthropogenic point sources (power plants and metal smelters; Carn et al., 2007), and the accessibility of absorption bands for remote sensing in the UV, IR and microwave spectral regions. The relatively high emission altitude (cf. anthropogenic sources) and consequent atmospheric lifetime of volcanic SO_2 (typically hours to days) is also conducive to satellite measurements, the latter being sufficiently long to enable detection and tracking of volcanic SO_2 plumes despite the large spatial scale of satellite sensor footprints, but short enough to prevent significant accumulation in the atmosphere and interference with fresh emissions (in contrast to CO_2). To date, the only point sources of anthropogenic SO_2 emissions observed to rival volcanic SO_2 plumes in terms of signal strength have been metal smelters (e.g., Khokhar et al., 2005; Carn et al., 2007; Walter et al., 2012; Bauduin et al., 2014) and rare sulfur fires (Carn et al., 2004; Kearney et al., 2009), although, on a global scale, estimated total anthropogenic sulfur emissions may exceed volcanic sulfur emissions by up to an order of magnitude, with significant uncertainty (e.g., Chin and Jacob, 1996; Graf et al., 1997; Andres and Kasgnoc, 1998). The main impediment to detection of volcanic CO_2 and H_2O emissions from space is the small volcanic contributions relative to the ambient atmospheric concentrations of these molecules (e.g., Gerlach, 2011). Whilst the detection of volcanic CO_2 from space has not yet been conclusively demonstrated, it is likely to occur as more data from instruments providing CO_2 retrievals, including GOSAT, OCO-2, SCIAMACHY, AIRS and IASI (Table 1), are analyzed (e.g., Schwandner and Carn, 2011; Schwandner et al., 2015). But, although several operational sensors retrieve profiles of atmospheric water vapor (Table 1), satellite detection of volcanic H_2O is extremely unlikely due to even greater background interference than for CO_2 , with the possible exception of volcanic H_2O injections into the dry stratosphere.

Satellite remote sensing of SO_2 exploits the molecule's absorption bands at UV, IR and microwave wavelengths (e.g., Carn, 2015a). Somewhat fortuitously, the overlap of SO_2 absorption features in the UV and IR with spectral regions used to retrieve higher priority measurements or atmospheric constituents (e.g., ozone, water vapor, clouds and surface temperature) has ensured that most operational satellite instruments have included channels suitable for SO_2 detection. UV measurements utilize SO_2 absorption bands at wavelengths of 300–340 nm, in the Huggins ozone absorption band; hence ozone and SO_2 must be retrieved simultaneously and large column amounts of ozone can impact the sensitivity of UV measurements to lower tropospheric or boundary layer SO_2 (e.g., Krotkov et al., 2008). IR measurements of SO_2 can exploit three vibrational absorption bands in the thermal IR at ~4 μm ($\nu_1 + \nu_3$ -band), ~7.3 μm (ν_3 -band) and ~8.6 μm (ν_1 -band). Of these, the 7.3 μm band is most often used since it is the stronger band (providing greater sensitivity), is less susceptible to interference by volcanic ash (e.g., Corradini et al., 2009), and provides sensitive measurements of SO_2 in the upper troposphere and above (e.g., Rinsland et al., 1995; Prata et al., 2003; Carn et al., 2005; Prata and Bernardo, 2007; Clarisse et al., 2008, 2012; Doeringer et al., 2012; Höpfner et al., 2013). It lacks sensitivity to lower tropospheric SO_2 (<3–5 km altitude; i.e., small eruptions and passive degassing) notwithstanding relatively rare coincidences of surface temperature inversions, extreme pollution levels and low humidity (e.g., Boynard et al., 2014; Bauduin et al., 2014). The typically poor sensitivity to lower tropospheric SO_2 at 7.3 μm is due to interference from water vapor absorption, but this also simplifies retrievals somewhat since the background 7.3 μm radiation originates from relatively homogeneous (on the scale of a volcanic cloud) lower tropospheric water vapor. The 4 μm and 8.6 μm bands are

weaker but are situated in atmospheric windows, allowing retrieval of lower tropospheric SO_2 , but with the added complexity of dependence on variable background emissivity, especially over land (e.g., Realmuto et al., 1994, 1997; Teggi et al., 1999; Realmuto, 2000; Watson et al., 2004; Prata and Bernardo, 2007; Prata and Kerkemann, 2007; Kearney et al., 2008), atmospheric water vapor (Realmuto and Worden, 2000), daytime solar reflection at 4 μm , and interference from volcanic ash at 8.6 μm (Corradini et al., 2009). Microwave limb sounding of SO_2 in the UTLS by the Microwave Limb Sounder (MLS) instruments has exploited rotational absorption bands of the molecule at frequencies of 200–240 GHz (Read et al., 1993; Pumphrey et al., 2015).

The era of satellite observations of volcanic degassing (primarily SO_2) can be broadly divided based on sensor specifications (mainly spectral and spatial resolution), which directly impact sensitivity to SO_2 abundance and altitude and hence to different styles of volcanic activity. Between October 1978 and December 2005, four TOMS missions provided near-continuous multi-spectral, global, nadir UV observations at low spatial resolution (39–50 km at nadir), with occasional small data gaps and a major 19-month gap in 1995–96 between the Meteor-3 and Earth Probe (EP) TOMS missions (Krueger et al., 1995, 2000; Fig. 1). The limited number of TOMS wavelengths restricted measurements to larger volcanic eruptions and extreme levels of passive SO_2 degassing (e.g., Bluth et al., 1993; Carn et al., 2003; Carn, 2004). SBUV (flown since 1978; Table 1) has also provided nadir UV observations of volcanic SO_2 clouds, albeit with poorer spatial sampling than TOMS, but only for the largest eruptions (McPeters et al., 1984; McPeters, 1993; Schnetzler et al., 1995). Coincident with TOMS, the High-resolution Infrared Radiation Sounder and subsequent iterations (HIRS, HIRS/2, HIRS/3, etc.), flown as part of the TIROS Operational Vertical Sounder (TOVS) package on NOAA polar-orbiters since October 1978, provide multi-spectral, global IR SO_2 measurements at 7.3 μm (Prata et al., 2003). The HIRS SO_2 data provide coverage of the 1995–96 TOMS data gap and also offer nighttime and high-latitude winter measurements, unlike TOMS.

Hyperspectral UV measurements, offering increased sensitivity to lower tropospheric SO_2 due to optimized spectral resolution (Krueger et al., 2009), began with the Global Ozone Monitoring Experiment (GOME) in July 1995 (Burrows et al., 1999). GOME provided some of the first satellite observations of lower tropospheric SO_2 from volcanic and anthropogenic sources despite its low spatial resolution (40 × 320 km at nadir; Eisinger and Burrows, 1998), and detected numerous volcanic eruptions during its operational lifetime (1995–2003; e.g., Afe et al., 2004; Thomas et al., 2005; Khokhar et al., 2005). However, a GOME inventory of volcanic SO_2 mass loading has not been widely reported to date and hence GOME SO_2 data are not included in the database presented here (Fig. 1). GOME was superseded by the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY; Bovensmann et al., 1999), launched on Envisat in March 2002 and operational from 2002 to 2012. SCIAMACHY provided increased sensitivity to tropospheric SO_2 over GOME due to higher spatial resolution (30 × 60 km at nadir), and also detected many volcanic SO_2 emissions (e.g., Afe et al., 2004; Lee et al., 2008, 2009; Penning de Vries et al., 2014) and provided the first satellite observation of volcanic OCIO (Theys et al., 2014), but its spatial coverage was limited by alternation between nadir and limb mode observations. Thus, SCIAMACHY-derived volcanic SO_2 mass loadings have also not been widely reported and are not used herein, although there is potential for further analysis of the SCIAMACHY limb mode data archive to constrain volcanic plume altitudes (e.g., Penning de Vries et al., 2014). The Ozone Monitoring Instrument (OMI), launched on NASA's Aura satellite in July 2004, has provided operational, hyperspectral nadir UV SO_2 measurements with a nadir spatial resolution of 13 × 24 km since September 2004 (Fig. 1; Krotkov et al., 2006; Yang et al., 2007). OMI SO_2 measurements have been the primary source of data for continuation of the TOMS record of volcanic SO_2 emissions, but with increased sensitivity to small eruptions and passive degassing (Carn et al., 2008b; Carn and Prata, 2010; McCormick et al., 2012, 2013; Campion et al., 2012; Carn et al., 2013).

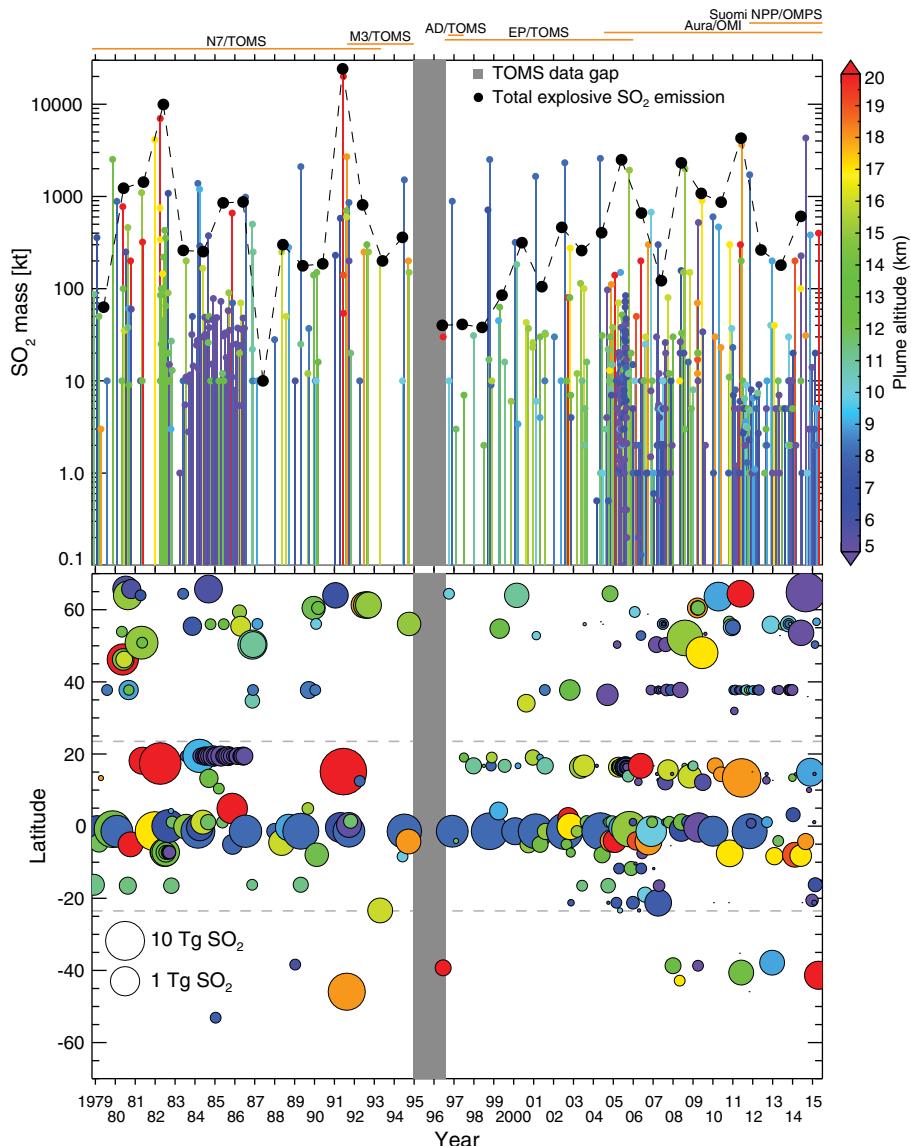


Fig. 1. Satellite measurements of SO₂ emissions by volcanic eruptions between October 1978 and April 2015. Measurements are primarily based on UV TOMS, OMI and OMPS data, supplemented by IR data from TOVS, AIRS and IASI. Eruptions are color-coded by estimated plume altitude, derived from a variety of sources, including Smithsonian Institution Global Volcanism Program volcanic activity reports, volcanic ash advisories, and satellite data (e.g., CALIOP lidar for eruptions since 2006). *Upper panel* shows SO₂ mass loading against time, with annual total explosive volcanic SO₂ production (omitting SO₂ discharge from effusive eruptions) shown in black. Orange lines above the plot indicate the operational lifetimes of the UV satellite instruments: Nimbus-7 (N7), Meteor-3 (M3), ADEOS (AD), and Earth Probe (EP) TOMS, OMI (currently operational) and OMPS (currently operational). The TOMS data gap in 1995–96 is shown in gray (but note that TOVS SO₂ data provide coverage during this time). *Lower panel* shows eruption latitude against time, with symbol size proportional to SO₂ mass loading. Horizontal dashed lines delineate the tropics. The charts only depict SO₂ emissions from discrete volcanic eruptions; continuous emissions from passive degassing and some smaller eruptions are not included. Over 600 eruptions are shown, releasing a total of ~100 Tg of SO₂. The database shown in this plot will soon be available from the NASA Goddard Earth Sciences (GES) Data and Information Services Center (DISC) as a level 4 MEaSUREs (Making Earth System Data Records for Use in Research Environments) data product.

Overlap between EP/TOMS in OMI in 2004–2005 provides opportunities for comparison of multi-spectral and hyperspectral SO₂ retrievals for several eruptions (Fig. 1; Table 2), to assess continuity between satellite missions. Second generation GOME (GOME-2) instruments were launched on the MetOp-A and MetOp-B satellites in October 2006 (Munro et al., 2006) and September 2012, respectively, offering higher spatial resolution than GOME (80 × 40 km) and high sensitivity to volcanic SO₂ and BrO emissions (e.g., Theys et al., 2009; Nowlan et al., 2011; Rix et al., 2012; Theys et al., 2013), but lower sensitivity to passive SO₂ degassing than OMI due to OMI's higher spatial resolution. However, as for GOME and SCIAMACHY, we have not made extensive use of GOME-2 data in this study. A further hyperspectral UV sensor, the Ozone Mapping and Profiler Suite (OMPS), was launched on the Suomi National Polar-orbiting Partnership (SNPP) satellite in October 2011 and will be flown on future NOAA Joint Polar Satellite System

(JPSS) polar-orbiters beginning in 2017. OMPS has variable spatial resolution (50 × 50 km in standard mode and 10 × 10 km in zoom mode, currently used once weekly on Saturdays) and comparable sensitivity to volcanic SO₂ to SCIAMACHY, OMI and GOME-2 (e.g., Yang et al., 2013) and is also being used to extend the UV volcanic SO₂ measurements in conjunction with OMI (Carn et al., 2015; Fig. 1).

IR satellite sensors have shown a similar evolution to UV instruments. Following on from TOVS/HIRS, multi-spectral TIR observations from the Moderate resolution Imaging Spectroradiometer (MODIS) and Advanced Spaceborne Thermal Emission and Reflection radiometer (ASTER) began following the launch of Terra in December 1999, and a second MODIS instrument was deployed on Aqua in May 2002. Both Terra and Aqua MODIS and ASTER continue to operate at the time of writing. MODIS and ASTER data can be used for IR SO₂ retrievals at moderate-to-high spatial resolution (90 m to 1 km) using channels at

Table 2SO₂ mass loadings for selected volcanic eruptions (1978–2014).

Volcano	Eruption date(s)	Total SO ₂ loading (Tg)	Sensor(s)	Reference
Nyamuragira	1979–2004	~25 ^a	TOMS	Bluth and Carn (2008)
Hekla	17 Aug. 1980	0.6 ± 0.1	TOMS	Carn et al. (2008a)
El Chichón	28 Mar. 1982	1.6	TOMS	Krueger et al. (2008)
	3–4 Apr. 1982	5.9	TOMS	Krueger et al. (2008)
Galunggung	Apr 1982–Jan. 1983	2.03	TOMS	Bluth et al. (1994)
Nevado del Ruiz	13 Nov. 1985	0.66	TOMS	Krueger et al. (1990)
Redoubt	15 Dec. 1989	0.18	TOMS	Schnetzler et al. (1994)
Hekla	17 Jan. 1991	0.4 ± 0.1	TOMS	Carn et al. (2008a)
Pinatubo	15 Jun. 1991	18 ± 4	TOMS	Guo et al. (2004)
		19 ± 4	HIRS/2	Guo et al. (2004)
		17	MLS	Read et al. (1993)
		12–15	SBUV/2	McPeters (1993)
Hudson	8–12 Aug. 1991	4	TOMS	Constantine et al. (2000)
Mt. Spurr	27 Jun. 1992	0.25	TOMS	Bluth et al. (1995)
	18 Aug. 1992	0.3	TOMS	Bluth et al. (1995)
	17 Sep. 1992	0.25	TOMS	Bluth et al. (1995)
Ruapehu	17 Jun. 1996	0.08	HIRS/2	Prata et al. (2003)
Hekla	26 Feb. 2000	0.2	TOMS/HIRS	Rose et al. (2003)
Miyakejima	18 Aug. 2000	0.043	Multiple	McCarthy et al. (2008)
Popocatepetl	23 Jan. 2001	0.017	MODIS 7.3 μm	Matiella Novak et al. (2008)
Nyiragongo	17 Jan. 2002	0.03	TOMS	Carn (2004)
Etna	30 Oct. 2002	0.025	AIRS	Carn et al. (2005)
		0.022	AIRS	Prata et al. (2007)
Anatahan	10–12 May 2003	0.11	TOMS	Wright et al. (2005)
Soufriere Hills	13 Jul. 2003	0.10	TOMS	Carn and Prata (2010)
		0.13	MODIS 7.3 μm	Carn and Prata (2010)
		0.12	AIRS	Carn and Prata (2010)
Soputan	Oct 2004–Oct. 2008	0.11 ^b	OMI	Kushendratno et al. (2012)
Manam	28 Jan. 2005	0.14	TOMS	<i>This study</i>
		0.14	OMI	<i>This study</i>
		0.13	AIRS	Prata and Bernardo (2007)
Anatahan	6 Apr. 2005	0.070	TOMS	<i>This study</i>
		0.075	OMI	<i>This study</i>
		0.077	AIRS	Prata and Bernardo (2007)
Sierra Negra	22 Oct. 2005	1.74	OMI	Yang et al. (2009a)
		0.33	TOMS	Thomas et al. (2009)
		1.27	MODIS 8.6 μm	Thomas et al. (2009)
Karthala	24 Nov. 2005	0.08	SEVIRI	Prata and Kerkmann (2007)
Soufriere Hills	20 May 2006	0.22	OMI	Carn and Prata (2010)
		0.23	AIRS	Carn and Prata (2010)
		0.18	SEVIRI	Prata et al. (2007)
P. Fournaise	4–9 Apr. 2007	0.23	OMI	Tulet and Villeneuve (2011)
Manda Hararo	12 Aug. 2007	0.03	OMI	Ferguson et al. (2010)
Jebel al-Tair	30 Sep. 2007	0.08 ± 0.02	Multiple	Eckhardt et al. (2008)
		0.05	IASI	Claris et al. (2008)
Chaitén	2–8 May 2008	0.01	OMI	Carn et al. (2009a)
Okmok	12 Jul. 2008	0.12	OMI	Spinei et al. (2010)
		0.3	AIRS	Prata et al. (2010)
		0.09	IASI	<i>This study</i>
Kasatochi	7 Aug. 2008	1.7	IASI	Karagulian et al. (2010)
		1.2	AIRS	Prata et al. (2010)
		1.7	Multiple	Kristiansen et al. (2010)
		2.2	OMI	Krotkov et al. (2010)
		0.9 ± 0.2	MODIS 7.3 μm	Corradini et al. (2010)
		2.7 ± 0.8	MODIS 8.6 μm	Corradini et al. (2010)
		1.6	IASI	Claris et al. (2012)
		1.6	GOME-2	Nowlan et al. (2011)
Dalafilla	3 Nov. 2008	0.23	IASI	<i>This study</i>
Redoubt	23 Mar.–12 Jun. 2009	0.54–0.61 ^c	OMI	Lopez et al. (2013a)
Sarychev Peak	15–16 Jun. 2009	1.2	IASI	Haywood et al. (2010)
Manda Hararo	28 Jun. 2009	0.03	OMI	Ferguson et al. (2010)
Eyjafjallajökull	1–12 May 2010	0.17	IASI + Model	Boichu et al. (2013)
	5–18 May 2010	1.2 ± 0.5	GOME-2	Rix et al. (2012)
	7 May 2010	0.18	IASI	Carboni et al. (2012)
Manda Hararo	21 May 2010	0.001	GOME-2+OMI	Barnie et al., 2015
Merapi	26 Oct.–4 Nov. 2010	0.44	Multiple	Surono et al. (2012)
Kizimen	13–31 Dec. 2010	0.06	IASI	<i>This study</i>
Kirishima	26 Jan. 2011	0.03	IASI	<i>This study</i>
Grimsövtin	21 May 2011	0.38	OMI + IASI	Sigmarsdóttir et al. (2013)
Cordon Caulle	4–6 Jun. 2011	0.2	IASI	Theys et al. (2013)
Nabro	12–28 Jun. 2011	4.5	Multiple	Theys et al. (2013)
Nyamuragira	6 Nov.–6 Dec. 2011	~1	OMI	Theys et al. (2013)
Tolbachik	27 Nov. 2012	0.09	IASI	<i>This study</i>
	Nov 2012–Aug. 2013	0.2	Multiple	Telling et al. (2015)
Copahue	22 Dec. 2012	0.5	IASI	<i>This study</i>
Paluweh	2 Feb. 2013	0.03	Multiple	Carn et al. (2015)
Kelut	14 Feb. 2014	0.2	OMI	<i>This study</i>
		0.19	IASI	<i>This study</i>

^a Total SO₂ release for 14 effusive eruptions.^b Total SO₂ release for 9 explosive eruptions.^c Total SO₂ release for several UTLS injections.

7.3 μm (MODIS) or 8.6 μm (MODIS and ASTER); these sensors have typically been used for specific volcanic eruption or degassing case studies that exploit their higher spatial resolution rather than the production of long-term volcanic SO₂ emissions inventories (e.g., Rose et al., 2003; Watson et al., 2004; Urai, 2004; Pugnaghi et al., 2006; Kearney et al., 2008; McCarthy et al., 2008; Matiella Novak et al., 2008; Corradini et al., 2009, 2010; Campion et al., 2010, 2012; Henney et al., 2012). This is largely due to the difficulty of processing the larger data volumes associated with high spatial resolution data, the limited spatial coverage of ASTER (a result of the trade-off between spatial resolution and swath width), plus the aforementioned interference from volcanic ash at 8.6 μm. Although high-resolution IR spectra were acquired from space as early as 1969 (Clarisso et al., 2011a), global, nadir hyperspectral IR measurements began from orbit in earnest with the launch of the Atmospheric Infrared Sounder (AIRS) on Aqua in 2002 (Chahine et al., 2006), which provides robust retrievals of volcanic SO₂ in the UTLS using the 7.3 μm band (Carn et al., 2005; Prata and Bernardo, 2007; Prata et al., 2007, 2010). AIRS spectra also cover the 4 μm band and part of the 8.6 μm SO₂ band but these data have not been widely exploited to date (Prata and Bernardo, 2007). AIRS SO₂ measurements provide particularly valuable observations of volcanic eruptions between 2002 and the beginning of OMI measurements in September 2004 (Fig. 1), in addition to ongoing complementary coverage of nighttime and high-latitude winter events. Hyperspectral IR measurements of SO₂ were augmented by the Infrared Atmospheric Sounding Interferometer (IASI), first launched on MetOp-A with GOME-2 in 2006 and followed by a second IASI instrument on MetOp-B in 2012. In addition to providing high-quality UTLS volcanic SO₂ measurements (e.g., Clarisse et al., 2008, 2012, 2014; Karagulian et al., 2010; Theys et al., 2013), IASI has provided new observations of other volcanogenic gas and aerosol species including hydrogen sulfide (H₂S; Clarisse et al., 2011a,b) and sulfate aerosol (Clarisse et al., 2010; Karagulian et al., 2010).

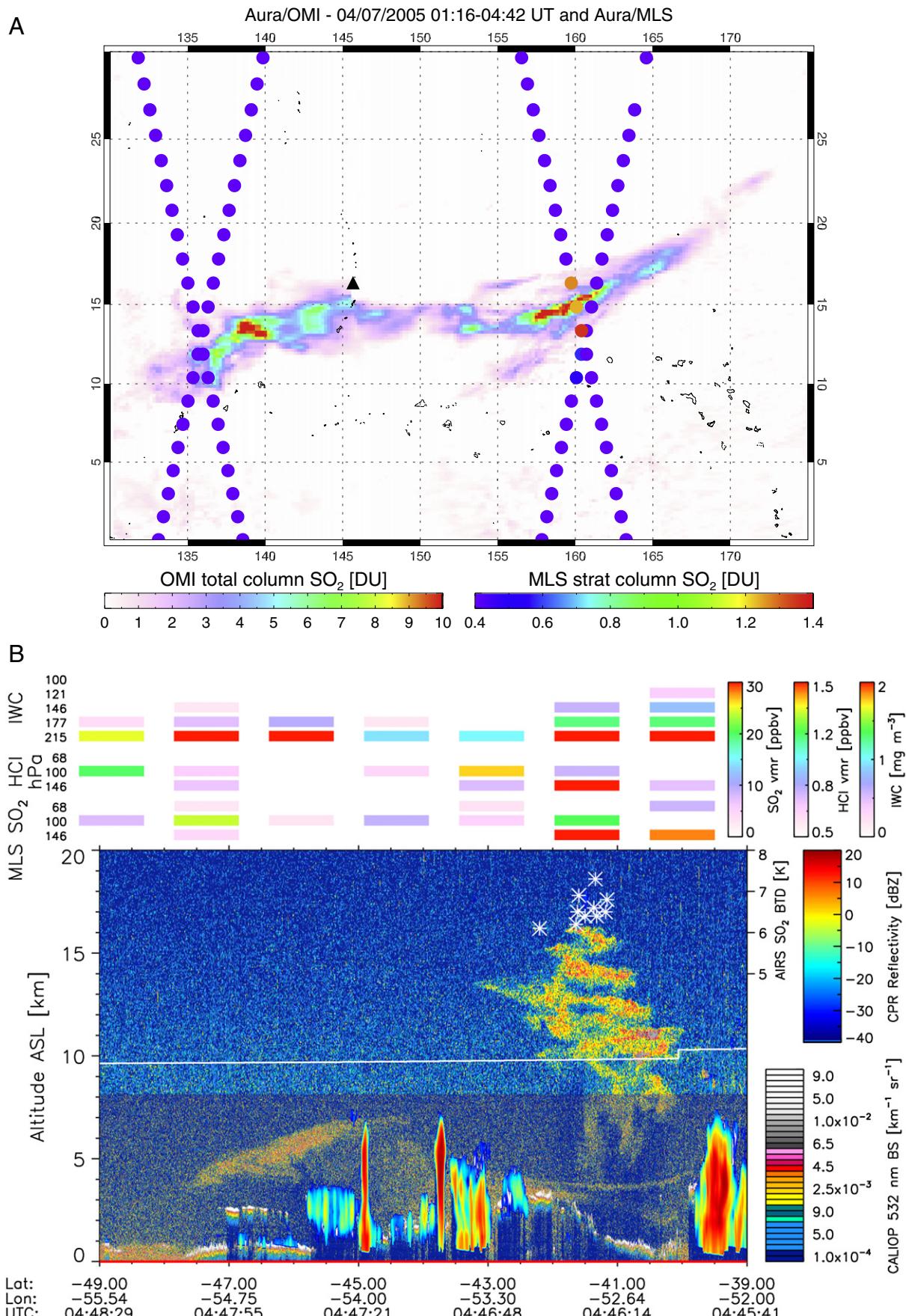
In tandem with the advent of hyperspectral measurements, a key advance in satellite remote sensing that has occurred in the past decade, and one that is particularly relevant to volcanic eruption detection, is the availability of altitude information for SO₂ (and other trace gases) and aerosols to complement measurements of SO₂ mass loading by the nadir UV and IR imagers and sounders described above. Although it has long been possible to estimate the altitude of a volcanic cloud by comparing observed SO₂ or ash cloud transport and winds aloft (e.g., from a radiosonde), by using IR cloud-top brightness temperatures for optically thick plumes, or by using trajectory models (e.g., Schoeberl et al., 1993; Carn et al., 2008a; Krotkov et al., 2010), there are now multiple sources of altitude information, with variable accuracy. Altitude information can be derived directly from satellite instruments in several ways including: active LiDAR or radar remote sensing of volcanic aerosols and hydrometeors (Fig. 2; e.g., Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) or CloudSat data; Carn et al., 2009a, 2015; Spinei et al., 2010; Lopez, 2011; Winker et al., 2012; Fromm et al., 2014; Prata et al., 2015); passive multi-angle imaging (e.g., the Multi-angle Imaging Spectroradiometer [MISR]; Kahn et al., 2007; Kahn and Limbacher, 2012); passive limb sounding of volcanic gases and aerosols in the UV, IR or microwave (e.g., MLS, OSIRIS, MIPAS, SCIAMACHY, ACE, OMPS; Read et al., 1993; Bourassa et al., 2010, 2012, 2013; Doeringer et al., 2012; Höpfner et al., 2013, 2015; Penning de Vries et al., 2014; Pumphrey et al., 2015); solar occultation measurements of volcanic aerosols (e.g., SAGE); or SO₂ altitude retrievals using passive nadir hyperspectral UV or IR measurements (e.g., OMI, GOME-2, IASI; Yang et al., 2009a,b, 2010; Nowlan et al., 2011; Rix et al., 2012; Clarisse et al., 2014). Information on the SO₂ vertical profile can also be gleaned from inverse trajectory modeling of satellite measurements (e.g., Eckhardt et al., 2008; Kristiansen et al., 2010; Hughes et al., 2012) or by comparisons of simultaneous UV and IR measurements, exploiting their different vertical sensitivity (e.g., Fig. 2; Carn et al., 2009b). Whilst it is rare to possess all the above information for a given volcanic eruption, the redundancy offered by multiple techniques

means that some altitude information is nearly always available. The operation of polar-orbiting satellites in close formation along the same orbit track, such as NASA's A-Train afternoon constellation (<http://atrain.nasa.gov/>; including OCO-2, Aqua, CALIPSO, CloudSat and Aura), providing near-coincident passive and active measurements from multiple platforms, has greatly facilitated analysis of volcanic cloud altitude (e.g., Fig. 2; Carn et al., 2009a, 2009b).

To summarize, there have been many improvements in satellite remote sensing of volcanic gases since 1978, largely due to the advent of hyperspectral UV and IR sensors and satellite constellations. Currently (June 2015), as several operational sensors continue to exceed their intended lifetimes, our capabilities for monitoring volcanic SO₂ emissions from space have reached a zenith, with up to ~18 daily SO₂ observations potentially available from nadir imagers and sounders. In chronological order, these are: AIRS and MODIS (Aqua; equator crossing at 1:30 am local time (LT)); VIIRS and CrIS (SNPP; 1:30 am LT), GOME-2B and IASI-B (MetOp-B; 9:00 am LT); GOME-2 A and IASI-A (MetOp-A; 9:30 am LT); MODIS (Terra; 10:30 am LT); AIRS and MODIS (Aqua; 1:30 pm LT); OMPS, VIIRS and CrIS (SNPP; 1:30 pm LT); OMI (Aura; 1:45 pm LT); IASI-B (9:00 pm LT); IASI-A (9:30 pm LT); and MODIS (Terra; 10:30 pm LT). This sequence does not include further observations from multiple consecutive overpasses for high latitude regions (e.g., Kamchatka, Iceland, Alaska), daily SO₂ observations from TOVS/HIRS, limb observations of SO₂ and HCl from MLS on Aura and occasional SO₂ observations by ASTER and the geostationary SEVIRI (for eruptions in Europe, Africa and the eastern Americas). Note that the times listed above are equatorial crossing times (ascending or descending nodes) and the gap between local satellite overpass times may be substantially longer at higher latitudes, although this is offset by the convergence of polar orbits towards the poles. But at low latitudes the maximum data gap between volcanic SO₂ measurements (UV or IR) is currently ~7.5 h (1:30 am–9:00 am and 1:30 pm–9:00 pm), which is useful for near real-time (NRT) monitoring applications (e.g., Brenot et al., 2014) and ensures that very few, if any, volcanic eruptions emitting significant SO₂ go undetected. Having numerous satellite SO₂ measurements available also permits inter-comparisons between UV and IR data and increases confidence in calculated SO₂ mass loadings. Furthermore, the recently launched Earth Polychromatic Imaging Camera (EPIC) on the NOAA Deep Space Climate Observatory (DSCOVR) will soon provide the first volcanic SO₂ measurements from the L₁ Earth-Sun Lagrange point (<http://www.nesdis.noaa.gov/DSCOVR/>). Beginning in 2016, EPIC will provide multi-spectral UV SO₂ measurements of the sunlit Earth disk several times per day, with spatial resolution similar to OMI, offering improved temporal resolution over LEO UV sensors. The L₁ orbit offers a unique perspective of Earth, distinct from a geostationary orbit (GEO). Like GEO, year-round L₁ observations will be best suited to eruptions at low- to mid-latitudes, but whereas GEO observations are always limited at high latitudes, EPIC will provide good coverage of the high latitude summer hemisphere as the Earth's axis tilts towards the Sun.

3. Satellite instruments and data

Here, we describe the specific datasets used in the multi-decadal analysis of volcanic SO₂ emissions in chronological order of sensor deployment, namely TOMS, TOVS/HIRS, AIRS, OMI, MLS, IASI and OMPS. We do not provide detailed descriptions of retrieval algorithms, since these have been extensively reported elsewhere and the focus of this review is interpretation of the measurements. Limitations and uncertainties in SO₂ retrievals will be acknowledged as appropriate, but we will also demonstrate broad agreement between SO₂ retrievals from independent UV and IR sensors (especially for the larger volcanic eruptions), providing confidence in the measurements. However, we also note that retrieval algorithms are likely to improve in the future due to continued improvements in spectroscopic data, additional constraints from ancillary data and better characterization of uncertainties.



Thomas and Watson (2009) and Carn (2015a) provide recent tabulations of satellite sensor specifications.

3.1. TOMS (UV)

A TOMS instrument was first deployed on the Nimbus-7 (N7) satellite that was operational between October 1978 and May 1993, and N7/TOMS provided the first satellite observation of volcanic SO₂ after the 1982 El Chichón eruption (Krueger, 1983; Krueger et al., 2008). Subsequently, three similar 6-channel TOMS sensors were launched on the Meteor-3 (M3; August 1991–December 1994), ADEOS (AD; September 1996–June 1997) and Earth Probe (EP; July 1996–December 2005) spacecraft. Following the 1982 El Chichón SO₂ cloud sighting, algorithms were developed for quantitative retrievals of SO₂ column amounts (Krueger et al., 1995, 2000). The N7, M3, AD and EP/TOMS instruments differed in their channel placement (after discovery of the volcanic SO₂ mapping capability, AD/TOMS and EP/TOMS wavelengths were optimized for SO₂ measurements; Gurevich and Krueger, 1997) and spatial resolution (24–50 km nadir footprint size), which impacts SO₂ sensitivity: the estimated volcanic SO₂ cloud detection limit ranges from ~2–4 kt (EP/TOMS) to ~11 kt (N7/TOMS; Carn et al., 2003). There was a TOMS data gap of ~19 months in 1995–96 between the M3 and EP/TOMS missions, but we use IR TOVS SO₂ measurements to cover this time period (Prata et al., 2003). The Solar Backscatter Ultraviolet (SBUV or SBUV/2) instruments flown on the N7 satellite with TOMS and later on NOAA polar-orbiting satellites could also detect volcanic SO₂ (e.g., McPeters et al., 1984; McPeters, 1993; Schnetzler et al., 1995), but with limited mapping ability (Table 1), and to date have only been applied to the largest eruptions (El Chichón and Pinatubo).

Most TOMS volcanic SO₂ data reported here have been produced using the iterative 4-band matrix inversion retrieval technique described by Krueger et al. (1995, 2000), using Sun.-normalized radiances produced by the TOMS ozone data production algorithm as input, and effective optical paths derived from a UV radiative transfer model. Krueger et al. (1995) report errors of ± 10–30% for TOMS SO₂ retrievals, highest at large solar zenith angles near the swath edge. Previous TOMS volcanic SO₂ analyses used TOMS production data version 6 (e.g., Bluth et al., 1992, 1993, 1997) or version 7 (Carn et al., 2003), whereas more recent analyses have used version 8 (the current TOMS ozone algorithm is version 9). Some, though not all, older eruptions have been reanalyzed using version 8 TOMS data, with little change from previously reported results (e.g., Krueger et al., 2008). TOMS production algorithm updates are largely focused on improving the accuracy of total ozone retrievals by correcting for the effects of aerosols, clouds and ozone profile variability (e.g., Bhartia and Wellemeyer, 2002), and hence small improvement in SO₂ retrievals may also result. TOMS SO₂ column retrievals are not publicly available at present, but will be released following a major reprocessing of the TOMS data archive that is expected to be complete by 2018 (in the meantime, TOMS SO₂ retrievals are available from the first author upon request). Currently, N7/TOMS and EP/TOMS level 2 (L2) ozone data (version 8) are publicly available from the NASA Goddard Earth Sciences (GES) Data and Information Services Center (DISK; <http://disc.sci.gsfc.nasa.gov/>), and the L2 TOMS ozone data files contain a SO₂ index (SOI) indicating contamination of ozone retrievals by volcanic SO₂.

As is the case for all satellite-based volcanic SO₂ measurements, opportunities to validate TOMS SO₂ data were rare and opportunistic,

relying on the chance overpass of a volcanic cloud over a ground measurement site. The best validation data have been obtained using ground-based UV Brewer spectrophotometers, which can be used to measure total column SO₂ and are operated as networks in several countries (e.g., Fioletov et al., 1998). A Brewer instrument in Toronto (Canada) detected SO₂ (column amounts up to ~100 DU) in the May 1980 Mt. St. Helens (WA, USA) eruption cloud as it drifted east, with general agreement between TOMS and Brewer SO₂ columns (Kerr and Evans, 1987). In 1984, a Brewer in Sweden detected SO₂ (>40 DU) emitted by the September 5, 1984 effusive eruption of Krafla (Iceland); in this case significant disagreement between Brewer and TOMS SO₂ observations was attributed to lower tropospheric SO₂ which was not detected by TOMS (Kerr and Evans, 1987). Perhaps the most robust TOMS SO₂ validation occurred when the Toronto Brewer measured SO₂ in a drifting volcanic cloud from Mt. Spurr (Alaska) in September 1992, with coincident N7/TOMS and Brewer SO₂ columns (~40–50 DU) agreeing to within 20% (Fioletov et al., 1998; Krueger et al., 2000). Attempts to compare TOMS SO₂ mass measurements with ground-based UV correlation spectrometer (COSPEC) SO₂ emission rates were made for the first time during the 1982 Galunggung eruption (Java, Indonesia; Bluth et al., 1994). In this case TOMS and COSPEC observed different emissions (eruptive and non-eruptive, respectively), precluding validation but demonstrating the complementarity of the satellite and ground-based measurements.

3.2. TOVS/HIRS (IR)

The High-resolution Infrared Radiation Sounder (HIRS/2) was first launched as part of the TOVS package on the TIROS-N satellite in October 1978, and subsequent iterations of the HIRS instrument (HIRS/3, HIRS/4) have been flown on all NOAA polar-orbiters and more recently on the European MetOp satellites, providing continuous coverage to the present. Prata et al. (2003) provide further details on TOVS/HIRS and describe a technique for retrieving UTLS SO₂ column amounts from HIRS/2 L1b radiance data, which we use to analyze volcanic eruptions that occurred during the 1995–96 TOMS data gap. However, the full HIRS/2 SO₂ dataset could potentially be used to generate a long-term volcanic SO₂ climatology for 1978–present, with greater temporal sampling than UV sensors, but reduced sensitivity to tropospheric SO₂. Prata et al. (2003) and Carn et al. (2008a) show good agreement between TOMS and HIRS/2 SO₂ retrievals for a small subset of eruptions that have been analyzed to date with both instruments (e.g., Table 2).

3.3. AIRS (IR)

AIRS, in Sun.-synchronous orbit on Aqua in the A-Train, provides hyperspectral IR measurements using 2378 channels at IR wavelengths between ~3.3–16.7 μm (Chahine et al., 2006). This wavelength range covers SO₂ absorption bands at 4 and 7.3 μm, and part of the 8.6 μm band (there is a gap from ~8.2–8.8 μm). The primary goal of AIRS is to provide global atmospheric soundings of temperature, moisture and other gases for numerical weather prediction and climate, but Carn et al. (2005) and Prata and Bernardo (2007) have shown that AIRS can be used to retrieve volcanic SO₂ at altitudes above ~3–5 km using the 7.3 μm band. AIRS scans a swath of ± 49° from nadir and the sensor's elliptical footprint dimensions vary from 15 × 15 km at nadir to

Fig. 2. Examples of A-Train synergy for characterizing volcanic cloud altitude. (A) OMI total column SO₂ and MLS UTLS column SO₂ (W. Read, pers. Comm.) for the April 6, 2005 eruption of Anatahan (CNMI; triangle). MLS retrievals are shown as filled circles along the Aura satellite daytime and nighttime overpass tracks; note that circle size is not representative of the MLS field-of-view. Whilst the nadir OMI measurements detect SO₂ east and west of Anatahan, MLS only detects UTLS SO₂ to the east, confirming the variable altitude of the mapped SO₂. (B) Composite of nighttime A-Train measurements of the Chaitén volcanic plume on May 7, 2008 at ~4:47 UTC. Main panel shows 532 nm total attenuated backscatter ($\text{km}^{-1} \text{sr}^{-1}$) from CALIOP, clearly showing aerosol in the layered volcanic cloud, overlain with reflectivity from the CloudSat Cloud Profiling Radar (CPR), which in this case only detects hydrometeors in the tropospheric meteorological clouds (indicating that the volcanic aerosol particle size is below the radar detection limit, i.e., sub-mm). White stars show locations of SO₂ detection by AIRS based on a 2-channel brightness temperature difference (BTD in K). The sub-horizontal white line indicates the tropopause altitude. MLS retrievals of SO₂ (ppbv), HCl (ppbv) and ice-water content (IWC; mg m⁻³) for several pressure levels (indicated top left in hPa) are plotted above the CALIOP data; these show clear enhancements in the volcanic cloud above 215 hPa.

18×40 km at the swath edge. A full description of the AIRS instrument and operational data processing can be found at <http://airs.jpl.nasa.gov>.

IR AIRS SO₂ retrievals used here are derived using the Level 1B AIRS IR geolocated and calibrated radiance product (AIRIBRAD version 5) and the Prata and Bernardo (2007) algorithm. AIRIBRAD data granules are publicly available from the NASA GES DISK (<http://disc.sci.gsfc.nasa.gov/>). The Prata and Bernardo (2007) algorithm is also used to produce NRT AIRS SO₂ products for aviation hazard mitigation, which can be obtained via the NASA GES DISK (<http://disc.sci.gsfc.nasa.gov/nrt/data holdings/airs-nrt-products>) and viewed on the Support to Aviation Control Service (SACS) website (<http://sacs.aeronomie.be/nrt>; Brenot et al., 2014). Prata and Bernardo (2007) quote a retrieval accuracy (or detection limit) of 6 DU SO₂ ($\sim 1.6 \times 10^{17}$ molecules cm⁻²) for a single AIRS pixel retrieval, mostly due to errors in accounting for background atmospheric water vapor and clouds. Thomas et al. (2011) report a similar AIRS SO₂ detection limit of 5 DU. A detection limit of 6 DU equates to ~ 30 –100 tons of SO₂ within a single AIRS footprint, depending on swath position, and assuming that a cluster of ~ 10 pixels would constitute a statistically significant volcanic cloud yields a minimum detectable SO₂ mass of ~ 0.3 –1 kt (kt) in the UTLS. In practice, minimum reported volcanic SO₂ mass loadings derived from AIRS (for SO₂ clouds consisting of multiple pixels) are ~ 0.3 –10 kt depending on eruption latitude (e.g., Prata and Bernardo, 2007; Carn and Prata, 2010; Bonny et al., in preparation), indicating slightly higher sensitivity to UTLS SO₂ than EP/TOMS in the 2002–2004 period of overlap between AIRS and TOMS. We also exploit the synergy between near-coincident AIRS 7.3 μm partial column SO₂ retrievals and OMI total column retrievals from the A-Train to infer volcanic SO₂ altitude, where OMI \approx AIRS > 0 implies UTLS SO₂, OMI > AIRS > 0 implies a vertically extensive SO₂ cloud straddling the lower troposphere and UTLS (or alternatively, a multi-layered volcanic cloud with layers residing at multiple altitudes), and OMI > AIRS \approx 0 implies SO₂ confined to the lower troposphere (e.g., Carn et al., 2009b).

To date, efforts to validate AIRS SO₂ measurements have focused on inter-comparisons with UV SO₂ retrievals from TOMS, OMI and OMPS. Good correlations have been found between AIRS and UV SO₂ burdens for volcanic clouds at high altitude (>8 km; e.g., Fig. 3; Prata and Bernardo, 2007; Carn and Prata, 2010; Thomas et al., 2011; Carn et al., 2015). The weak sensitivity of AIRS to lower tropospheric SO₂, along with incomplete coverage by the AIRS swath for some eruptions, results in a poorer correlation for eruptions to lower altitudes (Prata and Bernardo, 2007). As yet there have been few detailed comparisons between IR and UV SO₂ column retrievals at the pixel scale, principally due to problems with spatial and temporal collocation and typically non-uniform SO₂ distributions in volcanic clouds (e.g., Kearney et al., 2009; Carn et al., 2015).

3.4. OMI (UV)

OMI is a hyperspectral UV–Visible spectrometer (270–500 nm) using a two-dimensional charge-coupled device (CCD) detector to collect simultaneous spectral and spatial measurements of backscattered solar radiation across a 2600 km swath (Leveld et al., 2006). Spatial resolution is 13 \times 24 km (along-track \times across-track) at nadir, increasing to $\sim 28 \times 160$ km at the swath edge. The OMI UV-2 channel (307–383 nm) used for SO₂ retrievals has an average spectral resolution of 0.45 nm (Leveld et al., 2006), and this high spectral resolution coupled with the small OMI footprint size provides unprecedented SO₂ sensitivity for a satellite instrument, permitting detection of passive volcanic degassing and anthropogenic pollution (e.g., Carn et al., 2007, 2008b, 2013). OMI became fully operational in September 2004 and provided daily contiguous, global daytime mapping of ozone, SO₂ and other trace gases (NO₂, BrO, HCHO) until May 2008, when a blockage appeared in the sensor's field of view. This 'row anomaly' evolved dynamically (generally worsening) over time to obscure a significant proportion of the OMI swath from nadir to the east (see: <http://www.knmi.nl/omi/research/>

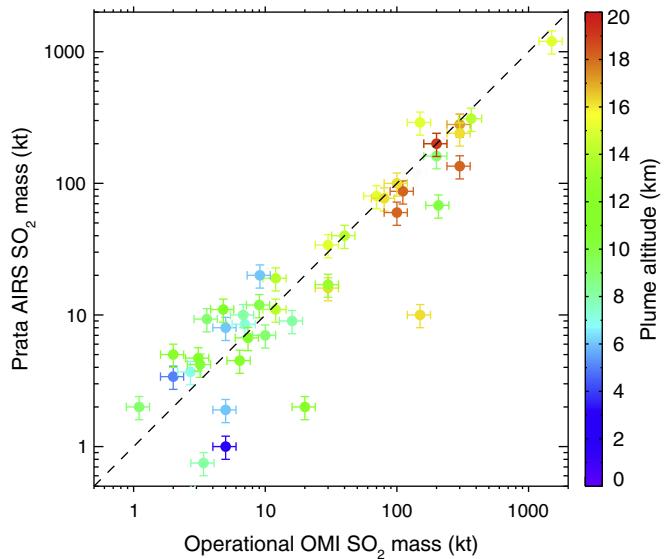


Fig. 3. SO₂ mass loadings (kiltons; log scale) derived from the operational OMI Linear Fit SO₂ algorithm (Yang et al., 2007) against those derived from the Prata and Bernardo (2007) AIRS SO₂ algorithm for selected volcanic eruptions in the 2004–2014 period. Eruptions are color-coded by estimated plume altitude (see text for discussion of sources) and 20% error bars are shown. Dashed line is the 1:1 line. Note broad correlation between OMI and AIRS SO₂ retrievals with greater scatter for smaller eruptions and/or lower altitude plumes, as expected.

[product/rowanomaly-background.php](http://rowanomaly-background.php)), ending daily contiguous coverage at low latitudes, although data unaffected by the anomaly remain of high quality. Future evolution of the row anomaly is hard to predict.

Several algorithms have been used to retrieve SO₂ from OMI UV radiances, with some differences in SO₂ column sensitivity and retrieval dynamic range (e.g., Krotkov et al., 2006; Yang et al., 2007, 2009a,b, 2010; Lee et al., 2009; Li et al., 2013; Carn et al., 2013; Theys et al., 2015). Most of the volcanic SO₂ measurements in the current inventory are derived from the Linear Fit (LF) algorithm (Yang et al., 2007), which has been used to generate the operational, publicly available OMI SO₂ product (OMSO2 collection 3; available from the NASA GES DISC). The LF algorithm uses 10 OMI wavelengths at 310–360 nm, covering regions of strong (shorter wavelengths) and weak (longer wavelengths) SO₂ absorption, to retrieve SO₂ column amounts (Yang et al., 2007). By using longer UV wavelengths, the LF algorithm avoids some of the non-linear saturation effects that limit algorithms using shorter wavelengths to lower SO₂ column amounts (~ 10 –20 DU; Krotkov et al., 2006). However, the use of a zero SO₂ a-priori constraint as a linearization point also limits the LF algorithm to column amounts of ~ 100 DU (Yang et al., 2007), and hence offline iterative algorithms have been developed to retrieve higher SO₂ columns (Yang et al., 2009a). Such high SO₂ column amounts (>100 DU) are typically only encountered in very fresh volcanic eruption clouds (<1 day old), after which SO₂ columns are rapidly depleted by upper atmospheric wind shear (e.g., Krueger et al., 2008), or close to the vent during SO₂-rich effusive eruptions (e.g., Yang et al., 2009a). With the exception of highly concentrated SO₂ plumes from effusive eruptions (>1000 DU; Yang et al., 2009a), the highest SO₂ columns detected from space in UTLS volcanic clouds to date were measured by N7/TOMS ~6 h after the April 4, 1982 El Chichón eruption (~600 DU; Krueger et al., 2008) and ~21 h after the paroxysmal June 15, 1991 eruption of Pinatubo (~540 DU; Guo et al., 2004). No explosive eruption comparable in SO₂ release to El Chichón or Pinatubo has occurred during the OMI mission, but iterative SO₂ retrieval techniques (e.g., Yang et al., 2009) would certainly be required for accurate SO₂ mass estimation in such cases. For comparison, peak SO₂ column amounts of 200–300 DU were measured by OMI in the August 2008 Kasatochi volcanic cloud, among the largest to have occurred since Pinatubo (Krotkov et al., 2010). In the current

volcanic SO₂ inventory, most eruptions have been analyzed using the operational LF algorithm, with several of the larger explosive eruptions (e.g., Okmok and Kasatochi in 2008; Nabro and Grimsvötn in 2011) also analyzed using offline algorithms to check for SO₂ column saturation. Other consistency checks, such as monitoring the temporal evolution of SO₂ loading in the days following an eruption, checking coincident ozone fields for artifacts (anomalies in ozone retrievals indicate that SO₂ absorption has been incorrectly accounted for), and comparing with IR SO₂ retrievals (e.g., Fig. 3), are also routinely used.

SO₂ retrieval algorithms require specification of SO₂ altitude due to the temperature- and pressure-dependence of SO₂ absorption and the air mass factor (the ratio of slant to vertical column density). In the LF algorithm, and others, this is achieved by retrieving separate SO₂ column amounts for three prescribed SO₂ vertical profiles representing different volcanic degassing styles (e.g., Yang et al., 2007; Rix et al., 2012; Carn et al., 2013; Theys et al., 2015). The vertical profiles used are (Carn et al., 2013): SO₂ in the lower troposphere (TRL; 0–5 km altitude with center of mass altitude (CMA) of 2.5 km); SO₂ in the mid-troposphere (TRM; 5–10 km altitude, CMA = 7.5 km) and SO₂ in the lower stratosphere (STL; 15–20 km altitude; CMA = 17 km). Corresponding styles of volcanic degassing are passive emissions in the lower troposphere (TRL), high-altitude volcanic degassing and/or small-to-moderate eruptions (TRM) and large eruptions with stratospheric injection (STL). Our approach for volcanic eruption analysis is to use available information on plume altitude (see Section 2, and below) to select the appropriate SO₂ CMA, or interpolate between CMAs for intermediate altitudes. Information on altitude can include direct SO₂ altitude retrievals using OMI measurements (e.g., Yang et al., 2009b, 2010). Provided that SO₂ column saturation is avoided and the assumed SO₂ vertical profile is approximately correct, LF SO₂ retrievals have an uncertainty of ~20% (Yang et al., 2007).

In addition to SO₂ mass, we also use an OMI SO₂ index (OSI) calculated from OMI ozone algorithm (OMTO3) residuals (Krotkov et al., 2006) to identify volcanic SO₂ emissions (e.g., Fig. 4). We use the OSI as a proxy for volcanic SO₂ to avoid complications related to calculation of SO₂ mass when SO₂ from multiple sources at different altitudes may be present (due to the difficulty of isolating SO₂ at different altitudes), a common problem in UV measurements which are sensitive to SO₂ anywhere in the atmospheric column in cloud-free conditions (e.g., Yang et al., 2007). This is less of an issue with IR SO₂ retrievals using the 7.3 μm band since these effectively screen out any lower tropospheric SO₂.

OMTO3 residuals are calculated using scaled, normalized reflectances (N-values) where N-values are defined as:

$$N_{\lambda} = -100 \log \left(\frac{I_{\lambda}}{F_{\lambda}} \right) \quad (1)$$

where I is the backscattered Earth radiance and F is the solar irradiance at wavelength λ . N-value residuals for a pair of wavelengths (res_j) are computed thus:

$$res_j = N_j^{measured} - N_j^{calculated} \quad (2)$$

$$N_j = N(\lambda_j^{short}) - N(\lambda_j^{long}) \quad (3)$$

where calculated N-values are derived from a forward radiative transfer model, accounting for Rayleigh scattering, ozone absorption, Ring effect (Raman scattering) and surface reflectivity, but not SO₂ absorption. The subscript j ($j = 1, 2, 3$) denotes wavelength pairs where pairs 1, 2 and 3 (P1, P2 and P3) correspond to wavelengths of 311.9–310.8 nm, 313.2–311.9 nm and 314.4–313.2 nm, respectively. These wavelength pairs represent maxima and minima in the UV SO₂ absorption cross-section (Krotkov et al., 2006; Carn et al., 2013) and this wavelength placement permits detection of SO₂ contamination (independent of

SO₂ column retrieval) if the following relationships are simultaneously satisfied (Carn et al., 2013):

$$\begin{aligned} res_{P1} &> 0 \\ res_{P2} &< 0 \\ res_{P3} &> 0 \end{aligned} \quad (4)$$

The OSI is defined using the N-value pair residuals as follows:

$$OSI = res_{P1} + abs(res_{P2}) + res_{P3} \quad (5)$$

The value of the OSI thus increases with the severity of SO₂ contamination (higher SO₂ column amount and/or SO₂ altitude), but since it uses short UV wavelengths it also saturates at a certain point for large eruptions. Here, we use the OSI as a semi-quantitative indication of volcanic SO₂ emissions and also to detect sources of passive volcanic degassing. In the latter case saturation is not an issue due to the much lower SO₂ column amounts and plume altitudes involved.

There have been several efforts to validate OMI SO₂ retrievals, including inter-comparisons with IR satellite measurements (e.g., Carn and Prata, 2010), ground-based SO₂ measurements (e.g., Spinei et al., 2010; Lopez, 2011; Theys et al., 2015; Lalongo et al., 2015) and airborne in-situ measurements (e.g., Krotkov et al., 2008; Carn et al., 2011). Results suggest that OMI SO₂ measurements are generally consistent with independent retrievals in dilute (aged) volcanic clouds, but validation of the very high SO₂ column amounts present in fresh volcanic eruption clouds remains a challenge, mainly due to the absence of ground-based or in-situ data in such situations.

To detect sources of passive SO₂ degassing using the OSI we have adopted the following procedure, which exploits the fact that the locations of all subaerial volcanoes (i.e., the sources of passive SO₂ emissions) are known and cataloged in the VOTW database. Detection and attribution of small, non-eruptive volcanic SO₂ plumes also requires sufficient spatial resolution to identify the source, which OMI provides in most cases although ambiguity can still arise in regions with several actively degassing volcanoes. For each day of global OMI SO₂ measurements, all pixels with identifiable SO₂ contamination (based on Eq. (4) above) are extracted from the OMSO2 data products and ranked based on their OSI values. In doing this, we attempt to exclude pixels affected by the South Atlantic radiation Anomaly (SAA) over South America (which have distinctive N-value residuals) and also exclude pixels with solar zenith angles >65° to reduce noise. Then, for each pixel we identify the closest active volcano within a 50 km radius of the pixel center coordinates using a modified version of the VOTW catalog, which is assigned as the presumed source of the detected SO₂; the 50 km distance threshold is arbitrary, but further afield the source of detected SO₂ becomes increasingly ambiguous. We acknowledge that there are uncertainties associated with this approach (e.g., the possible presence of more than one volcano within 50 km of the anomaly), but we attempt to mitigate these by cross-checking with available activity reports to exclude unlikely sources. Also, the modified volcano list used here includes the most likely potential sources of SO₂ emissions based on historical activity, and is necessary to facilitate source attribution in some regions (note that this modification does not impact SO₂ detection, only source identification). Once sources have been assigned to all the SO₂ contaminated pixels, the unique sources are identified (eliminating multiple detections of the same SO₂ plume; e.g., where multiple SO₂ contaminated pixels occur within a SO₂ plume from a single source volcano) and cataloged for each day. By repeating this process for the lifetime of the OMI mission we acquire an inventory of the dominant sources of passive SO₂ emissions detected from space. The analysis presented here is based on OMI SO₂ observations from the beginning of the mission (6 September, 2004) to the end of 2014, or ~3700 days of SO₂ measurements.

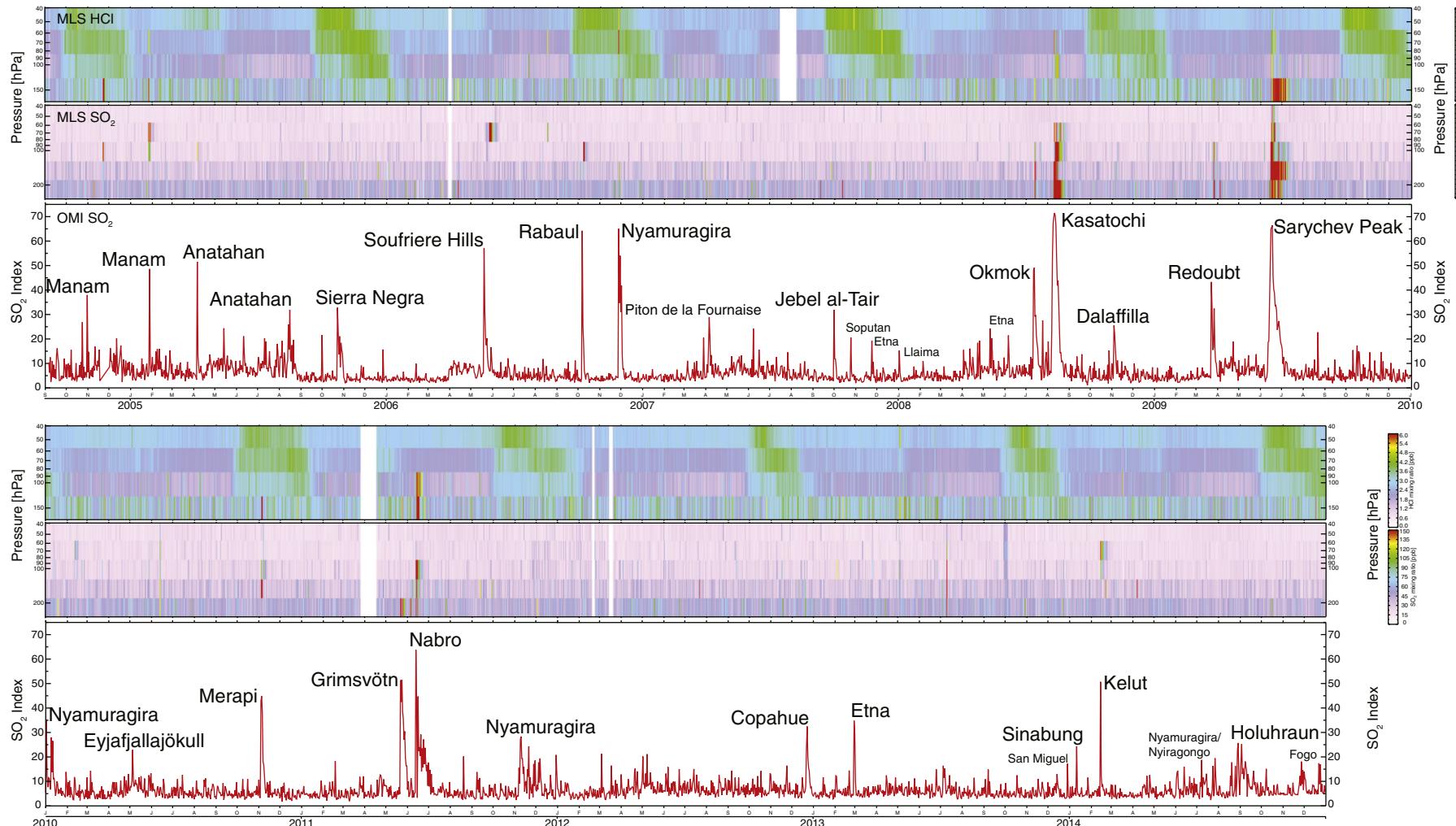


Fig. 4. Time-series of maximum daily OMI SO₂ index (OSI) and MLS UTLS SO₂ and HCl measurements from September 2004 to December 2014. OMI panels (red curve) show the maximum daily value of the OSI recorded in global OMI measurements. The OSI is sensitive to SO₂ loading and altitude (see text for definition). Some of the prominent volcanic eruptions responsible for OSI excursions are labeled. MLS panels show the maximum retrieved UTLS SO₂ and HCl mixing ratios on a pressure grid extending from 215 hPa (~11 km altitude) to 40 hPa (~20–21 km altitude) for SO₂ and from 150 hPa (~14 km altitude) to 40 hPa for HCl (note that although MLS HCl measurements are deemed invalid for scientific use below 100 hPa (Livesey et al., 2011), and clearly biased high at 147 hPa, it is clear that volcanic HCl is detected at this level). Note that noise in MLS measurements increases with pressure (i.e., towards lower altitudes) and the data shown here do not represent the full vertical extent of MLS measurements. Blank regions are MLS data gaps. MLS data have been screened using the criteria recommended by Livesey et al. (2011).

3.5. MLS (microwave) and IR limb sounders

The first MLS instrument was deployed on NASA's Upper Atmosphere Research Satellite (UARS) in September 1991, shortly after the Pinatubo and Cerro Hudson eruptions, and continued to operate until August 2001 but with only intermittent measurements after December 1993 (<http://mls.jpl.nasa.gov/uars/>). UARS MLS detected the tropical stratospheric SO₂ veil produced by the Pinatubo eruption (Read et al., 1993) and the large April 1993 eruption of Lascar volcano (Chile; http://mls.jpl.nasa.gov/joe/so2_lascar.html). UARS MLS data were used to constrain the altitude (~26 km), SO₂ decay e-folding time (~29–41 days) and total SO₂ mass (~17 Tg) of the Pinatubo SO₂ cloud (Read et al., 1993). However, UARS MLS data have not been analyzed in detail for this work. The second MLS was launched on Aura (with OMI) in July 2004 (Waters et al., 2006) and continues to operate at the time of writing. Aura/MLS measures microwave thermal emission from Earth's limb to retrieve vertical profiles of trace gas mixing ratios in the UTLS; its great value for volcanic gas detection is its ability to provide vertical profiles of SO₂ and HCl (and also ice water content; e.g., Fig. 2) coincident with nadir OMI and AIRS measurements from the A-Train. Hence we use daily Aura/MLS limb soundings of SO₂ and HCl to pinpoint UTLS injections of these species since September 2004 (e.g., Figs. 2, 4; Pumphrey et al., 2015). The main weakness of MLS is poor spatial sampling: profiles (with vertical resolution of ~3 km) are retrieved only along the sub-satellite Aura orbit track with horizontal spacing of ~167 km (Pumphrey et al., 2015). Hence smaller volcanic eruptions may not be sampled, but these are also less likely to reach the UTLS. We use version 3.3 of the level 2 MLS Daily SO₂ and HCl products, collection 3 (ML2SO2.003 and ML2HCL.003; Froidevaux et al., 2008). ML2SO2 and ML2HCL data products are available from the NASA GES DISK (<http://disc.gsfc.nasa.gov/>). Further details on MLS and SO₂ retrieval procedures are given by Pumphrey et al. (2015), and Livesey et al. (2011) provide information on the precision, accuracy and useful vertical range of MLS SO₂ and HCl retrievals, which are 3 ppbv, 5–20 ppbv and 215–10 hPa for SO₂ and 0.2–0.6 ppbv, 0.2–0.4 ppbv and 100–0.32 hPa for HCl, respectively. We note, however, that the very strong perturbations to SO₂ and HCl mixing ratios caused by volcanic eruptions can permit detection in MLS data at pressure levels outside the stated useful range.

In addition to MLS data, information on SO₂ vertical profiles in volcanic clouds for some eruptions has also been supplied by other limb sounding instruments (Table 1) such as MIPAS (2002–2012; Höpfner et al., 2013, 2015) and ACE (2003–present; Doeringer et al., 2012), and by Aura/TES (a nadir hyperspectral sounder; Clerbaux et al., 2008). MIPAS and ACE are both Fourier Transform Spectrometer (FTS)-based sensors operating in the TIR using the 7.3 μm SO₂ band, but MIPAS detected limb thermal emission whilst ACE uses solar occultation twice daily (sunrise and sunset). Both MIPAS and ACE data can suffer from poor spatial and temporal sampling (hence identification of the source of volcanic SO₂ perturbations can be ambiguous; e.g., Brühl et al., 2015); for example, Höpfner et al. (2013) analyzed monthly mean MIPAS limb spectra binned within 10°-wide latitude bands in order to detect small amounts of SO₂ in the upper stratosphere. Höpfner et al. (2015) analyzed single MIPAS limb scans and identified over 30 volcanic SO₂ injections into the UTLS between 2002 and 2012, some of which were from unidentified sources. However, our database of global volcanic SO₂ emissions based on nadir satellite observations can be used to pinpoint these sources: e.g., tropical UTLS injections of SO₂ from an unidentified source detected by MIPAS in July–August 2005 (Höpfner et al., 2015) can be confidently ascribed to eruptions of Anatahan (CNMI; Fig. 4).

3.6. IASI (IR)

IASI is a nadir-viewing Fourier-transform IR (FTIR) spectrometer first launched on the polar-orbiting MetOp-A satellite on 19 October 2006 (Clerbaux et al., 2007; Phulpin et al., 2007). A second, identical

IASI instrument was launched on MetOp-B on 17 September 2012. Both IASI instruments provide global coverage twice a day, with a nadir footprint size of 12 km (diameter) and a full swath width of 2200 km. The MetOp satellites are operational platforms; hence the IASI measurements are available in near real time. The IASI spectrometer provides continuous spectral coverage from 645 cm⁻¹ to 2760 cm⁻¹ (~3.6–15.5 μm) with no gaps (in contrast to AIRS), with full coverage of the ν₁, ν₃ and ν₁ + ν₃ SO₂ absorption bands. IASI measurement noise is also lower than AIRS; the IASI noise-equivalent temperature change (NEΔT) at 280 K is estimated to be 0.05 K in the ν₃ band and 0.12 K in the ν₁ band, significantly better than the expected 0.2–0.35 K. For comparison, the AIRS NEΔT is ~0.2 K (but note that this is at a scene temperature of 250 K; at the warmer temperature of 280 K quoted for IASI the AIRS NEΔT will be higher) and it only partially covers the ν₁ band (Carn et al., 2005).

IASI SO₂ retrievals (Figs. 7, 8) employ the technique described by Clarisse et al. (2012), using an SO₂ altitude retrieval algorithm (Clarisse et al., 2014) to derive accurate plume altitudes and hence more reliable estimates of SO₂ mass loadings. Inaccurate SO₂ loadings may still result if the plume altitude is incorrectly retrieved. Furthermore, as for most IR 7.3 μm SO₂ retrievals, the IASI SO₂ measurements are only reliable for SO₂ at altitudes above ~5 km (in mid-latitudes) or ~7 km (in the tropics) due to water vapor interference. SO₂ plumes at lower altitudes will be missed or mass loading will be underestimated (this could be mitigated in the future by exploitation of IASI data in the ν₁ SO₂ band). This is a pertinent issue for relatively long-lived eruptions with temporally variable plume altitudes, such as the 2010 Eyjafjallajökull, 2011 Nabro and 2014–15 Holuhraun eruptions.

3.7. Volcanic activity and eruption altitude information

Information on eruption occurrence, timing and VEI is derived from the Volcanoes of the World (VOTW) database curated by the Smithsonian Institution Global Volcanism Program (GVP; <http://www.volcano.si.edu>; Global Volcanism Program, 2013). The VOTW database is updated frequently (see http://www.volcano.si.edu/gvp_votw.cfm for latest updates) but for this analysis we have used VOTW v4.3.0, which includes eruptions through June 2014 (Global Volcanism Program, 2013). It is important to note that VEIs provided in the VOTW database (particularly for more recent eruptions) are assigned based on the best available information at the time (e.g., plume altitude, estimated eruption volume) and hence are subject to some uncertainty. Furthermore, a single VEI cannot capture any variability of explosive activity during long-lived eruptions; in such cases the assigned VEI corresponds to the largest event recorded in given time period.

The altitude of volcanic SO₂ injection determines its climate impact, and hence our database includes estimates of SO₂ altitude for all eruptions (Fig. 1). As discussed above, altitude estimates can be derived from several sources, with more frequent measurements of volcanic plume altitude available since 2002 (MIPAS), 2004 (MLS) and particularly since June 2006 (CALIOP and CloudSat). CALIOP, launched in April 2006 on the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite (Winker et al., 2009), has been of tremendous value to the volcano remote sensing community with its ability to determine precisely the altitude of aerosols and clouds (e.g., Vaughan et al., 2009). CALIOP browse and expedited browse imagery products produced at NASA Langley Research Center (<http://www.calipso.larc.nasa.gov/products/>) are routinely checked during eruptions for evidence of plume detection. Lidar backscatter data in conjunction with depolarization ratios (indicating aerosol phase) and color ratios (indicating relative particle size) can be used to confidently distinguish volcanic plumes from meteorological clouds (e.g., Winker et al., 2012), especially when matched with coincident nadir satellite observations from the A-Train (e.g., OMI or AIRS SO₂ data; Carn et al., 2009a, 2009b; Vernier et al., 2013). Although CALIOP detects aerosol particles and not SO₂ gas, it is often assumed that aerosols and SO₂ will be

approximately collocated (horizontally and vertically). Clarisse et al. (2014) found good agreement (within ~2 km in the vertical) between IASI SO₂ altitude retrievals and CALIOP aerosol layer altitudes for the 2011 Nabro eruption cloud. However, collocation may not always be a valid assumption and some separation between gas and aerosol phases is expected due to different sedimentation rates. The poor spatial sampling of CALIOP along the A-Train orbit track often precludes detection of fresh volcanic eruption plumes (~1–3 h old; where ash and hydrometeors may dominate; e.g., Rose et al., 2000) until they expand in size in subsequent days, although the February 2014 eruption of Kelut (Java, Indonesia) provided a rare CALIOP profile through an explosive eruption column only ~2 h after emission (Kristiansen et al., 2015). The Cloud-Proiling Radar (CPR) on the CloudSat satellite complements CALIOP with millimeter-wave (W-band; 3.2 mm wavelength) observations of larger cloud particles and precipitation, but is only sensitive to very fresh volcanic eruption clouds with high loadings of coarse ash and hydrometeors; to date very few confirmed CPR detections of volcanic clouds have been reported but there is evidence that a young plume (~4 h old) from Redoubt (Alaska) was detected on March 26, 2009 (S.A. Carn, unpublished data). Unfortunately, no CloudSat data are available for the February 2014 Kelut eruption. More typically, CALIOP samples volcanic plumes several hours after eruption at the earliest, but for the larger eruptions CALIOP observations can continue for many days or weeks as SO₂ gradually converts to sulfate aerosol and/or fine volcanic ash persists (e.g., Eckhardt et al., 2008; Shibata and Koulotsu, 2008; Carn et al., 2009a, 2009b; Spinei et al., 2010; Tulet and Villeneuve, 2011; Lopez, 2011; Vernier et al., 2011; Winker et al., 2012; Wang et al., 2013; Fromm et al., 2014; Clarisse et al., 2014; Carn et al., 2015). CALIOP observations of volcanic clouds since 2006 have also revealed the complex, multi-layered structure of some plumes (Fig. 2; e.g., Carn et al., 2009a), which has implications for the altitude assumptions made in nadir satellite SO₂ retrievals.

SO₂ altitude retrievals from nadir UV or IR satellite observations (e.g., Yang et al., 2009b, 2010; Nowlan et al., 2011; Rix et al., 2012; Carboni et al., 2012; Clarisse et al., 2014) offer the advantage of much better spatial sampling than CALIOP, although nadir-viewing satellite measurements are sensitive to a range of altitudes rather than a specific level; hence the retrieved SO₂ altitude typically represents the peak of a larger vertical distribution. SO₂ altitude retrievals require a minimum SO₂ column amount but are now routinely performed using IASI data (Fig. 7; e.g., Clarisse et al., 2014), although to date UV SO₂ altitude retrievals have only been applied to a small subset of eruptions (Yang et al., 2009b, 2010; Rix et al., 2012).

Prior to 2004–2006, and for many smaller eruptions since then which go undetected by CALIOP and/or provide insufficient signal for SO₂ altitude retrievals, the main sources of information on eruption altitude are Smithsonian GVP volcanic activity reports (which include pilot reports, satellite observations, and ground-based observations) and trajectory modeling.

4. Results and discussion

In presenting the results of our analysis, it is important to stress the ephemeral nature of any database pertaining to active volcanic processes. In essence, this work provides a snapshot of a dynamic inventory that is being continually revised as new eruptions occur or old events are reanalyzed (Carn, 2015c). We stress however, and demonstrate below, that the largest eruptions have been subject to greater scrutiny and are well-characterized using numerous satellite instruments; it is the smaller eruptions that are the source of most uncertainty. For larger eruptions, it may be possible to validate satellite SO₂ measurements using the mass of sulfate aerosol produced; e.g., for the 1982 El Chichón eruption the TOMS estimate of SO₂ loading is consistent with independent estimates of the mass of sulfate aerosol produced by SO₂ oxidation (Krueger et al., 2008). However, this is not possible for smaller eruptions. Although many eruptions are analyzed in close to real-time as they occur, providing preliminary estimates of SO₂ loading, post-

analysis of eruptions may continue many years or decades after the event as new retrieval algorithms are developed (e.g., Guo et al., 2004; Krueger et al., 2008). Table 2 lists many of the eruptions that have been subject to more detailed analysis (typically the larger events); this invariably results in more accurate estimates of SO₂ loadings and uncertainties. We therefore acknowledge that significant uncertainty remains on SO₂ loadings for some eruptions; estimates could potentially change by 50–100% in some cases though order of magnitude changes are considered extremely unlikely.

4.1. Eruptive degassing

4.1.1. General trends in eruptive degassing

An overview of the entire eruptive volcanic SO₂ inventory (Carn, 2015c) is given in Fig. 1, with more detailed time-series of OMI (September 2004–December 2014), AIRS (January 2003–December 2014) and IASI (October 2007–December 2014) SO₂ data shown in Figs. 4, 5, 6 and 7, and a map of average IASI SO₂ columns depicting the geographical extent of detected volcanic SO₂ plumes shown in Fig. 8. Fig. 9 shows a plot of cumulative SO₂ emissions against time for different eruption styles. In total, the database currently contains SO₂ measurements for ~700 eruptions or eruptive events, with a cumulative SO₂ emission of ~101 Tg. Figs. 1 and 9 display the main trends in eruptive SO₂ emissions observed since 1978, namely the large perturbations associated with the 1982 El Chichón and 1991 Pinatubo eruptions, a significant decline after Pinatubo until the late 1990s, followed by a general increase throughout the 2000s, with peaks in 2005, 2008 and 2011, and reduced emissions in 2012–14. Fig. 9 shows possible evidence for an increase in the rate of SO₂ production (primarily from explosive eruptions) beginning in 2004–2005, after a lull between the mid-1990s and early 2000s, although we interpret this cautiously since the change in slope corresponds to the onset of more sensitive SO₂ measurements by OMI, and hence may simply reflect an increased rate of eruption detection (the mean detection rate increases from 8 eruptions/year in 1978–2004 to ~40 eruptions/year in 2004–2014). In contrast, the time-averaged flux of SO₂ from effusive eruptions has been less variable over the past 3–4 decades, although a possible change in slope occurred in the early 2000s (Fig. 9), which may be partly related to an increased magma output rate at Nyamuragira (DR Congo) in this timeframe (e.g., Wadge and Burt, 2011).

No major eruptions were reported during the 1995–96 TOMS data gap (Fig. 1), but we have assessed several smaller events for SO₂ emissions using HIRS/2 data. An effusive eruption of Fernandina (Galápagos Islands) in January–March 1995 was not detected by HIRS/2 (Head, 2005), indicating that SO₂ emissions were confined to the lower troposphere. Some of the largest eruptions of the 1995–96 period occurred at Ruapehu (New Zealand), with significant SO₂ degassing suggested by ground-based COSPEC measurements (Christenson, 2000) and eruption plume altitudes of up to ~9 km (Prata and Grant, 2001), which is close to the tropopause in the mid-latitude southern hemisphere winter. However, Prata et al. (2003) report relatively minor SO₂ loadings of ~0.01–0.03 Tg for the June 17, 1996 eruptions. Analysis of HIRS/2 data for other eruptions in 1995–96 has not revealed any significant UTLS SO₂ emissions. We therefore conclude that the apparent decline in total volcanic SO₂ emissions between 1994 and 1996 (Fig. 1) is genuine; an inference that is also supported by the lack of major SAOD perturbations in that period (Fig. 9).

The latitudinal distribution of eruptive SO₂ emissions in 1978–2014 (Fig. 1) simply reflects the geographic distribution of active volcanoes, which has clusters at 0°–10°S (including Ecuador, the Galapagos islands, DR Congo, Indonesia and Papua New Guinea), 10°–20°N (including Mexico, central America, the West Indies, Afar, the Philippines, the Marianas, and Hawaii), and 50°–60°N (including Kamchatka, the northern Kuriles and the Aleutian islands), with a smaller peak at 30°–40°N (including Sicily, Italy and Japan; Siebert et al., 2010). Regular eruptive activity in the equatorial region is evident, largely due to frequent

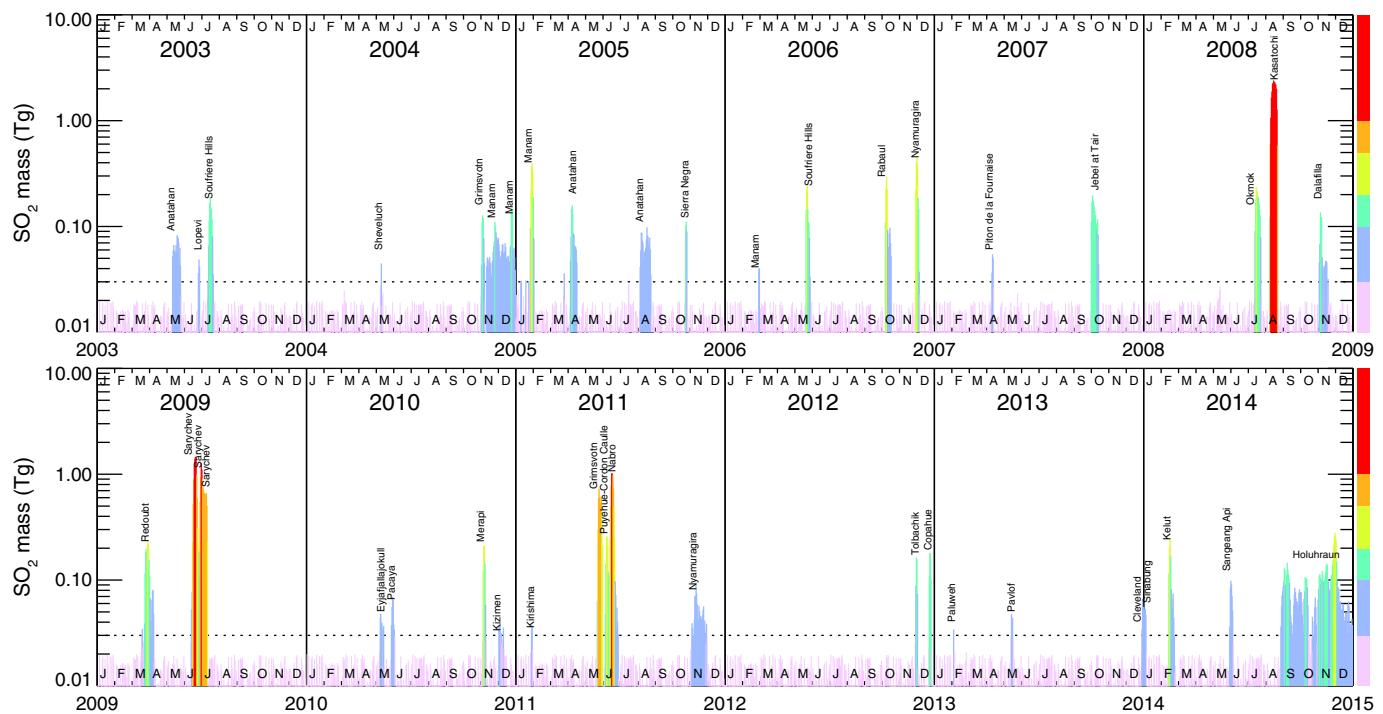


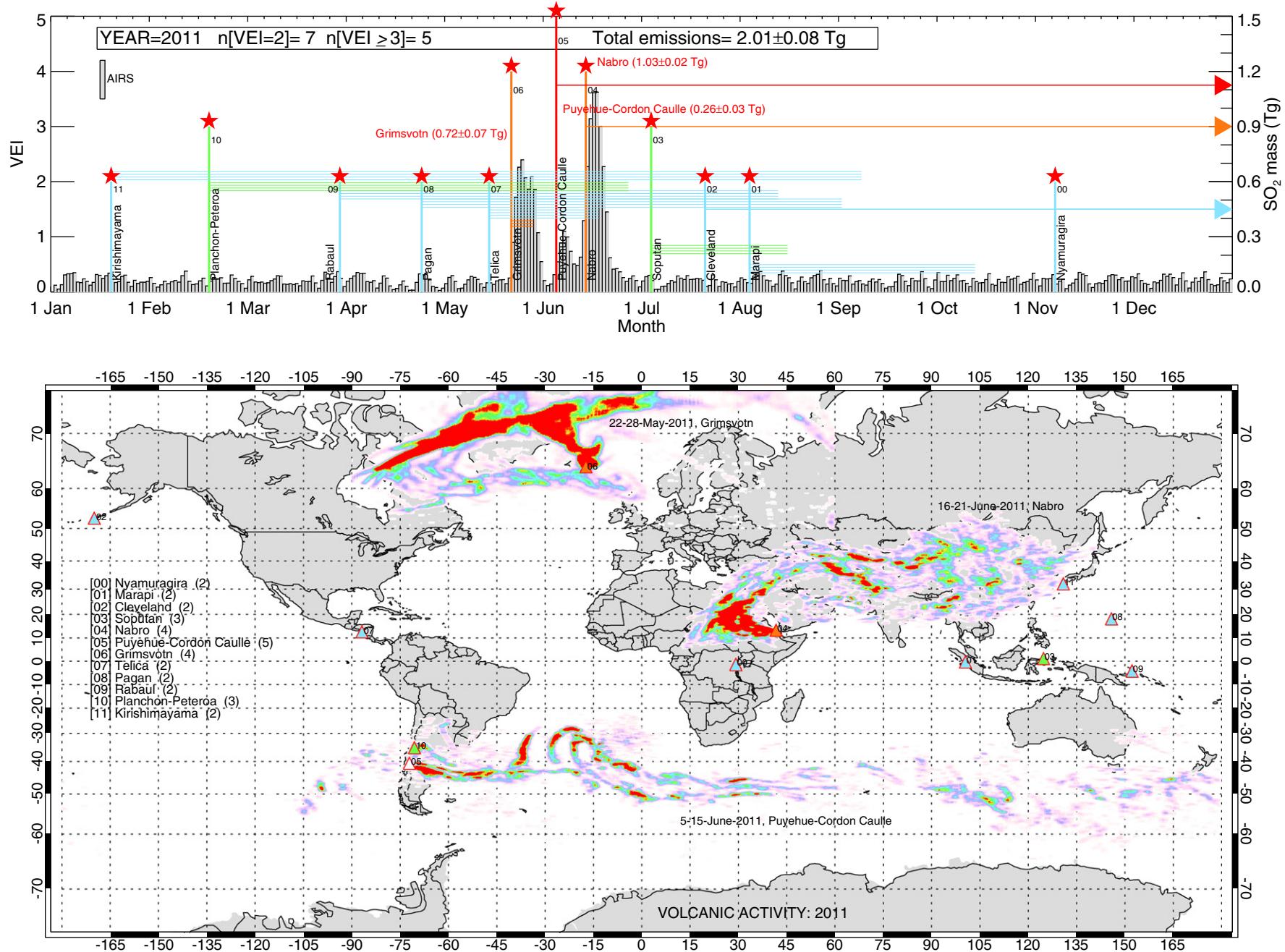
Fig. 5. Time-series of UTLS SO₂ emissions (Tg, log scale) detected by AIRS in 2003–2014 using the Prata and Bernardo (2007) SO₂ retrieval technique. Sources of the observed SO₂ emissions are indicated. SO₂ loadings are also colored based on the color bar to the right of the plot.

effusive eruptions of Nyamulagira (DR Congo; Carn and Bluth, 2003; Bluth and Carn, 2008; Smets et al., 2014) and the Galápagos Islands volcanoes (Head, 2005; Carn, 2015b). The spate of increased activity in southern Chile since 2008 (eruptions of Llaima, Chaitén, Cordon-Caulle, Copahue, and Calbuco) is also noteworthy in the otherwise less active extra-tropical southern hemisphere. The general dearth of southern hemisphere eruptions contrasts with the high-latitude northern hemisphere, where there have been relatively frequent eruptions in the Kuriles, Kamchatka, Alaska and Iceland, particularly since 2008 (Figs. 1, 6, 8).

The more sensitive SO₂ measurements by OMI since 2004 (Fig. 4) and IASI since 2007 (Fig. 7) provide more detailed insight into volcanic SO₂ emissions in the last decade. As previously discussed, use of the maximum daily OSI to detect volcanic SO₂ emissions in a continuous daily time-series (Fig. 4) avoids complications when SO₂ from multiple sources at different altitudes is present (accurate SO₂ loadings are shown in Fig. 1). Spikes in the OSI correlate with known eruptions as indicated on Fig. 4, with the OSI value roughly proportional to the maximum SO₂ column amount and altitude on a given day, and the larger (or longer-lived) eruptions have distinctive OSI tails due to the persistence of SO₂ in the atmosphere for days or weeks as it converts to sulfate aerosol. Comparison of the OSI with coincident maximum daily SO₂ and HCl mixing ratios measured by MLS (Fig. 4) clearly indicates the major UTLS injections (e.g., Manam in February 2005, Soufrière Hills in May 2006, Rabaul in October 2006, Kasatochi in August 2008, Redoubt in March 2009, Sarychev Peak in June 2009 (see animation provided as Supplementary Material), Merapi in November 2010, Nabro in June 2011 and Kelut in February 2014; Table 2), although many smaller injections are also apparent upon closer inspection (see also Pumphrey et al., 2015). The significance of the observed HCl emissions will be discussed later (Section 4.1.6). The advantage of SO₂ measurements from multiple platforms is also demonstrated by the large eruption of Manam on November 24, 2004: this occurred during an OMI data gap but the corresponding UTLS SO₂ and HCl injection was detected by MLS (Fig. 4) and the SO₂ cloud was measured by AIRS (Fig. 5), though it was not detected by MIPAS (Höpfner et al., 2015).

Much of the background variation in the OSI time-series in periods devoid of large eruptions is due to smaller eruptions or very high passive SO₂ emissions from volcanoes such as Nyiragongo (DR Congo; 2004–2005; Carn, 2004), Ambrym (Vanuatu; 2005; e.g., Bani et al., 2009, 2012), and Kilauea (Hawaii; strong summit degassing from March 2008), or extended periods of unrest at volcanoes such as Anatahan (CNMI; April–September 2005; see animation provided as Supplementary Material) and Kliuchevskoi (Kamchatka; May–July 2007). Section 4.2 discusses observed sources of passive SO₂ degassing in more depth. It is also likely that some seasonally variable anthropogenic SO₂ emissions are detected by the OSI: for example, the strong SO₂ emissions from the Norilsk metal smelter (Siberia, Russia; e.g., Walter et al., 2012) produce a strong signal in the OMI data in the northern hemisphere spring (March–April) each year (Fig. 4), due to the presence of SO₂ above highly-reflective snow-covered ground.

The AIRS and IASI measurements of UTLS SO₂ emissions and altitude (Figs. 5, 6, 7) are broadly consistent with the OMI SO₂ data, and also with MIPAS SO₂ observations (Höpfner et al., 2015) with the 2008 Kasatochi, 2009 Sarychev Peak and 2011 Nabro eruptions dominating the time-series in terms of SO₂ loading (Table 2). IASI SO₂ altitude retrievals for these eruptions indicate injection of SO₂ to a range of altitudes in the UTLS (Fig. 7), which is consistent with MLS data (Fig. 4). These vertical profiles contrast with the more localized UTLS SO₂ injections associated with the tropical eruptions of Soufrière Hills in May 2006, Rabaul in October 2006, Merapi in November 2010 and Kelut in February 2014 (Figs. 4, 7); this may be a consequence of variable eruption magnitude and style, or of diverse atmospheric dynamics in the tropics and mid-latitudes. An advantage of IASI and AIRS is that they provide robust measurements of high-latitude winter eruptions, such as the December 2010 eruption of Kizimen (Kamchatka; which was incorrectly ascribed to Shiveluch by Höpfner et al. (2015)), the November 2012 eruption of Tolbachik (e.g., Telling et al., 2015), and the 2014 eruption of Holuhraun (Bardarbunga, Iceland; Figs. 5, 7). Long-range dispersal of SO₂ emitted by the June 2011 eruption of Cordon Caulle (Chile), which occurred in the southern hemisphere winter, was also tracked more effectively by the IR sensors (Figs. 6, 8). Average SO₂ columns



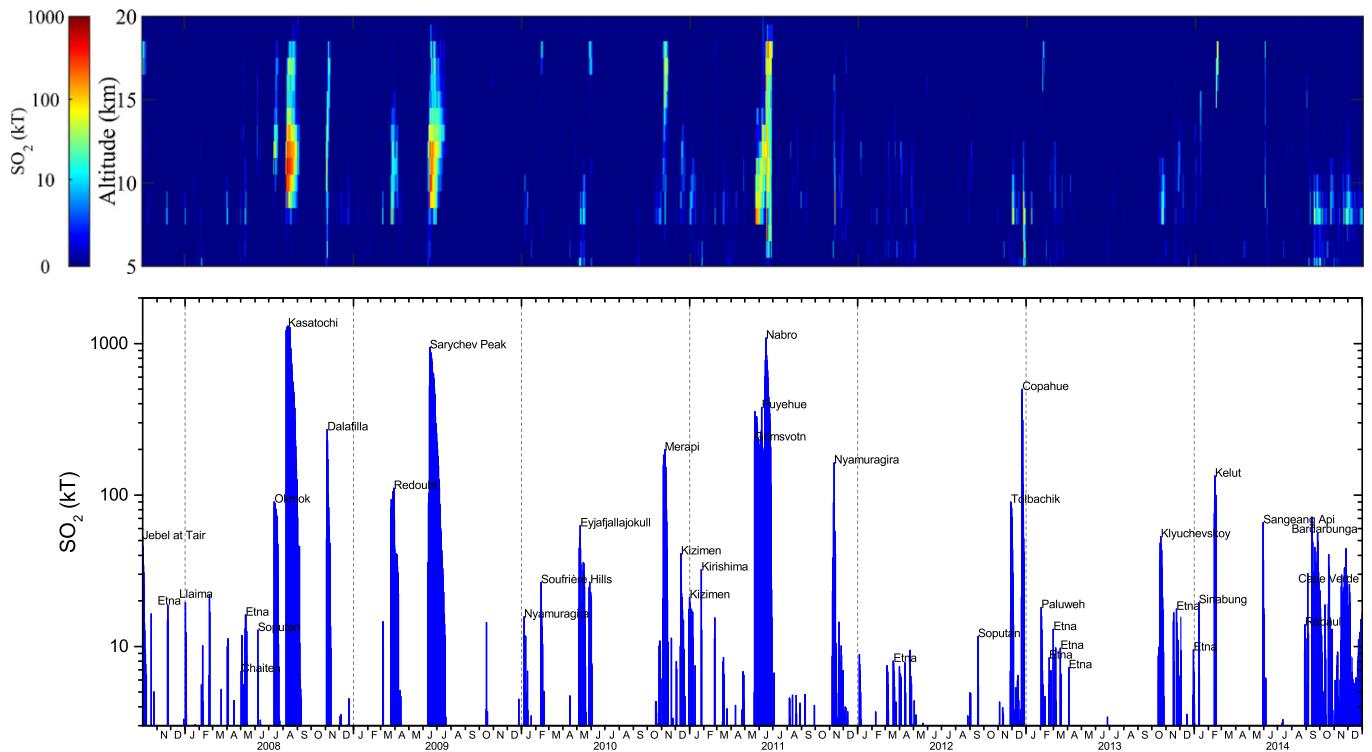


Fig. 7. Time-series of IASI total SO₂ mass (lower panel; logarithmic scale in kilotons) and altitude-binned SO₂ mass (upper panel; for altitudes > 5 km only) measurements from October 2007 to December 2014, derived using the retrieval techniques described by Clarisse et al. (2012, 2014). Prominent volcanic eruptions are labeled in the lower panel.

detected by IASI clearly show the imbalance between northern and southern hemisphere SO₂ emissions since 2007, and long-range tracking of SO₂ from several eruptions made possible by sensitivity to low SO₂ column amounts (Fig. 8). During a two-month period in May–June 2011, large eruptions occurred in Iceland (Grimsvötn), Chile (Cordon Caulle) and Eritrea (Nabro), resulting in UTLS SO₂ emissions spread over an unusually large latitudinal range (Fig. 6).

Comparisons between satellite SO₂ measurements in the overlap periods between missions provide confidence in the data for larger eruptions and in the multi-decadal continuity of the volcanic SO₂ database (e.g., Table 2). TOMS, MODIS, AIRS and MIPAS measured very similar SO₂ amounts (~0.1–0.12 Tg) in the July 2003 Soufrière Hills eruption cloud, which reached 16 km altitude (Table 2; Prata and Bernardo, 2007; Carn and Prata, 2010; Höpfner et al., 2015). The UTLS SO₂ injection from Anatahan on April 6, 2005 was observed by TOMS, OMI, AIRS and MIPAS, with each sensor measuring ~70–80 kt SO₂ (Table 2; Höpfner et al., 2015), with similar agreement observed for most other UTLS SO₂ emissions. Other eruptions, such as the October 2005 effusive eruption of Sierra Negra (Galapagos Islands) show discrepancies between satellite measurements (Table 2) but often there are good reasons for this; in this case a significant fraction of the emitted SO₂ was lower tropospheric, preventing detection by TOMS, which also had some data gaps. Variable spatial coverage of volcanic clouds can also produce differences (e.g., Prata and Bernardo, 2007), hence in general the largest SO₂ mass measured by any sensor is assumed to provide the best estimate.

The August 2008 eruption of Kasatochi, one of the largest explosive eruptions since Pinatubo, is a good example of an event that has been subject to detailed analysis with encouraging convergence of total SO₂ loading estimates from multiple satellite instruments and analysis

techniques (Table 2). Contemporary eruption reports suggested ash emissions to at least 11–14 km altitude during the Kasatochi eruption on August 7, 2008 (Global Volcanism Program, 2008). Multiple satellite instruments observed the emitted SO₂ cloud. Kristiansen et al. (2010) used SO₂ measurements from OMI, GOME-2 and AIRS in an inversion scheme to derive a total SO₂ discharge of 1.7 Tg, and a co-eruptive vertical SO₂ profile extending from 7 to 20 km altitude with a major peak at 12 km. They also estimated that ~1 Tg SO₂ (i.e., ~60% of the total SO₂ discharge) was injected into the local stratosphere (>10 km altitude), although this is dependent on the tropopause definition (Kristiansen et al., 2010; Clarisse et al., 2014; similarly, Eckhardt et al. (2008) estimated ~10–60% of SO₂ emitted by the October 2007 eruption of Jebel al-Tair (Yemen) was injected into the stratosphere, depending on tropopause definition). Kristiansen et al. (2010)'s SO₂ loading estimate for Kasatochi could be regarded as robust since the inversion technique includes multiple UV and IR satellite datasets. Individual satellite datasets yield similar results: Karagulian et al. (2010) report 1.7 Tg SO₂ based on IASI measurements using both the 7.3 μm and 4 μm SO₂ bands. A subsequent, improved analysis of IASI data by Clarisse et al. (2012) found a maximum of 1.6 Tg SO₂ measured on Aug. 11, 2008; in agreement with the 1.6 Tg SO₂ measured by GOME-2 (Nowlan et al., 2011). Prata et al. (2010) retrieved a total of ~1.2 Tg SO₂ using AIRS data, with the lower SO₂ amount detected perhaps due to lower sensitivity to low SO₂ columns and incomplete cloud coverage by AIRS. Krotkov et al. (2010) report a slightly higher value of ~2.2 Tg SO₂, injected to 10–12 km altitude, based on OMI measurements and trajectory modeling; in this case the estimated SO₂ loading was derived from extrapolation of the SO₂ decay trend back to the eruption time. This procedure is subject to some uncertainty as the prevailing SO₂ removal processes may not be the same in the early and aged volcanic plume, plus

Fig. 6. Time-series (upper panel) and cumulative map (lower panel) of SO₂ emissions detected by AIRS in 2011 (data for other years in 2003–2014 are provided as Supplementary Material). Upper panel shows daily SO₂ amounts (Tg) retrieved by AIRS using the Prata and Bernardo (2007) algorithm (gray bars) and reported volcanic eruptions (colored bars and stars; color denotes VEI). Horizontal bars indicate eruption duration reported by the Smithsonian GVP; horizontal arrows indicate eruptions or activity continuing into the following year. Note that not all the indicated eruptions produced SO₂ detected by AIRS. Lower panel shows cumulative SO₂ column amounts retrieved by AIRS with major eruptions indicated.

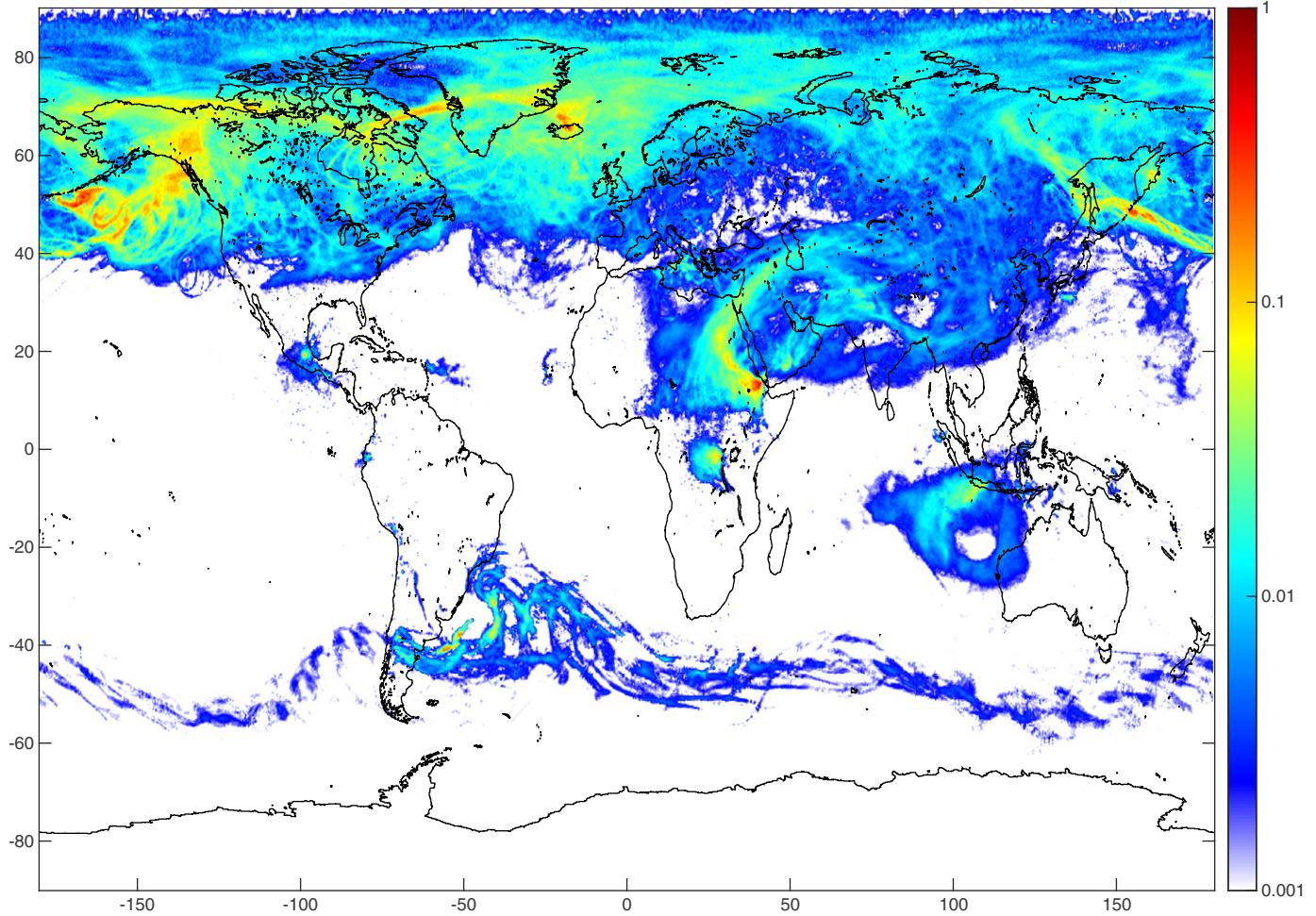


Fig. 8. Average SO₂ columns measured by IASI over ~7 years (October 2007–December 2014). The scale is logarithmic ($\log[1 + (\text{mean SO}_2 \text{ column in DU})]$). The more prominent eruptions evident in this map include Kasatochi (Alaska; August 2008), Sarychev Peak (Kurile Islands; June 2009), Merapi (Indonesia; November 2010), Grimsvötn (Iceland; May 2011), Cordón Caulle (Chile; June 2011), Nabro (Eritrea; June 2011) and Copahue (Chile; December 2012).

the observed e-folding time for SO₂ removal was relatively short (~9 days; Krotkov et al., 2010), possibly due to proximity to the tropopause (e.g., Campbell et al., 2012), and this impacts the extrapolated SO₂ mass estimate. Note that the largest SO₂ mass measured by OMI in a single, coherent cloud was 1.8 Tg on August 10 (Krotkov et al., 2010). MODIS SO₂ retrievals for Kasatochi, including 8.6 μm data corrected for ash interference (Corradini et al., 2010), are broadly consistent with the other estimates within stated uncertainties (Table 2). Thus, when all UV and IR satellite measurements are considered we find general convergence on a total SO₂ emission of ~2 Tg for the 2008 Kasatochi eruption, a value that can be considered very robust.

4.1.2. Eruptive SO₂ flux

Knowledge of the time-averaged flux of volcanic SO₂ into the UTLS is crucial for interpretation of trends in SAOD and assessment of the impact of volcanic activity on climate. Using TOMS SO₂ data for 1978–1993, Bluth et al. (1993) estimated an annual volcanic SO₂ flux from explosive volcanism of ~4 Tg, clearly influenced by the large El Chichón and Pinatubo eruptions in that era. Pyle et al. (1996) calculated a medium-term (~100 year) flux of ~1 Tg/yr. volcanic SO₂ to the stratosphere (0.3–3 Tg/yr. range), and a short-term (10–20 year) flux due to small eruptions (10^{10} – 10^{12} kg or VEI 3–5) of ~0.4 Tg/yr. Here, we provide updated estimates of the volcanic SO₂ flux to the UTLS based on recent satellite measurements, with improved data for smaller eruptions.

Table 3 reports the total annual SO₂ emissions measured by satellites from 1978 to 2014 for all detected eruptions and for explosive eruptions

only (i.e., excluding effusive eruptions; also plotted in Fig. 1). The latter have greater potential to reach the UTLS and impact climate. Our distinction between explosive and effusive eruptions in the SO₂ emissions database (Carn, 2015c) is based on the dominant style of activity; we define effusive eruptions as those dominated by effusion of basaltic lava flows (typically in intraplate settings). Eruptions involving extrusion of silicic lava domes (e.g., Soufrière Hills, Montserrat) are categorized as ‘explosive’ since it is the discrete, explosive emissions from these eruptions that are most effectively measured from space. To estimate total SO₂ emissions from effusive eruptions we have used the procedure described by Bluth and Carn (2008), which accounts for rapid, daily removal of SO₂ in lower tropospheric plumes.

A notable feature of the volcanic SO₂ data is significant inter-annual variability, with total explosive SO₂ emissions varying from 10 kt (in 1987) to ~24 Tg (in 1991; Table 3). On average, ~50% of annual measured SO₂ emissions have been sourced from explosive eruptions (Table 3), indicating the important contribution of sulfur-rich effusive basaltic eruptions to the global flux. The mean annual SO₂ fluxes from all detected eruptions and explosive eruptions are 2.8 Tg (range = 0.04–26 Tg) and 1.6 Tg (range = 0.01–24 Tg), respectively; the latter can be considered an estimate of the SO₂ flux to the UTLS, and if we assume that ~60% of this SO₂ enters the stratosphere (based on detailed analyses of the 2008 Kasatochi and 2007 Jebel al-Tair eruptions; Eckhardt et al., 2008; Kristiansen et al., 2010) then the stratospheric flux is ~1 Tg/yr. SO₂, similar to earlier estimates of medium-term and longer-term (~ 10^3 years) fluxes (Pyle et al., 1996). We can

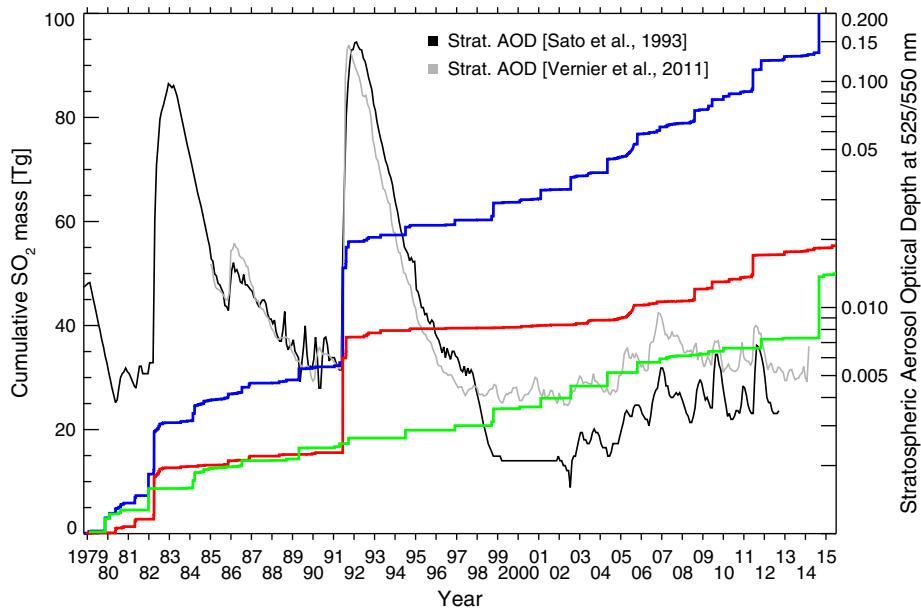


Fig. 9. Cumulative SO₂ emissions from volcanic eruptions against time for 1978–2015, showing emissions for all eruptions detected from space (blue line), explosive eruptions only (red line) and effusive eruptions only (green line). Black line shows global stratospheric aerosol optical depth (SAOD) at 550 nm for 1978–2012 from Sato et al. (1993) and recent updates through December 2012 (<http://data.giss.nasa.gov/modelforce/strataer/>); gray line shows the tropical (30°S–30°N) SAOD at 525 nm data of Vernier et al. (2011; with recent updates) for 1985–2014 based on SAGE II, GOMOS and CALIPSO observations of aerosol between 15 and 40 km altitude. Differences in the SAOD datasets arise from different sources of SAOD observations, interpolation methods, latitude range (the Sato et al. (1993) data plotted are global whereas the Vernier et al. (2011) data are for the tropics; thus high-latitude eruptions produce a weaker signal in the latter) and altitude ranges (the Vernier et al. (2011) data cover altitudes of 15–40 km and hence includes part of the tropical upper troposphere, resulting in larger AODs in some periods).

conclude that the flux of volcanic SO₂ to the stratosphere in the past 3–4 decades has not been abnormally high or low. The relationship between SO₂ emission and eruption magnitude is explored in more detail in Section 4.1.3.

The exact proportion of emitted SO₂ that enters the stratosphere during an explosive eruption will vary with eruption magnitude, location and style. Although we attempt to separate volcanic SO₂ injected into the UTLS from that injected into the stratosphere sensu stricto, volcanic SO₂ injected into the upper troposphere below the tropopause also has the potential to impact the stratosphere. Effusive eruptions are not generally regarded as significant sources of stratospheric pollution, notwithstanding large-volume fissure eruptions with high eruption rates (e.g., Stothers et al., 1986), yet they frequently emit prodigious quantities of SO₂ into the upper troposphere (e.g., Krueger et al., 1996; Carn and Bluth, 2003; Head, 2005; Bluth and Carn, 2008; Hughes et al., 2012; Höpfner et al., 2015) where it has the potential to cross the tropopause. MIPAS detected SO₂ in the UTLS (at altitudes of up to 18–22 km) after several eruptions of Nyamuragira (in 2002, 2006 and 2010) and the 2009 Fernandina eruption (Galápagos Islands; Höpfner et al., 2015), suggesting that such eruptions should not be neglected as a possible source of stratospheric SO₂. The June 12, 2011 eruption of Nabro, which was initially (and, it transpired, incorrectly) reported as tropospheric but produced a significant SAOD anomaly (e.g., Sawamura et al., 2012; Uchino et al., 2012; Bourassa et al., 2012; Mateshvili et al., 2013), ignited a debate over the relative roles of direct injection and cross-tropopause transport by other processes such as deep convection or isentropic ascent in lofting volcanic SO₂ into the stratosphere (Bourassa et al., 2012; Fromm et al., 2013; Vernier et al., 2013; Bourassa et al., 2013; Fairlie et al., 2014; Fromm et al., 2014; Penning de Vries et al., 2014; Clarisse et al., 2014). In addition to the evidence presented in these works, which indicate that both early direct injection and subsequent slow ascent of SO₂ and sulfate aerosol contributed to Nabro's stratospheric impact, daily MLS SO₂ retrievals (Fig. 10) also show SO₂ close to or above the tropopause (at 68 hPa) as early as June 13–14 plus a slower ascent of SO₂ (up to ~46 hPa or 19–20 km altitude) in the subsequent two weeks. Interestingly, MLS data do not

show the same evidence for substantial stratospheric HCl input (Fig. 10), likely due to more efficient HCl removal processes in the troposphere. Overall, the Nabro eruption demonstrates that volcanic eruptions can be complex, multifaceted events and require analysis on a case-by-case basis to unravel their stratospheric impact. The Nabro eruption displayed characteristics of both explosive and effusive eruptions in that it initially produced an energetic eruption column (e.g., Fee et al., 2013) that penetrated the UTLS, yet it did not produce a substantial ash cloud and emitted a long lava flow, with tropospheric SO₂ emissions continuing for several weeks after the initial paroxysm (e.g., Theys et al., 2013; Goitom et al., 2015). It can thus be considered a relatively rare, hybrid event sharing features of two 'end-member' eruptions: a sulfur-rich, effusive eruption injecting SO₂ predominantly into the troposphere (e.g., similar to Nyamuragira) and an explosive eruption with direct stratospheric injection (e.g., the January 2005 Manam eruption; Fig. 4).

4.1.3. VEI-SO₂ loading relationships

Analysis of the relationship between SO₂ production and eruption magnitude (VEI) can inform studies of the atmospheric and climate impact of eruptions prior to the satellite measurement era. In comparing measured SO₂ emissions with reported eruption VEIs, we extend earlier work based on TOMS SO₂ data by Schnetzler et al. (1997) with the addition of many more observations of smaller eruptions due to improved satellite data quality. We also attempt to estimate how much volcanic SO₂ could elude detection from space based on VEI-SO₂ mass relationships. We note that more precise (i.e., continuous) measures of eruption size, such as magnitude and intensity, are more useful for such analyses than the more limited cardinal (or discrete) VEI scale, since each VEI category encompasses a significant range of eruption size (e.g., Pyle, 1995, 2015). However, magnitudes and intensities are only available for a relatively small number of eruptions, and here we use the Smithsonian GVP database of VEIs from VOTW 4.3.0. Some small modifications to the VOTW database were necessary to account for long-duration eruptions assigned a single VEI; hence continuous activity at Anatahan in 2005, included as a single VEI 3 eruption in the VOTW catalog, was

Table 3Total annual eruptive volcanic SO₂ emissions, 1978–2014.

Year	Total SO ₂ emissions (kt)	Explosive SO ₂ emissions (kt)	Explosive/Total SO ₂ (%)
1978 ^a	87	87	100
1979	2945	63	2.1
1980	2882	1228	43
1981	5562	1430	26
1982	11,033	9950	90
1983	348	260	75
1984	3957	252	6.4
1985	1241	852	69
1986	2086	872	42
1987	38	10	26
1988	580	300	52
1989	2329	177	7.6
1990	186	186	100
1991	26,082	24,214	93
1992	810	810	100
1993	450	450	100
1994	1874	360	19
1995 ^a	TOMS data gap		
1996 ^a	987	100	10
1997	41	41	100
1998	3265	38	1.2
1999	130	85	65
2000	653	336	51
2001	1783	122	6.8
2002	2626	271	10
2003	679	679	100
2004	2997	410	14
2005	4634	2501	54
2006	1347	661	49
2007	712	122	17
2008	2625	2318	88
2009	1934	1379	71
2010	1470	867	59
2011	6030	4310	71
2012	763	563	74
2013	185	180	97
2014	5296	608	11
Total	100,646	57,090	
Mean^b	2873	1629	57
1σ^b	4621	4317	

^a Denotes incomplete year or data gap.^b Excluding 1978 and 1995.

subdivided into discrete VEI 2 and 3 events based on reported plume altitudes.

As pointed out by Pyle (2015), there are few metrics other than VEI, magnitude and intensity that are routinely used to describe eruption size. However, we contend that SO₂ measurements are the most accurate way of quantifying the impact of an eruption on climate (and atmospheric composition generally), which is ultimately among the main goals of any eruption magnitude scale, given that volcanic ash has only minor climate impacts (e.g., Robock, 2000). Since SO₂ emissions are not clearly linked to other magnitude scales (which are more relevant to impacts determined by emissions of solid ejecta such as volcanic ash and lava, e.g., on aviation or the environment), except in the broadest sense (e.g., Schnetzler et al., 1997; Blake, 2003; Sciallet et al., 2003), there may be a need for multiple measures of eruption magnitude and impact. Intensity (which determines eruption column altitude) may be a more relevant metric than magnitude or VEI for assessment of climate impacts.

Tables 4 and 5 summarize the measured SO₂ emissions per VEI category for explosive eruptions and all detected eruptions, respectively, and the data for individual eruptions are plotted in Fig. 11. Fig. 11 indicates that, when effusive eruptions (as defined in Section 4.1.2) are excluded, there is a broad correlation between VEI and SO₂ emission; i.e., there is roughly an order of magnitude increase in mean SO₂ discharge with increasing VEI (Table 4). A similar correlation was found by Schnetzler et al. (1997) using a smaller

dataset. Based on the mean SO₂ loading for each VEI (Table 4) we find the following relationship:

$$\log_{10}(SO_2, \text{Tg}) = 0.71\text{VEI} - 3.15 \quad (1)$$

Cross-checking with the VOTW database shows that SO₂ emissions by all reported eruptions with VEI ≥ 4 have been quantified (consisting of 26, 4, and 1 eruptions of VEI 4, 5, and 6, respectively, reported in the VOTW database), confirming that satellite measurements provide excellent constraints on SO₂ emissions by the largest eruptions. Note that some eruptions are assigned high VEIs due to large cumulative erupted volumes over long periods (years), but may not feature any discrete large eruptive events; e.g., activity at Kliuchevskoi (Kamchatka) in 1986–1990 (where SO₂ was occasionally detected by TOMS), or long-term dome-building eruptions at Shiveluch (Kamchatka) in 1999–2014, during which time a few relatively small SO₂ emissions have been detected. In such cases we include these eruptions as ‘detected’ if any SO₂ emissions were measured at any time during the reported eruption period, although the total eruptive SO₂ emissions may be poorly constrained. Emissions from such continuous eruptions are more akin to passive volcanic degassing, with similar challenges for detection from space.

For eruption magnitudes below VEI 4, the SO₂ emission record becomes increasingly incomplete. Based on reported VEIs, around 50% of VEI 3 eruptions ($n = 162$) have been detected by satellite SO₂ measurements (Tables 4, 5). The VEI 3 category likely includes some phreatic or phreatomagmatic eruptions that may generate substantial eruption columns (>3 km altitude required for VEI 3) but which would not emit large quantities of SO₂. Some VEI 3 events also correspond to extended periods of volcanic unrest, often with continuous SO₂ emissions (e.g., Ambrym, Vanuatu). Other VEI 3 eruptions only appear to produce ash, with no detectable SO₂; e.g., many eruptions of Bezymianny (Kamchatka) produce little measurable SO₂, but based on Smithsonian GVP reports it appears that the larger explosive eruptions are detected. However, Bezymianny clearly does not appear to be a major source of SO₂ emissions based on detected events. We also stress that, for the lower magnitude eruptions, not all satellite data has been analyzed in detail due to the increasingly large number of events. However, it is likely that any significant SO₂ emissions would have been noticed and recorded.

At magnitudes of VEI ≤ 2, it is clear that the majority of eruptions (87% or more) are not detected (Tables 4, 5), most likely due to insufficient SO₂ release. However, it is also certain that not all the >1000 reported VEI 0–2 eruptions have been checked for SO₂ emissions in satellite data. By definition, non-explosive, effusive eruptions (e.g., in Hawaii, DR Congo, the Galápagos Islands and Iceland) are generally assigned a VEI of 0–2, but these are all detected due to their typically SO₂-rich emissions (e.g., Carn and Bluth, 2003; Sharma et al., 2004; Head, 2005; Bluth and Carn, 2008; Carn, in press), which obfuscate any relationship between SO₂ emission and VEI (e.g., Fig. 11). In fact, the only VEI 0 eruptions with confirmed SO₂ emissions are effusive eruptions (Tables 4, 5; Fig. 11). Although the undetected, explosive VEI 0–2 eruptions are unlikely to be significant from a climate perspective (due to low column heights and/or low SO₂ amounts), quantifying their emissions may be important for the global sulfur budget and sulfur cycle, and for the budgets of other, longer-lived volcanic gases such as CO₂, but would require more extensive ground-based measurements of this class of eruption. Indeed, at the lower end of the VEI scale the distinction between continuous, passive degassing and frequent small eruptions becomes tenuous.

Despite the poor sampling of low magnitude eruptions, a notable feature of the SO₂ data is the high variability of SO₂ emissions from eruptions of intermediate size (VEI 2–4), with standard deviations similar to or exceeding the mean SO₂ loading for each VEI (Tables 4, 5; Fig. 11). The range of SO₂ emissions for intermediate VEIs is larger than reported by Schnetzler et al. (1997) due to improved data for

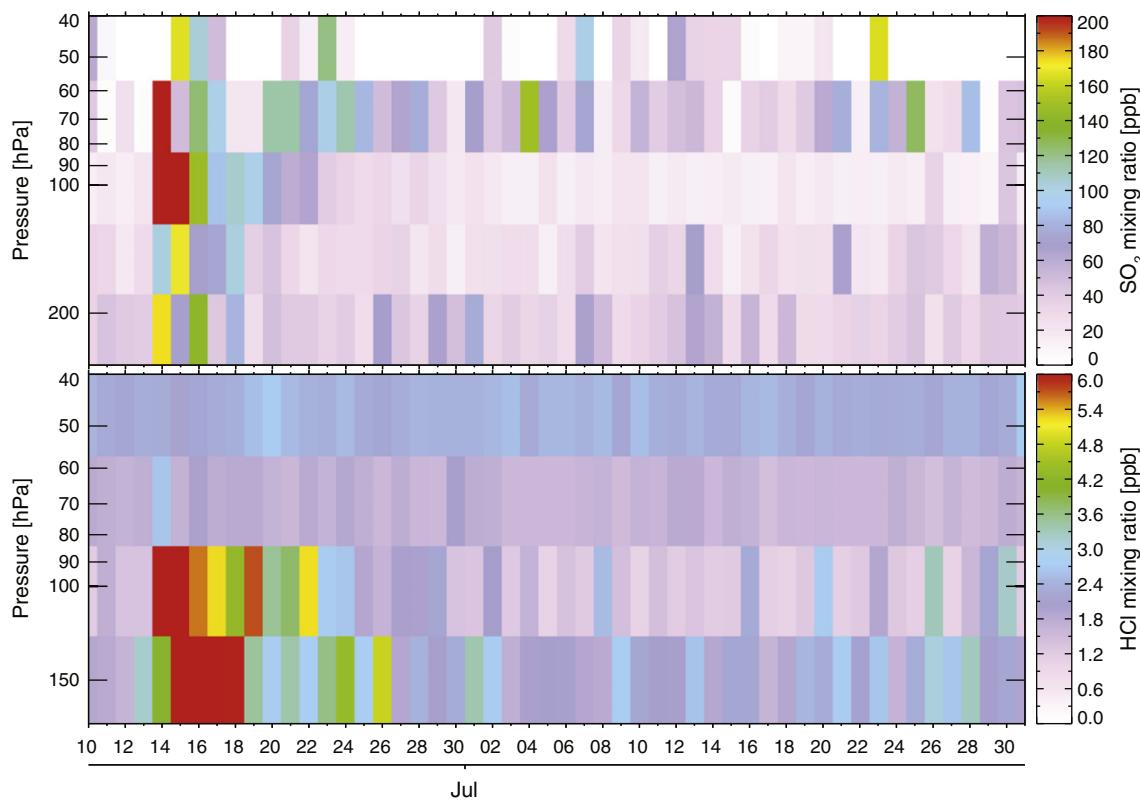


Fig. 10. Daily maximum UTLS mixing ratios of SO₂ (upper panel) and HCl (lower panel) measured by MLS between June 10 and July 30, 2011, showing the impact of the June 12, 2011 eruption of Nabro (Eritrea).

smaller eruptions; thus the increased variability may reflect sampling of eruptions with a larger range of magmatic sulfur contents. Since all VEI 4 eruptions since 1978 have been detected, we have good constraints on the variability of SO₂ discharge from this magnitude class, which covers ~2–3 orders of magnitude. The greatest variability is found for VEI 3 eruptions (Tables 4, 5), which range over ~3 orders of magnitude (Fig. 10). The actual variability may be even greater, since only 50% of VEI 3 eruptions have been detected (likely the more SO₂-rich ones). This extreme variability makes it challenging to estimate SO₂ emissions from past eruptions based on VEI alone, but this analysis at least provides some potential error bars on such predictions. One likely cause of the observed variability is that some VEIs may be inaccurate, which relates to our earlier point that SO₂ emissions offer a useful alternative indication of eruption magnitude. For example, both the 2011 Nabro and 2008 Kasatochi eruptions were assigned a VEI of 4, but their total SO₂ emissions (~2–4 Tg) are the highest in this VEI class and are similar

to or greater than the mean SO₂ loading for VEI 5 eruptions. Indeed, for Nabro, Goitom et al. (2015) estimate a magnitude of 5.1 based on total observed SO₂ emissions (Theye et al., 2013) and magmatic sulfur content. Similarly, the magnitude of the Kasatochi eruption has been estimated as 4.5–4.7 (Pyle, 2015), although we note that the 2008 Chaitén eruption (which produced <20 kt of detectable SO₂) was also a magnitude 4.7–5 event. Nevertheless, refinement of eruption magnitudes would likely improve the correlation between SO₂ emission and VEI shown in Fig. 11, and the SO₂ measurements provide perhaps the most robust basis for adjusting VEIs for recent eruptions. Finally, we note that Scaillet et al. (2003) also found an increased spread of estimated SO₂ yield with decreasing eruption magnitude based on petrological data, which they attributed to the variability in bulk sulfur contents of magmas. Thus the variability in observed SO₂ loadings for smaller eruptions (Fig. 10) may represent sampling of magmas with different bulk sulfur content.

Table 4
Explosive volcanic SO₂ emissions per Volcanic Explosivity Index (VEI) class.

VEI	Eruptions ^a	Detected (%)	Measured SO ₂				Est. undetected SO ₂	
			Mass (Tg)	Flux (Tg/yr) ^b	Mean (Tg)	1σ (Tg)	Mass (Tg)	Flux (Tg/yr) ^b
0	222	0	–	–	–	–	0.22	0.006
1	507	4	0.26	0.007	0.006	0.006	2.9	0.081
2	479	13	3.22	0.09	0.02	0.02	8.35	0.23
3	162	50	8.18	0.23	0.05	0.11	5.67	0.16
4	26	100	13.05	0.36	0.45	0.75	0	0
5	5	100	11.28	0.31	2.26	2.82	0	0
6	1	100	20	0.56	20	–	0	0
Total	1402		55.97			17	0.48	

^a Extracted from the Global Volcanism Program Volcanoes of the World 4.3.0 database (accessed 19 Sept. 2014) for Oct. 1978–Sep. 2014. Listed eruptions without an assigned VEI (75 eruptions) were ignored.

^b Mean annual SO₂ flux for the ~36 year period of observations.

Table 5
Total volcanic SO₂ emissions per Volcanic Explosivity Index (VEI) class.

VEI	Eruptions ^a	Detected (%)	Measured SO ₂			
			Mass (Tg)	Flux (Tg/yr) ^b	Mean (Tg)	1σ (Tg)
0	222	5	6.6	0.18	0.6	1.28
1	507	4	4.6	0.13	0.05	0.15
2	479	13	21.4	0.59	0.07	0.3
3	162	50	22.4	0.62	0.14	0.45
4	26	100	13.2	0.37	0.44	0.74
5	5	100	11.3	0.31	2.26	2.82
6	1	100	20	0.56	20	–
Total	1402		99.6			

^a Extracted from the Global Volcanism Program Volcanoes of the World 4.3.0 database (accessed 19 Sept. 2014) for Oct. 1978–Sep. 2014. Listed eruptions without an assigned VEI (75 eruptions) were ignored.

^b Mean annual SO₂ flux for the ~36 year period of observations.

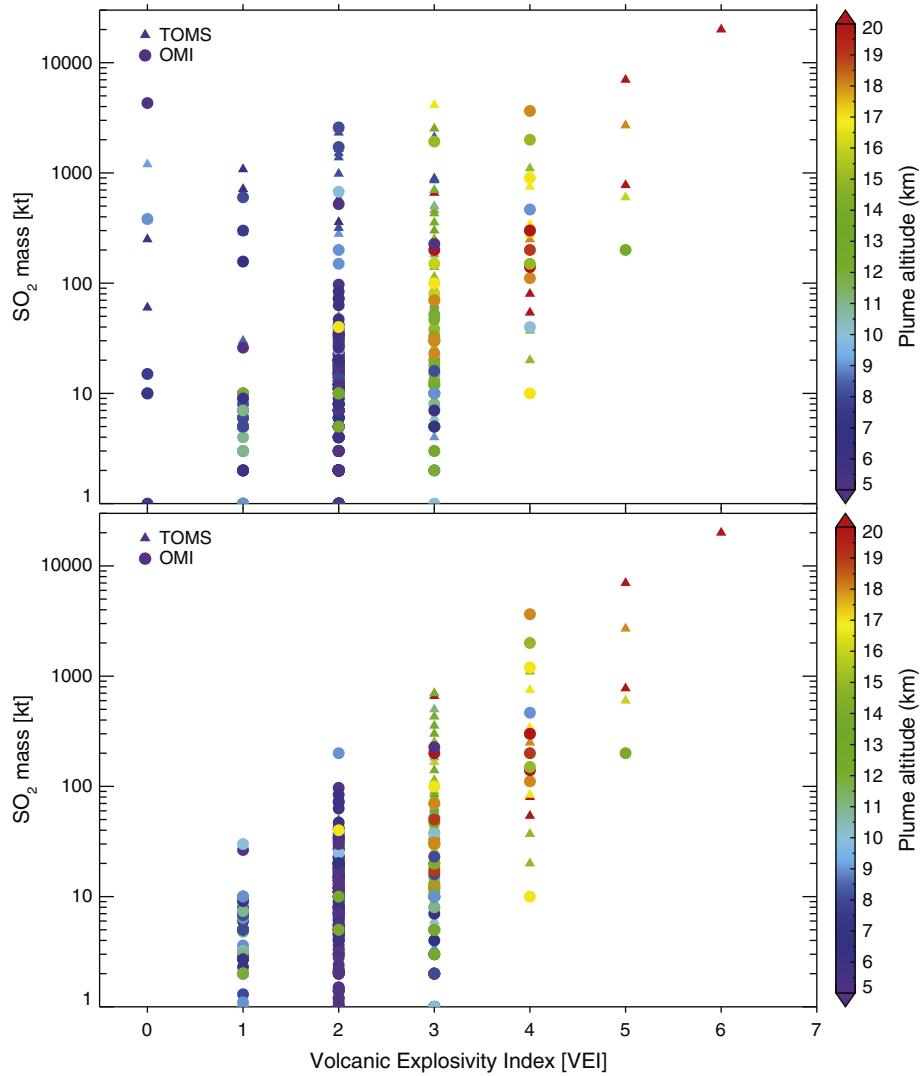


Fig. 11. Upper panel: SO_2 mass loading against Volcanic Explosivity Index (VEI) for all measured eruptions (1978–2014). Triangles and circles denote eruptions detected by TOMS (i.e., in 1978–2004) and OMI (i.e., in 2004–2014), respectively. Lower panel: SO_2 mass loading against VEI for explosive eruptions only (i.e., excluding effusive eruptions regardless of VEI). VEIs are derived from the Smithsonian Global Volcanism Program (GVP) database (VOTW 4.3.0).

To assess the SO_2 emissions from low-magnitude explosive eruptions that elude detection from space, we can use the mean SO_2 loading for detected eruptions, the VEI- SO_2 regression (Eq. (1)), or an estimate of the sensor detection limit. For example, the VOTW 4.3.0 catalog includes 507 eruptions with VEI 1, of which only 4% have detected SO_2 emissions (Table 4), i.e., 487 eruptions are unaccounted for. Using the mean SO_2 emission for detected VEI 1 eruptions (excluding effusive eruptions) of 0.006 Tg SO_2 yields an estimated SO_2 emission of $487 \times 0.006 = 2.9$ Tg SO_2 from undetected eruptions (Table 4). Using Eq. (1) yields an estimated SO_2 loading of 0.004 Tg per eruption and a total undetected amount of 1.9 Tg SO_2 . We consider these upper limits, since it is likely that the larger SO_2 emissions in each VEI category would be detected, and hence the mean SO_2 mass may be biased high, and some eruptions are known to have produced only ash emissions and/or no measurable SO_2 (e.g., phreatic or phreatomagmatic eruptions, submarine eruptions). Using the same logic for all VEI categories with missing data (VEI 0–3) we calculate the estimates of undetected SO_2 emissions and equivalent annual mean fluxes for the ~36 years of satellite measurements listed in Table 4. Hence, the total estimated unmeasured SO_2 discharge for VEI 0–3 eruptions is ~0.48 Tg/yr. SO_2 , or ~17 Tg SO_2 in total (Table 4), i.e., equivalent to a single Pinatubo-scale event, or ~20% of the ~100 Tg total measured SO_2 emissions. Most of the undetected SO_2 is sourced from VEI 2 eruptions (~50%; Table 4).

Again, we consider these estimates to be upper bounds for the reasons mentioned above. The undetected SO_2 emissions are commensurate with one VEI 4 eruption per year, or a single VEI 6 eruption, but although this appears significant we stress that the unquantified SO_2 is sourced from low-magnitude eruptions and hence is unlikely to significantly impact the stratosphere or climate. However, the emissions will impact the tropospheric SO_2 budget and the budgets of other volcanic gases (e.g., CO_2). Due to variable eruption rates, the unmeasured fraction may be more significant in some years than others, but we find an overall uncertainty of ~20% on total eruptive SO_2 emissions to be very acceptable. A further consideration in calculating the mean SO_2 emission for each VEI category is whether the probability density function (PDF) of SO_2 loading for measured eruptions is Gaussian or lognormal, since this affects calculation of the mean. Although data are limited, the PDF appears approximately Gaussian for some VEIs (VEI 4), but may be more lognormal for lower VEIs. Further statistical analysis of the database is encouraged.

In summary, we conclude that the flux to the stratosphere (or UTLS) from VEI 4+ eruptions is very well constrained (Tables 4, 5), but significant uncertainty remains on the flux from smaller eruptions (as for passive volcanic degassing). However, more eruptions of low VEI eluded detection during the TOMS missions (1978–2004) than since 2004 with the onset of more sensitive OMI and IASI SO_2 measurements, so

the uncertainty on SO₂ emissions from small eruptions is lower for the last decade.

4.1.4. Sources of eruptive volcanic SO₂ emissions

In total, around 110 volcanoes are responsible for the eruptive SO₂ emissions detected from space in the current database (Fig. 12). With the exception of Antarctica and the South Sandwich Islands, at least one eruption has been detected in each of the 19 volcanic regions defined by the Smithsonian GVP (Siebert et al., 2010; Fig. 12). A further ~30 volcanoes have produced passive (non-eruptive) SO₂ emissions detected in satellite data (see later discussion), resulting in a total of ~140 global volcanoes with detectable SO₂ emissions since 1978. This is only ~9% of the 1545 volcanoes with known or inferred Holocene eruptions, or ~17% of the 858 volcanoes with dated Holocene eruptions, listed in the Smithsonian GVP catalog (Siebert et al., 2010). Table 6 lists the sources of detected eruptive SO₂ emissions over 10 kt between 1978 and 2014 in decreasing order of total measured SO₂ production, along with the number of eruptions generating the observed SO₂ emissions, and a qualitative assessment of degassing style. Only eruptive SO₂ emissions are reported in Table 6, i.e., passive degassing from volcanoes with variable degassing styles such as Etna and Kilauea is excluded. The global distribution of eruptive SO₂ sources is shown in Fig. 12.

The sources in Table 6 highlight the significant contribution of effusive volcanic eruptions to the eruptive volcanic SO₂ flux (e.g., Nyamuragira, Sierra Negra, Holuhraun, Fernandina, Cerro Azul, Mauna Loa, Wolf), though the SO₂ emitted by these events is mostly tropospheric and less significant for climate. However, effusive eruptions have the potential to inject SO₂ at or above tropopause altitudes, particularly at higher latitudes (e.g., Stothers et al., 1986; Krueger et al., 1996), and have been linked to SAOD perturbations in the past (e.g., a SAOD disturbance in 1979 was partly attributed to the 1979 Sierra Negra eruption; Fig. 13; Rosen and Hofmann, 1980). Hence, given the frequency of these eruptions (e.g., Figs. 1, 9), they constitute an important source of SO₂ in the UTLS. Otherwise, the dominant eruptive SO₂ emissions (>2 Tg SO₂) have been produced by the first recorded eruptions of previously dormant volcanoes (mostly in subduction zones), typically after centuries or more of quiescence (e.g., 1991 Pinatubo: previous dated eruption in 1450; 1982 El Chichón: previous known eruption in 1360 or 1850; 2011 Nabro: first known eruption; 2008 Kasatochi: previous confirmed minor eruption in 1760; 2003 Anatahan: first

documented eruption; Siebert et al., 2010). However, there are exceptions: the large 1991 eruption of Cerro Hudson followed relatively recent, albeit smaller, eruptions in 1891 and 1971 (Siebert et al., 2010), although in this case the large SO₂ emission in 1991 may have been supplied by a large basaltic dyke intrusion that triggered the eruption (e.g., Kratzmann et al., 2009). One possible explanation for the broad correlation between inter-eruptive quiescence and SO₂ emission is that it reflects the accumulation of a vapor phase in long-lived magma chambers during repose (e.g., Wallace and Gerlach, 1994; Scaillet et al., 2003; Wallace, 2005). Although many of the dominant sources have limited data on inter-eruptive degassing (no significant SO₂ emissions have been detected from space, but weak, fumarolic emissions would not be measured), they appear to release the vast majority of their SO₂ during eruptions. Whether this SO₂ originates from a stored vapor phase or syn-eruptive melt degassing is an open question that requires more extensive petrological studies. However, the behavior of other volatile species such as CO₂ may be different (e.g., there could be substantial inter-eruptive outgassing of CO₂ exsolved at depth), but would require extensive ground-based CO₂ flux measurements to resolve.

After the dominant explosive SO₂ sources, which tend to produce single, large injections of SO₂ into the UTLS, there are a number of volcanoes that produce smaller but more frequent emissions. The latter include Rabaul, Manam, Ulawun, Soputan, Kelut, Soufrière Hills, Sarychev Peak and Hekla (Table 6), which generally produce <1 Tg SO₂ per eruption but erupt relatively frequently (e.g., Tupper and Kinoshita, 2003; Tupper et al., 2004, 2007; Carn and Prata, 2010; Kushendratno et al., 2012) and in some cases may be persistently active between larger eruptions (e.g., Rabaul, Manam). Some volcanoes, such as Manam and Soputan, are prolific SO₂ emitters with 100% eruption detection rates where explosive degassing appears to dominate the total SO₂ flux, whereas passive SO₂ emissions appear more significant at others (e.g., Tungurahua (Ecuador) and Nevado del Ruiz (Colombia)). Soufrière Hills volcano (SHV) is unusual in that the SO₂ emissions listed in Table 6 are all associated with lava dome collapses or explosive events during a single, long-lived dome-forming eruption (Carn and Prata, 2010); these events trigger the release of a deep, SO₂-rich vapor phase (e.g., Christopher et al., 2010) and are not supplied by syn-eruptive degassing. Although these are not explosive eruptions in the conventional sense, they are still capable of injecting SO₂ into the UTLS and impacting SAOD (e.g., the July 2003, May 2006 and February

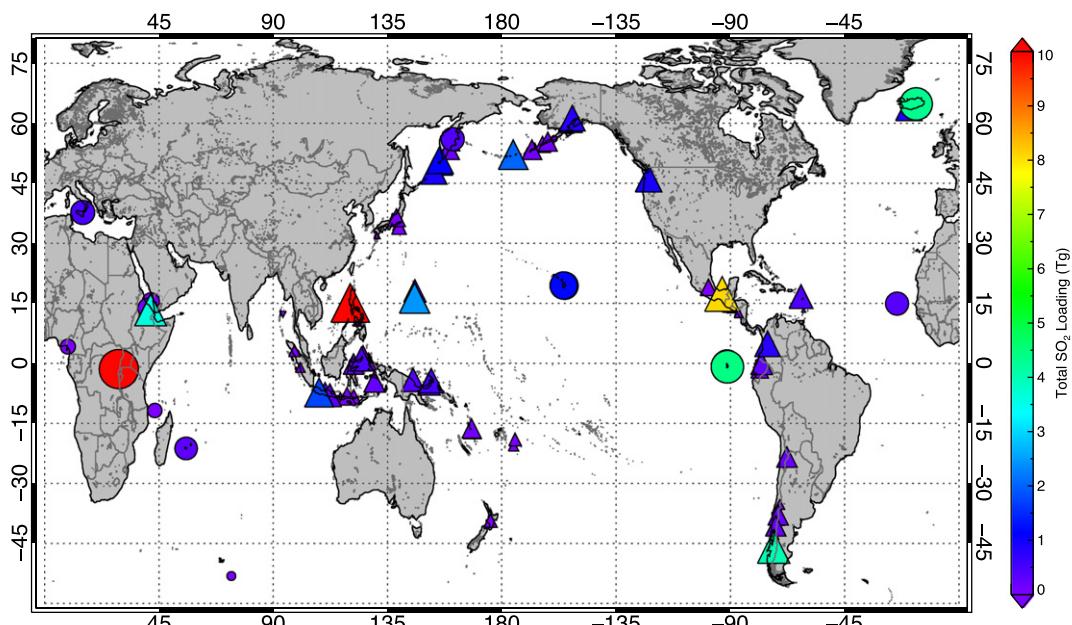


Fig. 12. Global map of sources of eruptive SO₂ emissions detected from space in 1978–2014. Symbol size is proportional to total SO₂ emissions measured for each volcano (Table 6), which is also indicated by the symbol color scale. Triangles indicate explosive eruptions and circles indicate effusive eruptions.

Table 6Sources of eruptive volcanic SO₂ emissions (>10 kt), 1978–2014.

Volcano	Total SO ₂ emission (Tg)	Eruptions	Degassing style*
Nyamuragira	25.149	17	1
Pinatubo	20.194	1	0
El Chichon	8.090	1	0
Sierra Negra	4.452	2	0
Holuhraun ^a	4.305	1	0
Hudson	4.001	2	0
Nabro	3.65	1	0
Anatahan	2.818	2	1
Kasatochi	2.000	1	0
Fernandina	1.821	5	0
Galunggung ^b	1.730	24	0
Cerro Azul	1.229	3	0
Sarychev Peak	1.200	1	0
Mauna Loa	1.197	1	0
Kilauea ^b	1.147	39	3
Alaid	1.110	2	0
Wolf	1.080	1	0
Mt St Helens	0.910	1	1
Hekla	0.883	4	0
Chikurachki	0.830	7	0
Spurr	0.800	3	0
Ruiz	0.762	3	3
Krafla	0.685	3	0
Rabaul	0.531	3	2
Etna	0.505	51	3
Copahue	0.500	1	1
Eyjafjallajökull	0.466	1	0
Soputan	0.455	19	1
Lascar	0.455	3	3
Soufriere Hills ^b	0.450	17	3
Manam	0.420	12	2
Kluchevskoi	0.403	6	1
Fogo ^a	0.382	1	0
Kelut	0.350	2	0
Grimsvötn	0.350	4	0
Ulawun	0.347	5	2
P. de la Fournaise	0.334	9	0
Pagan	0.320	1	1
Redoubt	0.315	2	1
Merapi	0.310	2	2
Banda Api	0.250	1	0
Zhupanovsky	0.227	1	0
Colo	0.200	1	0
Marchena	0.200	1	0
Cordon Caulle	0.200	1	0
Tolbachik	0.200	1	0
Dalaffilla	0.150	1	0
Okmok	0.150	1	0
Lopevi	0.149	5	3
Pavlof	0.135	4	1
Ambrym	0.122	3	3
Asama	0.101	2	3
Sangeang Api	0.100	1	0
Tungurahua	0.090	5	3
Ruapehu	0.090	2	1
Popocatepetl ^b	0.087	5	3
Reventador	0.084	1	2
Ruang	0.080	1	0
Jebel at Tair	0.080	1	0
Shishaldin	0.063	1	1
Kizimen	0.060	2	0
Manda Hararo	0.060	2	0
Makian	0.050	1	0
Karkar	0.050	1	1
Mayon	0.050	1	3
Cameroon	0.048	2	0
Miyakejima	0.043	1	3
Karthala	0.041	4	0
Llaima	0.040	2	1
Paluweh	0.040	1	1
Bromo	0.031	1	3
Augustine	0.030	2	1
Nyiragongo	0.030	1	3
Bezymianny ^b	0.026	5	1
Home Reef	0.025	1	0

Table 6 (continued)

Volcano	Total SO ₂ emission (Tg)	Eruptions	Degassing style*
Pacaya	0.025	2	3
Oshima	0.022	1	1
Lokon-Empung	0.020	1	3
Sinabung	0.020	1	1
H. T. Hungaapai	0.014	1	0
Gamalama	0.013	2	1
Sheveluch ^b	0.012	2	3
Santa Ana	0.012	1	3

0: Entirely eruptive degassing. No significant passive SO₂ degassing known or reported.

1: Some passive degassing likely/measured/reported but eruptive degassing dominant.

2: Approximately equal passive/eruptive degassing.

3: Some eruptive degassing likely/measured/reported but passive degassing dominant.

* Qualitative assessment of degassing style:

^a Preliminary estimate of SO₂ emissions through the end of 2014.^b 'Eruptions' listed correspond to discrete eruptive events producing the measured SO₂ (although only a single long-lived eruption may be recorded in the VOTW 4.3 database).

2010 events at SHV; Figs. 4, 7, 13; Prata et al., 2007; Carn and Prata, 2010).

Smaller eruptive SO₂ emissions may be a consequence of shorter inter-eruptive repose times, precluding accumulation of a magmatic gas phase, and/or persistent degassing between eruptions that may bleed off accumulating gases (Table 6). In general, volcanoes exhibiting higher levels of passive degassing appear less prone to large explosive eruptions, although even Etna (one of the strongest sources of passive volcanic SO₂ emissions; Section 4.2) has a significant eruptive SO₂ flux (Table 6). Emissions from frequent, moderate-sized eruptions dominate the short-term eruptive volcanic SO₂ flux and in the tropics they have a higher climate impact than larger eruptions on a time-averaged basis due to their higher frequency (e.g., Miles et al., 2004). In the tropics, small eruptions can have disproportionate impacts on the UTLS due to vertical transport of SO₂ assisted by deep convection (e.g., Tupper et al., 2009; Tupper and Wunderman, 2009). Several of these modest tropical eruptions (e.g., 2002 Ruang, 2005 Manam, 2006 Soufriere Hills, 2006 Rabaul) have been responsible for small SAOD perturbations since 2000 (e.g., Figs. 9, 13), maintaining the stratospheric aerosol layer above background (non-volcanic) levels and likely playing a role in the global warming 'hiatus' observed since 1998 (e.g., Solomon et al., 2011; Vernier et al., 2011; Neely et al., 2013; Ridley et al., 2014; Santer et al., 2014, 2015; Carn et al., 2015).

A detailed examination of recent trends in SAOD (Figs. 9, 13) confirms the dominant influence of volcanic eruptions, but also shows that no major SO₂ emissions have eluded detection by satellite instruments. All major perturbations to SAOD since 1979 can be attributed to a known volcanic eruption, and the more comprehensive record of volcanic SO₂ emissions presented here also elucidates some of the more subtle SAOD variations apparent in Figs. 9 and 13. For example, the September 1994 eruption of Rabaul appears responsible for a small increase in SAOD above the post-Pinatubo background (Figs. 9, 13), and this and other eruptions (e.g., Manam in December 1996) may have prolonged the decay of the Pinatubo aerosol to background levels. It has been shown that the overall increase in SAOD observed since 2002 (Figs. 9, 13) is not due to increased anthropogenic SO₂ emissions, as had previously been suggested (Hofmann et al., 2009), but to moderate-sized volcanic eruptions (Neely et al., 2013). The frequency of UTLS SO₂ injections since 2002 has been sufficient to prevent the return of tropical SAOD to background conditions, although a slight decline in SAOD is apparent from 2012 to 2014 (Figs. 9, 13) due to reduced levels of volcanic activity (at the time of writing, more recent SAOD data were not available to gauge the atmospheric impact of the large eruptions of Kelut in February 2014 and Calbuco in April 2015). We note, however, that there are still discrepancies between SAOD observations and climate model simulations of stratospheric aerosol, even when the latter include forcing by the larger eruptions since 2002 (Neely

et al., 2013). This appears to be due to a combination of incomplete records of volcanic SO₂ forcing used to initialize model simulations, an over-emphasis on VEI as an indication of potential climate impact, and incorrect date assignments for some eruptions. For example, both Neely et al. (2013) and Santer et al. (2014) list a VEI 3 eruption of Anatahan on April 12, 2004. Although this is the date given for the onset of the 2004–2005 Anatahan eruption by the Smithsonian GVP, it actually represents the beginning of an extended phase of activity, which peaked in April–August 2005 with several VEI 3 eruptive events and UTLS SO₂ injections (Figs. 4, 5). Similarly, Vernier et al. (2011) ascribe minor stratospheric impact to an eruption of Shiveluch in May 2001 (based on its VEI of 4), but this produced no detectable SO₂ emissions and we therefore contend that a smaller (VEI 3) eruption of Ulawun on April 30, 2001 (which injected at least ~30 kt SO₂ into the tropical UTLS) was more significant. Thus in these cases the satellite measurements provide the key information on the timing and location of SO₂ emissions required for accurate climate model simulations. We expect that use of our comprehensive volcanic SO₂ emissions database will likely resolve some of the remaining discrepancies between model simulations and observations, and also assist efforts to identify periods when the stratosphere was truly free of any volcanic influence.

4.1.5. Lifetimes of SO₂ in the UTLS

The lifetime of SO₂ in UTLS volcanic plumes (typically represented by an e-folding time; i.e., the time for SO₂ mass reduction by a factor of e⁻¹) is one of the key factors determining the climate impact of an eruption (along with SO₂ amount, injection altitude and latitude; Robock, 2000). Observations of SO₂ lifetime can also be used to test climate model simulations of volcanic plumes and improve the chemical schemes embedded in such models. With growing numbers of volcanic eruptions observed from space, we have increasingly accurate constraints on SO₂ lifetimes in volcanic plumes at a range of altitudes and latitudes, and in all seasons. Fig. 14 shows a representative set of e-folding times for SO₂ removal from volcanic plumes, based on IASI

measurements for recent eruptions (since 2007) and values from the literature for older events and lower tropospheric SO₂ plumes. Note that the e-folding times shown in Fig. 14 characterize SO₂ removal (e.g., via gas- or aqueous-phase oxidation, cloud processing or wet/dry deposition) and not sulfate aerosol removal from the UTLS, which also influences climate impact.

The data in Fig. 14 confirm the strong correlation between SO₂ altitude and lifetime, with increasing variability apparent for lower tropospheric plumes. However, it is not absolute altitude but rather altitude relative to the local tropopause that is the main determinant of e-folding time, as evinced by the high-latitude eruptions of Sarychev Peak, Kasatochi, Cordon Caulle and Grimsvötn, among others (Fig. 14). These eruptions exhibit comparatively large e-folding times relative to the main trend defined by tropical eruptions, which is presumably due to the lower tropopause altitude at mid- to high-latitudes. Lower solar insolation at higher latitudes may also extend SO₂ lifetimes by reducing the rate of some photochemical reactions (e.g., the production of OH⁻). The 1991 Cerro Hudson eruption, which is also mid-latitude, is somewhat of an anomaly (Fig. 14), but a shorter than expected SO₂ lifetime in this case may be due to the timing of the eruption in the late southern hemisphere winter. With the exception of Cordon Caulle (which also has a slightly lower e-folding time; Fig. 14) the more recent high latitude eruptions all occurred in the spring or summer. There is thus evidence for both altitude and seasonal impacts on the SO₂ lifetime in volcanic plumes, which may be further elucidated as more eruptions are analyzed. The present dataset is too limited (and recent eruptions too small) to draw any conclusions regarding the effects of SO₂ loading alone on SO₂ lifetime, for example, if excessive stratospheric SO₂ loading could sufficiently deplete OH levels to promote extended SO₂ lifetimes (e.g., Bekki, 1995).

4.1.6. Volcanic HCl emissions into the UTLS

A striking feature of Fig. 4 that we highlight here is that MLS has detected injection of HCl into the UTLS by several explosive eruptions

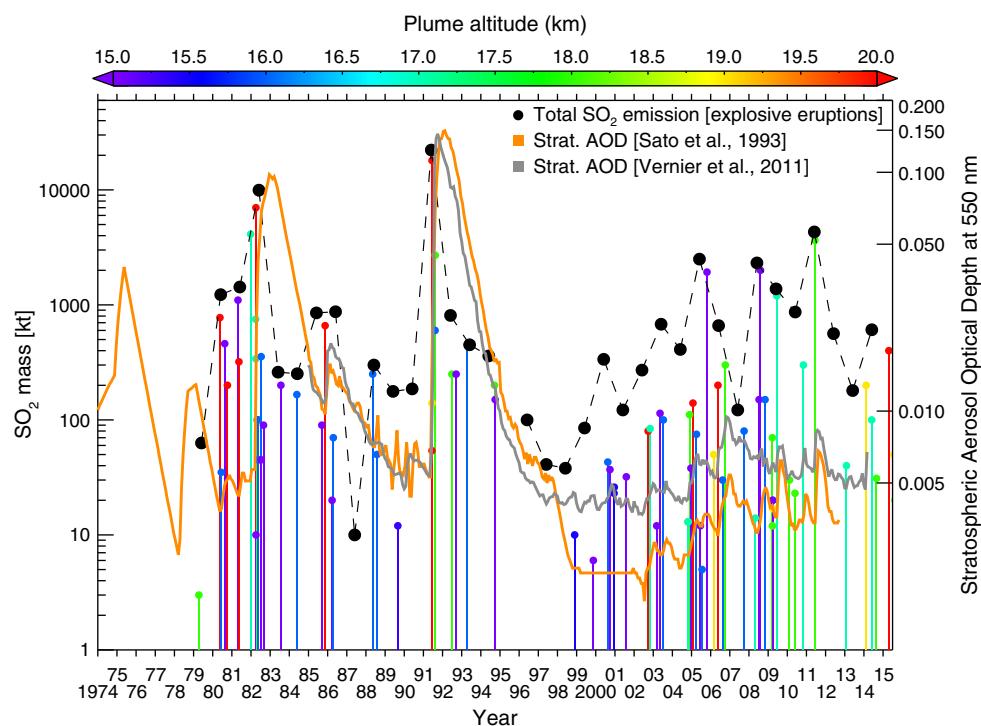


Fig. 13. Time-series of SO₂ mass loading similar to Fig. 1, but with only eruptions reaching altitudes of ≥ 15 km shown. The time axis is extended to include the 1974–1978 period prior to the satellite remote sensing era. Orange line shows stratospheric aerosol optical depth (SAOD) at 550 nm from Sato et al. (1993) and recent updates through December 2012 (<http://data.giss.nasa.gov/modelforce/straterr/>); gray line shows the tropical (30°S–30°N) SAOD at 525 nm data of Vernier et al. (2011; with recent updates) for 1985–2014 based on SAGE II, GOMOS and CALIPSO observations of aerosol between 15 and 40 km altitude. Differences between the SAOD datasets are explained in Fig. 9.

since 2004. We do not provide a detailed analysis of the MLS HCl data here, but rather point out the value of the MLS measurements (also including SO₂ and potentially other halogen species such as CH₃Cl; Table 1), which have not been widely used by the volcanological community (see also Pumphrey et al., 2015). HCl is an important halogen compound in the stratosphere. It is the dominant chlorine reservoir species and although it does not react directly with ozone, it is nevertheless a potential source of reactive halogen species (such as ClO) that destroy ozone, and hence any volcanic input of HCl into the stratosphere could be significant. Also notable in Fig. 4 is an inverted ‘tape-recorder’-like signal in the MLS HCl data (absent in the SO₂ data), with higher HCl mixing ratios transported to lower altitudes in the northern hemisphere winter. Since the major source of stratospheric HCl is the UV photolysis of chlorine-containing compounds (e.g., CFCs) in the upper stratosphere (note that Fig. 4 only extends to 47 hPa pressure), we assume that this pattern is due to tropical upwelling of HCl-poor air during the northern hemisphere summer, HCl production at higher altitudes, followed by HCl redistribution by the Brewer-Dobson circulation. The MLS HCl data at 147 hPa are noisy and although MLS HCl retrievals at pressures above 100 hPa are not recommended for scientific use (Livesey et al., 2011), the volcanic HCl signal in large eruption clouds is clearly strong enough to be detected at this level (Fig. 4).

Over the past few decades, opinions have varied on the potential for volcanic injection of HCl (and other halogens) into the stratosphere. Symonds et al. (1988) estimated an annual global volcanic HCl flux of 0.4–11 Tg and suggested that <10% of these emissions originated from large explosive eruptions that would transmit them efficiently to the stratosphere. However, direct measurements of stratospheric HCl after major eruptions found relatively modest (1982 El Chichón) or barely detectable (1991 Pinatubo) increases in HCl loadings above the stratospheric background (Mankin and Coffey, 1984; Mankin et al., 1992; Wallace and Livingston, 1992), although the observations were made weeks or months after the eruptions (and both eruptions produced measurable ozone depletion). Subsequent modeling work by Tabazadeh and Turco (1993) suggested that scavenging of volcanic HCl by condensed supercooled water in Plinian eruption columns could effectively remove more than 99% of emitted chlorine, precluding stratospheric HCl injection, whilst having minimal impact on less soluble SO₂. However, Textor et al. (2003) pointed out flaws in the Tabazadeh and Turco (1993) study, namely that they used a relatively simple treatment of the dynamics, omitted some detailed cloud microphysical processes, and assumed that all hydrometeors were liquid (ice was later discovered to be dominant in some volcanic clouds; e.g., Rose et al., 1995). Accounting for the effects of ice particles, Textor et al. (2003) concluded that ice could scavenge 50–90% of HCl and 10–30% of SO₂ in eruption columns, and predicted that >25% and 80% of emitted HCl and SO₂, respectively, could potentially reach the stratosphere during explosive eruptions.

Recent satellite observations seem to confirm the predictions of Textor et al. (2003), and suggest that explosive eruptions frequently inject detectable HCl into the stratosphere (Fig. 4; e.g., Prata et al., 2007; Theys et al., 2014). Note that daily maximum SO₂ and HCl mixing ratios retrieved by MLS are shown in Fig. 4, and some weaker volcanic signals may be omitted, although the larger eruptions clearly stand out. Furthermore, due to the limited spatial sampling of MLS (Section 3.5) some smaller eruptions may not be detected, or the regions of highest SO₂ and HCl concentrations in volcanic clouds may not be sampled. Table 7 lists some HCl and SO₂ mixing ratios measured by MLS in volcanic clouds since 2004, along with estimated HCl/SO₂ mass ratios calculated from collocated SO₂ and HCl vertical profiles. Actual volcanic SO₂ and HCl concentrations are likely to be underestimated by MLS since the vertical extent of volcanic plumes is typically much smaller than the MLS vertical resolution of ~3 km (Livesey et al., 2011; Theys et al., 2014). Furthermore, since SO₂ and HCl are retrieved using data from separate MLS radiometers with different fields of view, the averaging effects differ for the two gases: the vertical × horizontal (cross-

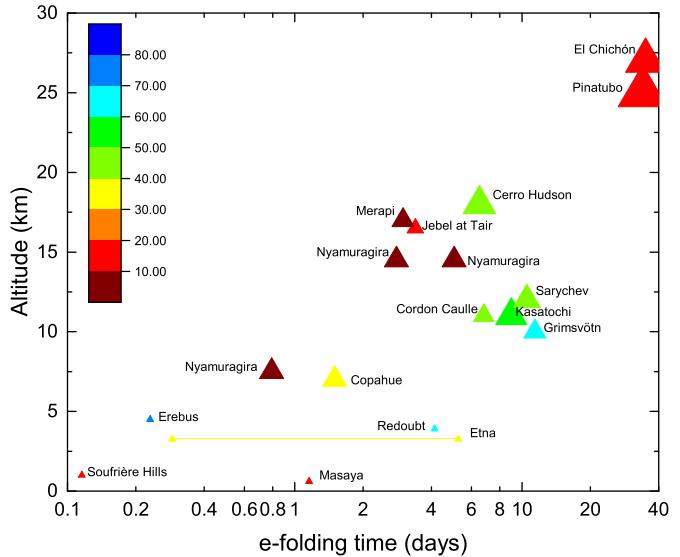


Fig. 14. Altitude of SO₂ injection against e-folding time for SO₂ removal for selected volcanic eruptions and emissions since 1980, with e-folding times derived from the literature (pre-2007) and from IASI SO₂ measurements (2007–2014). Symbol size is proportional to total SO₂ emission, and symbol color indicates volcano latitude. The SO₂ emissions represented are from: Erebus 1980 (Radke, 1982), Nyamuragira 1981 (Krueger et al., 1996; Bluth and Carn, 2008), Nyamuragira 1980–2004 (Bluth and Carn, 2008), El Chichón 1982 (Bluth et al., 1997), Pinatubo 1991 (Bluth et al., 1997), Etna 1983 (Martin et al., 1986), Redoubt 1990 (Hobbs et al., 1991), Cerro Hudson 1991 (Constantine et al., 2000), Soufrière Hills 1996 (Oppenheimer et al., 1998), Masaya 2003 (McGonigle et al., 2004b), Soufrière Hills 2004 (Rodríguez et al., 2008), Jebel at Tair 2007 (Clarisse et al., 2008), Kasatochi 2008 (Krotkov et al., 2010), Sarychev Peak 2009 (Haywood et al., 2010), Cordon Caulle 2011 (Theys et al., 2013), Merapi 2010 (IASI derived), Grimsvötn 2011 (IASI derived), and Copahue 2012 (IASI derived). Several e-folding times below 0.1 days were reported by Oppenheimer et al. (1998) for lower tropospheric SO₂ plumes but are not shown here.

track) × along-track resolution of MLS SO₂ and HCl retrievals in the lower stratosphere is ~3 × 6 × 180 km and ~3 × 3 × 300–400 km, respectively (Livesey et al., 2011). Thus, since MLS averages volcanic SO₂ and HCl concentrations over different atmospheric volumes, coupled with limited spatial sampling, we treat the HCl/SO₂ ratios in Table 7 with caution. Nevertheless, some broad conclusions can be drawn. The HCl/SO₂ ratios observed in volcanic eruption clouds (~0.01–0.03; Table 7) are around an order of magnitude lower than values (~0.1–0.3) estimated for other eruptions based on petrological data or in-situ measurements (e.g., Westrich and Gerlach, 1992; Gerlach et al., 1996; Rose et al., 2006). This may partly reflect uncertainties on the MLS-derived ratios, but would also be consistent with greater scavenging of HCl relative to SO₂ in eruption plumes (Textor et al., 2003). Observed variations in HCl/SO₂ ratios between eruptions may be a consequence of variable HCl/SO₂ ratios in the source emissions (related to depth of degassing) or may reflect variable plume liquid water or total water content. Fig. 4 shows a notable contrast in HCl signal between the 2008 Kasatochi and 2009 Sarychev Peak eruptions, which were similar in magnitude. Sampling issues notwithstanding, a possible explanation for the difference is that liquid water was more abundant in the Kasatochi eruption plume, since it erupted through a pre-existing crater lake (Waythomas et al., 2010), resulting in more effective scrubbing of HCl in the emissions. The 2011 Nabro eruption also produced a surprisingly strong UTLS HCl signal (Figs. 4, 10), given its non-subduction tectonic setting (northeast Afar, Eritrea) where Cl enrichment from subducted seawater is absent. However, in this case the MLS-derived HCl/SO₂ ratio of 0.03 is roughly commensurate with ratios measured in tropospheric plumes at other East African Rift volcanoes (~0.04 at Erta ‘Ale and ~0.03 at Nyiragongo; Sawyer et al., 2008a, 2008b), perhaps suggesting minimal scavenging of HCl in the Nabro plume (or similar scavenging efficiencies of SO₂ and HCl).

4.2. Passive volcanic SO₂ degassing

We have shown above that the flux of SO₂ from volcanic eruptions is well constrained, with the exception of lower magnitude events (VEI ≤ 2). However, there is more uncertainty on the global flux of SO₂ emitted by passive (i.e., non-eruptive) degassing (e.g., Oppenheimer et al., 2011; Shinohara, 2013), which also propagates into estimates of volcanic fluxes of other gases, such as CO₂ (e.g., Burton et al., 2013). The ground-based perspective on global volcanic degassing must be aggregated from diverse sources, including journal publications, volcano observatories, instrument networks (e.g., Galle et al., 2010), and Smithsonian GVP reports, and consequently takes considerable time to generate and update. Satellite measurements offer the significant advantage of providing a daily, global snapshot of volcanic SO₂ emissions, collected by a single instrument using the same retrieval technique, and thus provide more timely and consistent assessments of the dominant volcanic SO₂ sources at a given time.

Currently, OMI, GOME-2 and OMPS can provide daily observations of tropospheric volcanic SO₂ plumes from space (e.g., Carn et al., 2007, 2008b, 2013; McCormick et al., 2012, 2013; Yang et al., 2013), and the quality of measurements will improve with future sensors such as the Tropospheric Monitoring Instrument (TROPOMI; <http://www.tropomi.eu>). Measurement noise and variable spatial resolution limits the SO₂ sources that can be detected in a single, daily satellite measurement to the strongest emitters (OMI has greater sensitivity than GOME-2 or OMPS, due to higher spatial resolution), but spatial and temporal data averaging techniques can also be used to detect weaker SO₂ sources (e.g., Fioletov et al., 2011, 2013). The latter are less useful for volcano monitoring purposes, where timeliness is critical, but can be used to generate accurate, annual SO₂ emissions inventories based on satellite observations. Here, we have used the technique described in Section 3.4 to identify the most persistent volcanic SO₂ sources detected by OMI in the 2004–2014 period (Fig. 15).

Results of the daily OMI SO₂ data analysis are shown in Fig. 15. In these plots higher average values of the OSI generally indicate stronger SO₂ emissions; hence the observations of Anatahan and Nevado del Ruiz, for example, indicate shorter periods of strong SO₂ degassing in 2004–2013. The SO₂ emissions from four volcanoes (Ambrym, Kilauea, Popocatepetl and Nyiragongo-Nyamulagira) have been notably more persistent than others in the ~10 years of satellite data, and SO₂ emissions from Ambrym and Nyiragongo were also detected prior to 2004 in TOMS SO₂ data (e.g., Carn, 2004). Three of these persistent SO₂ sources host at least one active lava lake (Ambrym, Kilauea, and Nyiragongo-Nyamulagira), reflecting efficient and persistent SO₂ outgassing from low-viscosity, basaltic magmas. Although many of the detected sources (e.g., Etna, Bagana, Nevado del Ruiz) figure prominently in previous volcanic SO₂ emissions inventories (e.g., Andres and

Kasgnoc, 1998), the global OMI measurements permit identification of sources in poorly monitored regions such as Papua New Guinea, Indonesia, Vanuatu and Kamchatka, and also provide unique insight into the interannual variability of SO₂ emissions (Fig. 15). For example, Sabancaya (Peru) and Sinabung (Indonesia) do not appear as strong SO₂ sources based on OMI measurements in 2004–2013 (Fig. 15A), but are prominent in 2014 due to recently increased activity (Fig. 15B; Global Volcanism Program, 2014).

Although we do not convert the OMI SO₂ measurements into SO₂ emission rates here (techniques for doing this are under development; e.g., Carn et al., 2013), recent compilations of ground-based SO₂ measurements confirm high SO₂ fluxes (exceeding ~10 kt/day at times) at Ambrym, Kilauea, Popocatepetl, Nyiragongo, Etna and Bagana, among others (e.g., Oppenheimer et al., 2011; Elias and Sutton, 2012; Shinohara, 2013). We also note that several of these volcanoes (Nyiragongo, Popocatepetl, Ambrym, Etna) are regarded to be dominant sources of volcanic CO₂ emissions (Burton et al., 2013). However, other sources with previously reported high SO₂ emission rates, such as Láschar (N. Chile; 2.4 kt/day; Mather et al., 2004) are conspicuously absent from Fig. 15, and this appears to be corroborated by more recent (post-2004) ground-based SO₂ measurements indicating substantially reduced SO₂ emissions from Láschar (~0.1–0.6 kt/day; Henney et al., 2012; Tamburello et al., 2014; Lopez et al., 2015). We therefore contend that UV satellite measurements of tropospheric SO₂ plumes provide arguably the timeliest information on the status of global volcanic SO₂ sources, especially considering that it is crucial to constrain the strongest sources in order to estimate global or regional SO₂ fluxes (e.g., Mori et al., 2013). We are not aware of any volcano with a reported SO₂ emission rate of 1 kt/day or more since 2004 that has not been detected at some point in OMI SO₂ measurements. As SO₂ fluxes from an increasing number of weaker volcanic SO₂ sources are being measured using sensitive ground-based or airborne techniques (e.g., McGonigle et al., 2004a; Mori et al., 2006; Bani et al., 2013; Lopez et al., 2013b; Tamburello et al., 2014; Saing et al., 2014; Lopez et al., 2015; Smeekens et al., 2015; Stebel et al., 2015), combining these new data with satellite observations will result in more accurate volcanic SO₂ emissions inventories, encompassing a wider range of sources.

Whilst the tropospheric SO₂ plume detection method employed here (Section 3.4) aims to omit major eruptive emissions (since eruptive SO₂ clouds typically drift rapidly away from the source, beyond the 50 km search radius), SO₂ plumes associated with continuous eruptive activity may still be captured. It is either known or likely that much of the SO₂ detected at Dukono, Anatahan, Kliuchevskoi, Zhupanovsky, Soufrière Hills, Sakura-jima and perhaps others is associated with continuous eruptions, i.e., the emissions are not passive in the strict sense (involving no coeval eruption of magma). Both Kilauea's and Bagana's SO₂ emissions are also associated with ongoing, long-term effusive eruptions (Elias and Sutton, 2012; McCormick et al., 2012) and hence could be regarded as eruptive discharges. Satellite observations of volcanic ash could potentially be used to distinguish plumes emitted by continuous eruptions, but ash measurements are typically less sensitive and noisier than SO₂ measurements. Although of minor importance when attempting to estimate total volcanic SO₂ fluxes to the atmosphere, the distinction between passive and eruptive degassing becomes important when assessing volcanic hazards or calculating volumes of degassed magma stored in the crust based on observed SO₂ emissions.

To visualize the geographical distribution and temporal variation of the major tropospheric volcanic SO₂ sources, the latitudes of the tropospheric SO₂ plumes detected by OMI in 2004–2014 are shown in Fig. 16 (note that some eruptive degassing is also apparent). Persistent volcanic SO₂ sources are manifested as continuous horizontal 'bands' on this plot (Fig. 16): prominent near-continuous bands occur at ~20°N (Kilauea and Popocatepetl; with increased SO₂ degassing apparent in March 2008 associated with the summit eruption at Kilauea; Elias and Sutton, 2012); ~1–2°S (Nyiragongo and Nyamuragira); ~4–9°S (volcanoes of Indonesia and Papua New Guinea); and ~16°S (Ambrym and

Table 7
HCl and SO₂ mixing ratios measured by Aura/MLS in volcanic clouds.

Volcano	Eruption Date	HCl (ppbv)	SO ₂ (ppbv)	Pressure (hPa)	HCl/SO ₂
Manam	Jan 27, 2005	4–6	279	68–100	0.01
Anatahan	Apr 6, 2005	3.5	133	100	0.02
Soufrière Hills	May 20, 2006	3	200	68	0.01
Chaitén	May 6, 2008	1.6	28	147	0.03
Okmok	Jul 12, 2008	5	212	147	0.01–0.02
Kasatochi	Aug 7, 2008	5–6	392	68–215	0.01–0.014
Redoubt	Mar 26, 2009	4–5	175	100–215	0.02
Sarychev Peak	Jun 15, 2009	7–9	529	32–215	0.03
Merapi	Nov 5, 2010	6–7	172	100–215	0.03
Cordón Caulle ¹	Jun 4, 2011	2–3	77	147–215	0.03
Nabro	Jun 15, 2011	9	306	46–215	0.03
Paluweh	Feb 2, 2013	2.2	129	100–215	0.01
Kelut	Feb 14, 2014	7	398	32–147	0.01
Sangeang Api	May 31, 2014	2–3	53	146	0.03

1. Data from Theye et al. (2014).

other Vanuatuan volcanoes such as Aoba and Gaua). Other notable features of Fig. 16 include elevated SO₂ emissions from Anatahan (~16°N) in 2005–2008 with a subsequent decline in 2009–2014 (McCormick et al., 2015), increased SO₂ emissions from Nevado del Ruiz (~5°N) in early 2012 (Global Volcanism Program, 2012), and at Turrialba (~10°N) in 2009–2010 (Campion et al., 2012). There is also a striking contrast between the tropics and high latitudes (Fig. 16). This partly reflects the distribution of SO₂ sources (most of the dominant volcanic SO₂ sources are in the tropics) but is also a result of seasonal variations in the availability and sensitivity of UV measurements at high latitudes, and higher wind speeds at extratropical latitudes (which dilute SO₂ column amounts in tropospheric plumes). Detection of degassing from Etna (~37°N), Kamchatkan volcanoes (~50–60°N) and southern Chilean volcanoes (~35–40°S) shows a clear seasonal dependence with wintertime observational gaps. However, the absence of a persistent SO₂ degassing

signal in Iceland (~63–66°N) is probably genuine, due to a lack of eruptive SO₂ emissions from Icelandic volcanoes.

One conclusion that can be drawn from this analysis is that more ground-based measurements of SO₂ emissions from high-latitude volcanoes (e.g., in Kamchatka, the Kurile Islands and the Aleutian Islands), especially in winter, are required to improve volcanic gas emissions budgets. However, ground-based UV remote sensing techniques are also likely to be challenging in the relative darkness and adverse weather conditions of high-latitude winter, although IR spectroscopy and camera techniques have potential as they are not dependent on UV radiation (e.g., Lopez et al., 2013b; Prata and Bernardo, 2014; Lopez et al., 2015). Moderate- to high-resolution IR satellite retrievals of lower tropospheric SO₂ (e.g., MODIS, ASTER) may also be possible for some plumes but routine detection of passive SO₂ degassing is likely to be restricted by thermal contrast and sensitivity issues.

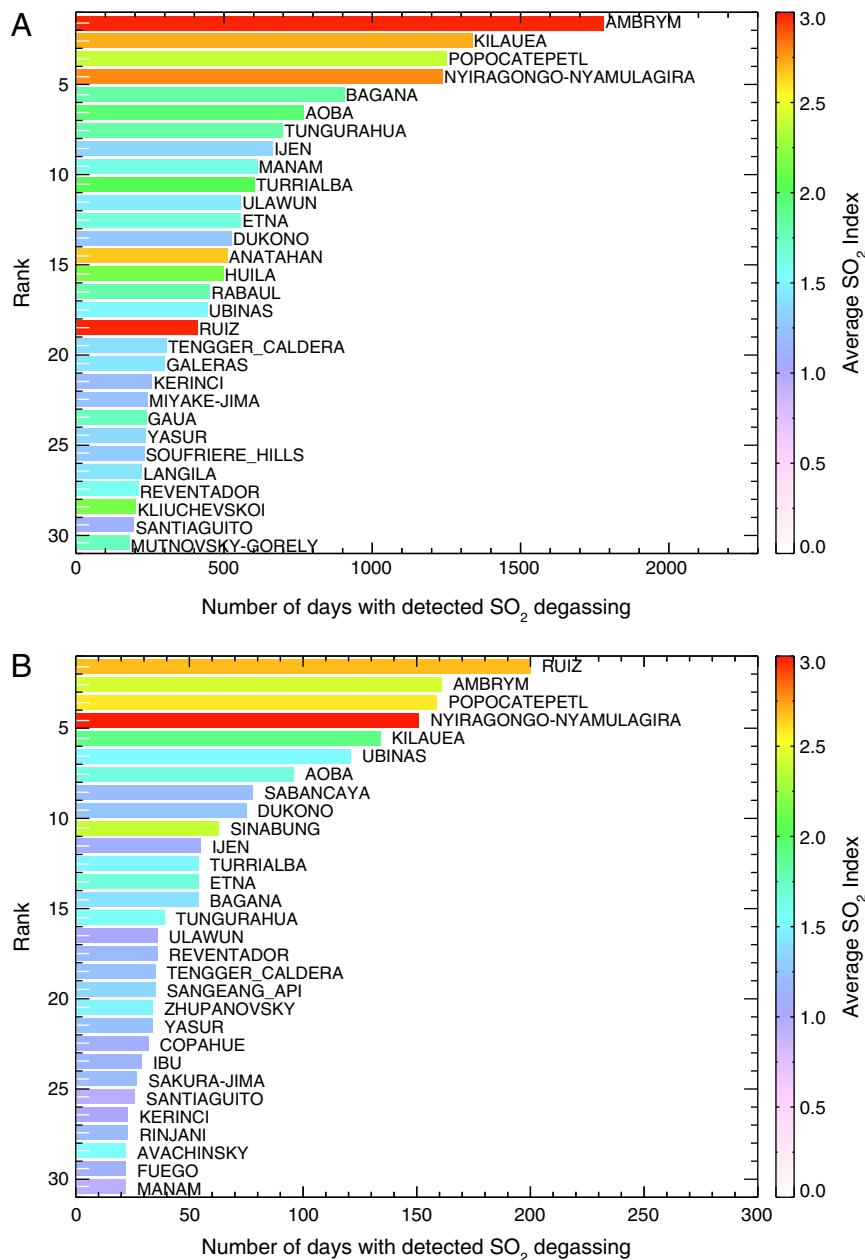


Fig. 15. Sources of passive volcanic SO₂ emissions detected by OMI: (A) Most persistent sources (in rank order) detected in OMI data collected between September 2004 and December 2013; (B) Most persistent sources detected by OMI in 2014. Colors correspond to the average value of the OSI for each volcano. Note that for some volcanoes in close proximity, the source of observed SO₂ emissions can be ambiguous (e.g., Nyiragongo-Nyamulagira, DR Congo; Mutnovsky-Gorely, Kamchatka).

5. Discussion

With at least 16 satellite instruments capable of measuring volcanic SO₂ emissions currently in orbit (Table 1), the flow of SO₂ observations from space has never been more profuse. This availability of remote sensing assets for detecting volcanic SO₂ is set to continue for at least the next decade, as degrading instruments or those nearing the end of their operational lifetimes are supplanted by sensors such as TROPOMI, ensuring that the SO₂ flux from volcanic eruptions will remain well constrained for the foreseeable future. Furthermore, should another Pinatubo-scale or larger eruption occur in the near future, current satellite measurements are capable of providing much higher quality data than they were in 1991. A Pinatubo-scale SO₂ emission could potentially be tracked for weeks or months with the improved sensitivity of existing sensors relative to TOMS. Open questions to consider include whether an even larger SO₂ release would saturate current SO₂ retrieval algorithms during the initial emission, precluding accurate SO₂ mass loading measurements until the cloud began to disperse, and how a persistent, widespread SO₂ and sulfate aerosol veil from such a large event would impact remote sensing measurements during its atmospheric residence.

Although SO₂ emissions are reasonably well constrained, a major unresolved question concerns the magnitude of global volcanic emissions of other sulfur species, most notably H₂S, which have been previously estimated at 1.4–35 Tg/yr. of S (e.g., Halmer et al., 2002; Oppenheimer et al., 2011). Hydrogen sulfide can comprise up to ~40% of total sulfur gases (SO₂ + H₂S) in high-temperature volcanic gas samples (Oppenheimer et al., 2011), and petrological evidence suggests that it may be dominant in some eruptions. Luhr et al. (1984) proposed that most of the sulfur gases released by the 1982 El Chichón eruption were in the form of H₂S, and Hobbs et al. (1982) reported significant quantities of H₂S in the May 1980 Mt. St Helens eruption cloud. Although IASI is capable of detecting volcanic H₂S from space (Table 1), the only reported observations to date are for the 2008 Kasatochi and 2011 Grimsvötn eruptions (Clarisse et al., 2011a,b; Sigmarsson et al., 2013). These new observations of eruptive H₂S emissions are valuable, though satellite measurements of H₂S will never be as comprehensive as SO₂ measurements since the IR H₂S absorption lines are weak and hence sensitivity is low (Clarisse et al., 2011b).

There appears to be significant scope for more work on the rates of key reactions involved in H₂S oxidation, but McKeen et al. (1984) suggest that H₂S is oxidized to SO₂ relatively quickly (~3 day chemical lifetime) provided OH is readily available. Hence the degree to which satellite measurements will capture H₂S emissions converted to SO₂ will depend on the relative rates of H₂S oxidation to SO₂ and SO₂ conversion to sulfate, i.e., if H₂S is oxidized relatively quickly, the SO₂ produced will be detected and factored in to the total eruption sulfur budget. However, if OH concentrations were sufficient to promote efficient H₂S oxidation, then it also likely that SO₂ would be simultaneously oxidized. Oxidation of H₂S has been invoked to explain increases in measured SO₂ observed in the days following some explosive eruptions, such as the 1982 El Chichón and 1992 Mount Spurr eruptions (e.g., Bluth et al., 1995; Schneider et al., 1999; Rose et al., 2000, 2001), but such increases could also be reasonably attributed to other causes such as a reduction in volcanic cloud optical depth, reduced ash content, or sequestration of SO₂ by ice in the young volcanic cloud (e.g., Rose et al., 2000, 2001), and are often subject to some ambiguity.

Although some H₂S is likely to be present in all volcanic eruption plumes, it may be safe to assume that SO₂ dominates the sulfur inventory in emissions from volcanoes in subduction zone environments (where most of the major volcanic SO₂ sources are located), where oxidized magmas with high SO₂:H₂S ratios are typical. Magmas in hot-spot and divergent plate contexts may also be relatively oxidized (Oppenheimer et al., 2011). Aiuppa et al. (2005) report a SO₂/H₂S molar ratio of 20 in emissions from Etna in 2003, and H₂S was not detected in emissions from Gorely volcano (Kamchatka) in 2011 (Aiuppa

et al., 2012), although Edmonds et al. (2010) found a relatively significant quantities of H₂S in gas emissions from SHV (Montserrat) in July 2008 during an eruptive pause (SO₂/H₂S = 1.9–3.6). Emissions of H₂S may be more significant during precursory activity at wet volcanoes, where phreatomagmatic eruptions may precede the main magmatic eruption phase, as was suggested for Kasatochi (Clarisse et al., 2011b). Clearly, however, further work is needed to better constrain volcanic H₂S emissions on a global scale. Global S emissions from volcanic arcs have been estimated to be ~10–11 Tg/year (Hilton et al., 2002; Shinohara, 2013), equivalent to ~20–22 Tg/yr. SO₂. Assuming ~16 Tg/yr. SO₂ emitted by passive degassing in volcanic arcs (Shinohara, 2013) and ~2 Tg/yr. SO₂ emitted by explosive eruptions (Table 3) yields a total of ~18 Tg/yr. SO₂ or 9 Tg/yr. S. Based on an estimated annual emission of 10–11 Tg S, this leaves a 1–2 Tg/yr. S shortfall, which if emitted as H₂S would be ~1.2–2.1 Tg/yr. H₂S.

One approach that could provide further insight into eruptive emissions of H₂S is to identify significant discrepancies between petrological estimates of S in pre-eruptive magmatic gas phases (Scaillet and Pichavant, 2003; Scaillet et al., 2003) and observations of SO₂ emissions. Such disagreement could indicate the discharge of substantial H₂S emissions that were not detected in satellite data (or alternatively, rapid removal of SO₂). One possible example is the April 1979 eruption of Soufrière St Vincent, for which Scaillet et al. (2003) predicted a total S release of ~0.5 Tg from the melt and gas phase (i.e., ~1 Tg SO₂). However, only ~3 kt of SO₂ (0.3% of the predicted SO₂ yield) was detected by TOMS following the eruption, despite adequate measurement conditions (a volcanic ash cloud was detected). Furthermore, the eruption has been linked to a large SAOD anomaly in 1979–80 (Fig. 13; Rosen and Hofmann, 1980), which would not be commensurate with such a low SO₂ release. Taken together, this evidence may suggest that the bulk of the sulfur emitted by Soufrière St Vincent was in the form of H₂S, which may have been oxidized sufficiently slowly to prevent any SO₂ detection by TOMS.

Finally, although we do not discuss them in detail here, UV satellite measurements are also providing unique observations of reactive halogen species such as BrO and OCIO in volcanic clouds (e.g., Afe et al., 2004; Theys et al., 2009; Rix et al., 2012; Hörmann et al., 2013; Theys et al., 2014). Such measurements permit assessment of the potential impacts of volcanic eruptions on stratospheric ozone (e.g., Kutterolf et al., 2013). The HCl and SO₂ profile measurements from MLS are also advantageous, since the impact of a volcanic eruption on the stratospheric ozone layer will depend primarily on the altitude of the volcanic emissions relative to the peak in the ozone concentration. Theys et al. (2014) found significant differences in the quality of volcanic BrO and OCIO retrievals from available UV satellite instruments (SCIAMACHY, OMI and GOME-2), with certain sensors optimized for particular volatile species, highlighting the fact that multiple instruments are required to ensure accurate measurements of all volcanic emissions. Another potentially interesting but as yet unexplored aspect of satellite measurements of reactive halogens in volcanic clouds is the different overpass times of the sensors. For example, we might expect systematic differences between BrO measurements by GOME-2 (9:30 am overpass) and OMI (1:45 pm overpass) due to photochemical effects on BrO production in volcanic plumes (e.g., von Glasow et al., 2009), although other factors such as spatial resolution and plume aerosol loading will also play a role.

6. Conclusions

In this contribution we have reviewed ~36 years of satellite measurements of volcanic SO₂ emissions from 1978 to 2014. During this period, SO₂ emissions from every significant volcanic eruption (VEI ≥ 4) have been detected and quantified, providing accurate constraints on the mean volcanic SO₂ flux to the UTLS (~1–2 Tg/yr) and allowing robust interpretation of trends in SAOD. Measurements from multiple UV and IR satellite instruments are required to achieve optimum

coverage, to ensure detection of eruptions at all latitudes and at night, and to reduce uncertainty on measured SO₂ loadings for individual eruptions. Microwave sensors such as MLS can provide crucial complementary information on the altitude of volcanic SO₂ and HCl injections. Although there is greater uncertainty on SO₂ emissions from smaller eruptions (VEI 0–3), this SO₂ is not expected to be significant for climate. New techniques being adopted for ground-based measurements of volcanic SO₂ (e.g., UV and IR imaging cameras) will likely improve constraints on SO₂ fluxes from small eruptions, which may be an important component of the tropospheric volcanic SO₂ budget in years with low total SO₂ emissions or no large eruptions. Furthermore, CO₂ emissions from such activity may be more significant due to the longer atmospheric residence time of CO₂ relative to SO₂.

Reanalysis of the relationship between VEI and SO₂ emissions with the inclusion of recent satellite measurements confirms a first-order relationship between eruption magnitude and SO₂ loading. However, due to significant variability in SO₂ loading for individual VEI classes (coupled with the inherent challenges in assigning accurate VEIs), the use of VEI to assess the potential climate impact of eruptions in the remote sensing era is discouraged. Current satellite measurements can provide accurate and timely estimates of both SO₂ loading and altitude, with this capability expected to continue in the next decade, obviating the need for any proxy for climate impact (but the VEI certainly remains a useful indicator of the overall significance of an eruption). Specification of eruption altitude is crucial for climate modeling (e.g., Robock, 2000; Arfeuille et al., 2014), and one of the weaknesses of our current volcanic SO₂ emissions database is that eruptions are characterized by a single altitude, whereas in reality the vertical profile of an explosive eruption column can be complex and variable. The assumption of a single SO₂ injection altitude can result in significant errors in the UTLS SO₂ budget, especially for complex, continuous eruptions (e.g., the 2010 Eyjafjallajökull, 2011 Nabro and 2014–15 Holuhraun eruptions), which will propagate into estimates of climate impacts. In this regard, the application of inverse modeling techniques to estimate the syn-eruptive vertical profile of SO₂ emissions appears particularly powerful, and is facilitated by the availability of multiple UV and IR satellite measurements of volcanic SO₂ emissions. Such techniques could be used to determine the peak injection altitude or multiple injection altitudes.

For the past decade, satellite instruments have also provided new constraints on passive volcanic degassing of SO₂. A synthesis of ~10 years of OMI SO₂ measurements reveals the most persistent volcanic SO₂ sources detected from space, several of which have minimal or no ground-based monitoring currently in place. Our analysis demonstrates the key role of satellite observations in identifying the major volcanic SO₂ sources at any given time. It has been postulated that a few dominant SO₂ sources exert a major control over regional and global volcanic SO₂ fluxes (e.g., Mori et al., 2013; Shinohara, 2013), and hence a combination of satellite SO₂ measurements of the major SO₂ emitters with the increasingly comprehensive ground-based SO₂ data for volcanoes with lower SO₂ emission rates will lead to vastly improved SO₂ emissions budgets. However, observations of SO₂ emissions from high-latitude volcanoes (e.g., in Kamchatka, the Kurile Islands and the Aleutian Islands) remain challenging, especially in winter.

Collation of a multi-decadal record of volcanic SO₂ emissions allows us to draw some broad conclusions from the patterns that emerge. Volcanoes that exhibit minimal levels of passive or inter-eruptive SO₂ degassing are responsible for sporadic ($\geq 10^2$ years), large (≥ 1 Tg) SO₂ emissions with potentially significant impacts on climate or SAOD (e.g., Pinatubo, El Chichón, Kasatochi, Nabro); it is highly likely that the next, major SO₂-producing eruption will occur at a volcano that has not erupted for centuries or more, where accumulation of a stored vapor phase may be occurring. We also note that such volcanoes may emit substantial amounts of CO₂ in apparently quiescent periods, and more widespread ground-based or airborne CO₂ flux measurements would be desirable; the challenge is to select appropriate targets from the many currently dormant volcanoes (active ground deformation may be one useful criterion). Perhaps more significant for climate on shorter timescales are the restless, volatile-rich systems exhibiting both passive and eruptive SO₂ degassing (e.g., Rabaul, Manam, SHV, Ulawun) that produce smaller (VEI ≤ 4) but frequent UTLS eruptions, which may have significant cumulative impacts on climate. For such systems, the magnitude of SO₂ release is likely limited (≤ 1 Tg is typical) by near-continuous activity and bleeding of volatiles from resident magma at depth. This effect is even stronger for other systems where passive SO₂ degassing dominates the total SO₂ discharge and large eruptive SO₂ emissions appear unlikely unless passive degassing is curtailed (e.g., Popocatepetl, Tungurahua).

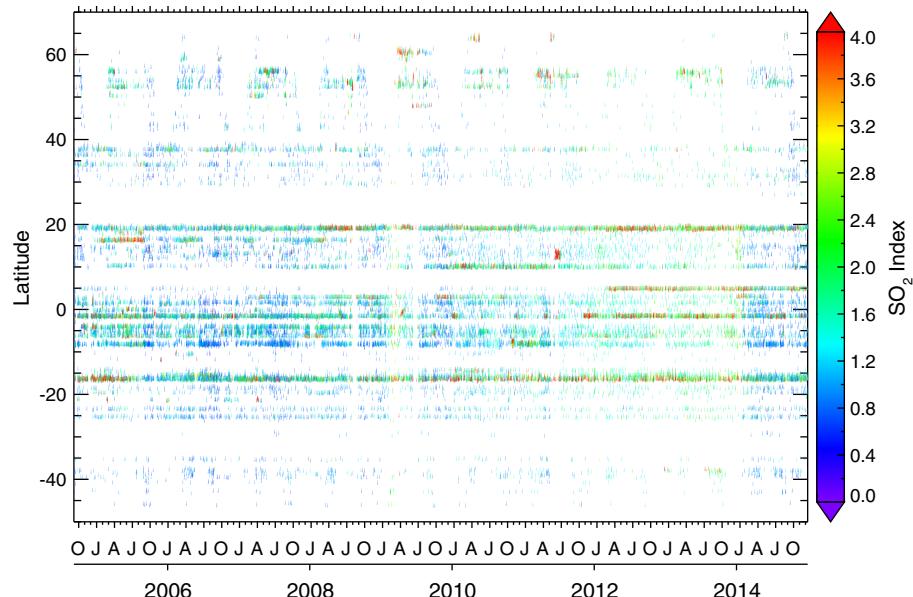


Fig. 16. Latitude and strength of persistent volcanic SO₂ sources against time based on OMI SO₂ measurements from September 2004 to December 2014. For each day, the latitudes of all OMI pixels that show the highest daily values of the OMI SO₂ Index (OSI; see Section 3.4 for definition) and are located within 50 km of an active volcano are plotted. Persistent SO₂ sources thus appear as horizontal bands on this plot. Although most eruptive SO₂ emissions are excluded from this plot (see Section 3.4), short discontinuities in the data are due to large SO₂-rich eruptions temporarily overwhelming the OSI measurements (e.g., the Kasatochi eruption in August 2008, the Sarychev Peak eruption in June 2009, and the Nabro eruption in June 2011).

Although the SO₂ data products from both UV and IR satellite instruments enjoy a long heritage and can be deemed relatively mature, some outstanding questions remain. Unraveling the causes of differences between UV and IR SO₂ retrievals, or between IR retrievals in different spectral bands, found for some eruptions (e.g., Table 2) is needed to reduce the uncertainties on some eruption SO₂ loadings. Such differences are most likely to originate from a complex combination of effects of SO₂ altitude or vertical profile, overpass timing, plume aerosol loading (e.g., ash, sulfate, hydrometeors), sensor spatial and spectral resolution, atmospheric water vapor, and total column ozone, and may be very challenging to untangle. Efforts should be made to determine the ‘optimum’ satellite SO₂ retrieval for each eruption. Validation of satellite SO₂ measurements remains a challenge, particularly for the largest volcanic clouds, but may become more achievable using unmanned aerial vehicles (UAVs) or other rapidly-deployable in-situ measurement techniques. Finally, volcanic H₂S emissions remain poorly constrained on a global scale and may be a significant fraction of the total S budget for some eruptions.

Although much can be achieved using satellite instruments deployed on multiple polar-orbiting platforms (and such observations are crucial for high latitude volcanoes), monitoring of volcanic SO₂ emissions from space would greatly benefit from a global, geostationary capability, particularly in the UV. Currently, only the European SEVIRI and Japanese Advanced Himawari Imager (AHI) sensors have IR channels capable of detecting SO₂ emissions from geostationary orbit (Table 1). However, prospects for high temporal resolution UV monitoring of SO₂ from geostationary or alternative orbits are improving. The recently deployed EPIC instrument on the DSCOVR satellite at the L₁ Earth-Sun. Lagrange point will soon provide multiple daily UV SO₂ retrievals for the sunlit Earth disk (including some coverage of high latitudes during the summer months). The SO₂ sensitivity of EPIC will likely be intermediate between TOMS and OMI, permitting detection of large eruptions and strong passive SO₂ degassing. Later in the decade (around 2017), the Tropospheric Emissions: Monitoring of Pollution (TEMPO) instrument will be launched on a geostationary satellite, providing UV observations of North America (including Mexico). Since SO₂ is a criteria pollutant affecting air quality, volcanic SO₂ surveillance will indirectly benefit from efforts (such as the TEMPO mission) to measure anthropogenic SO₂ emissions more frequently and track intercontinental transport of air pollution. TEMPO will form part of a global geostationary constellation for pollution monitoring, including UV instruments from Europe (Sentinel 4) and Asia (the Geostationary Environment Monitoring Spectrometer [GEMS] being developed in Korea). We are therefore on the cusp of another major advance in our ability to measure volcanic SO₂ emissions from space.

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SO₂ emissions database described in this paper is available from the NASA Goddard Earth Sciences (GES) Data and Information Services Center (DISK) as a level 4 MEASUREs (Making Earth System Data Records for Use in Research Environments) data product (Carn, 2015c).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.jvolgeores.2016.01.002>.

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