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First synoptic analysis of volcanic degassing in Papua New Guinea

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[1] We report the first satellite-based survey of volcanic sulphur dioxide (SO_2) degassing in Papua New Guinea, using Ozone Monitoring Instrument (OMI) data. OMI is sensitive to low-level passive degassing. These observations are useful for volcano monitoring, hazard assessment (particularly aviation hazard) and assessment of arc geochemical budgets and are of immense value in remote regions with little ground-based instrumentation, such as Papua New Guinea. We identify Bagana, Manam, Rabaul, Ulawun and Langila as the active sources of volcanic SO_2 in Papua New Guinea, with Bagana being the largest source. We present an OMI SO_2 time series for 2005–2008 and a total detected regional output of $\sim 1.8 \times 10^9$ kg SO_2. About 40% of emissions were released by major eruption events at Manam (January 2005), Bagana (June 2006) and Rabaul (October 2006). Over the past century however, we estimate that major explosive eruptions contribute <5% of the arc-scale SO_2 emission budget. Ground-based DOAS measurements of SO_2 degassing at five of Papua New Guinea's volcanoes are compared with our OMI observations. The total OMI SO_2 output is only ~20% of the total extrapolated from DOAS, a discrepancy which we demonstrate is consistent with other volcanic arcs. Therefore, the true total regional SO_2 output may be considerably higher than that detected by OMI. Uncertainties in the OMI SO_2 data include the effects of in-plume chemical processing and dilution of SO_2 prior to the satellite overpass, OMI's reduced sensitivity to low levels of SO_2 in the planetary boundary layer and interference by meteorological clouds.

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1. Introduction

[2] Papua New Guinea ranks among the most active volcanic regions on Earth, with seventeen historically active volcanoes (Figure 1). Since 2000, major explosive eruptions of Volcanic Explosivity Index (VEI) 4 have occurred at Rabaul (October 2006), Manam (January 2005) and Ulawun (September 2000) [*Venzke et al.*, 2002–2011]. These volcanoes, and Langila, have also shown more modest-scale eruptive activity throughout this period and near-continuous lava effusion has been observed at Bagana since 1947 [*Bultitude*, 1976]. Mild activity has been observed at Pago and the Garbuna Group since 2000 [*Venzke et al.*, 2002–2011]. A powerful explosive eruption occurred at Rabaul in 1994 [*Rose et al.*, 1995; *Roggensack et al.*, 1996]. A period of intense activity occurred during 1972–75, with major eruptions at Ritter Island and Karkar after ~80 years repose, the largest eruption of Langila for a century and strong activity at Manam, Ulawun and Bagana [*Johnson*, 1976; *Cooke et al.*, 1976]. Significant earlier explosive eruptions are known from Bagana (1952), Lamington (1951), Rabaul (1937) and Manam (1919) [*Venzke et al.*, 2002–2011].

[3] An understanding of Papua New Guinea's volcanoes is very important. Major volcanic eruptions in the region have caused widespread injury, fatalities, health hazards, settlement destruction and crop damage [*Cooke et al.*, 1976; *Venzke et al.*, 2002–2011]. Papua New Guinea lies near the busy air corridor connecting Australia and New Zealand to Japan and southeast Asia and regular explosive activity at several volcanoes requires airline pilots to divert their course around volcanic plumes [*Tupper et al.*, 2004]. Gas emissions, particularly those of SO_2 due to its relative ease of measurement, are widely used as a tool in volcanic monitoring and hazard management [*Edmonds et al.*, 2003; *Aiuppa et al.*, 2008; *Oppenheimer*, 2010] making it important to understand the patterns of degassing at active volcanic systems, especially preceding and following eruptions. Furthermore, the region plays an important, if poorly constrained, role in terms of the global volcanic degassing budget; a widely cited study of global volcanic SO_2 degassing lists Manam, Langila, Ulawun, Rabaul and Bagana among the major sources [*Andres and Kasgnoc*, 1998]. Accurate quantification of regional and global volcanic degassing budgets is a key goal for understanding volcanic impacts on the atmosphere and environment over local, regional and global scales [*Graf et al.*, 1997; *Robock*, 2000; *Delmelle*, 2003]. This is especially true in terms of SO_2 as, again due to its relative ease of measurement, budgets of other atmospherically important volcanic gaseous emissions are often estimated by scaling using the SO_2 flux (e.g., the halogens [*Pyle and Mather*, 2009]). Better constraints on the SO_2 emissions of major volcanic arcs are also crucial to our understanding of the role they play in returning subducted volatiles to Earth's atmosphere [e.g., *Wallace*, 2005]. Knowledge of the recycling of sulphur through subduction zones and the resulting fluxes into the atmosphere relies on, among other things, accurate measurement of the style, volume and persistence of degassing [*Wallace*, 2005; *Mather et al.*, 2006; *Wallace and Edmonds*, 2011].

[4] Degassing from Papua New Guinea's volcanoes remains comparatively understudied however; barring an isolated ground-based study in 2003 [*McGonigle et al.*, 2004a], volcanic SO_2 release is poorly quantified. This can partly be attributed to the logistical difficulties of extended field campaigns in what is a largely inaccessible and sometimes politically unstable (Bougainville) region. Furthermore, the remote and heavily vegetated volcanoes do not present easy targets for installing and maintaining continuous monitoring systems and there is little funding for such work. An opportunity to redress this lack of data is offered by the Ozone Monitoring Instrument (OMI), a satellite instrument with sensitivity to lower tropospheric SO_2, whose spatial and temporal coverage enables the generation of good quality long-term emission data sets [*Carn et al.*, 2008; *Bani et al.*, 2011]. In Papua New Guinea, such data sets constitute the only regular degassing information available, and when combined with other observations (ground-based measurements or observations, pilot reports or other satellite-based data) may provide powerful and synoptic insights into the behavior of the volcanoes in remote volcanic regions.

[5] This satellite-based study is the first detailed long-term assessment of volcanic SO_2 degassing in Papua New Guinea. We present a four-year (2005–2008) time series of daily OMI measurements of volcanic SO_2 mass burdens over the region, and identify the actively degassing volcanoes. We determine the relative strength of each source, and illustrate the patterns apparent in the OMI degassing data set via comparison with ground-based observations of regional volcanism, additional satellite-based measurements where available and previously published studies. We place new constraints on Papua New Guinea's overall volcanic SO_2 budget and evaluate the relative importance of explosive SO_2 emissions versus non-explosive and non-eruptive degassing to the total burden measured. Last,

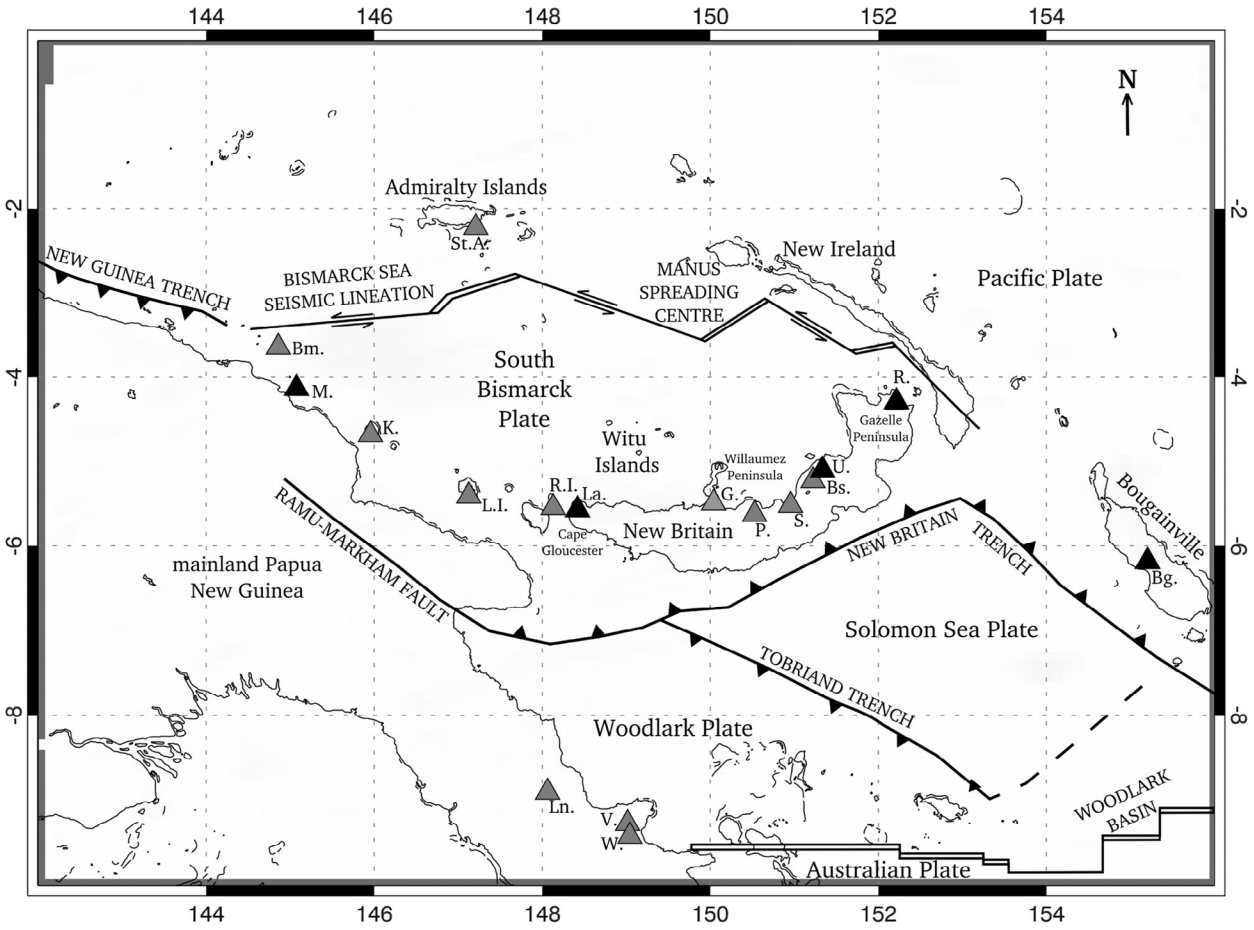

Figure 1. Geological setting of the study region, Papua New Guinea. Major tectonic and geographical features are shown. Triangles represent historically active volcanoes. Black triangles represent those volcanoes shown by this study to be current active sources of SO_2. Volcano names are abbreviated as follows: Bam (Bm.), Manam (M.), Karkar (K.), Long Island (L.I.), St Andrew's Strait (St.A.), Lamington (Ln.), Ritter Island (R.I.), Langila (La.), Victory (V.), Waiowa (W.), Garbuna Group (G.), Pago (P.), Sulu Range (S.), Bamus (Bs.), Ulawun (U.), Rabaul (R.), Bagana (Bg.).

we assess the applicability of OMI observations to detecting and monitoring volcanic SO_2 emissions in the region, particularly from non-explosive or passive degassing, and propose future avenues of research to exploit the great potential offered by the OMI SO_2 data set further.

1.1. Geological Setting

[6] Papua New Guinea lies in the southwest Pacific Ocean in a tectonically complex buffer zone between the obliquely converging Pacific Ocean and India-Australian tectonic plates (Figure 1). The interactions of the smaller Woodlark, South Bismark and Solomon Sea plates govern the expression of volcanism and tectonics in the region [*Honza et al.*, 1987; *Tregoning et al.*, 2000; *Woodhead et al.*, 2010]. Most active volcanism in Papua New Guinea is concentrated along the Bismarck arc, a chain of volcanoes stretching ~1000 km from Bam Island in the northwest of the region to the Gazelle Peninsula of eastern New Britain in the east [*Johnson*, 1976]. The other main center of activity currently is on Bougainville Island, in the northern Solomons [*Bultitude*, 1976]. Historical eruptions are also known from the Admiralty Islands and southeastern mainland Papua regions [*Davies and Smith*, 1971; *Page and Johnson*, 1974; *Smith*, 1981]. The Bismarck arc volcanism is driven by the northward subduction of the Solomon Sea plate beneath the South Bismarck Plate, at a rate of 90–120 mm yr^{-1} [*Woodhead et al.*, 1998, 2010]. West of New Britain, this convergence is accommodated along the Ramu-Markham fault, at ~67 mm yr^{-1} [*Tregoning et al.*, 2000] with the volcanoes of the western Bismarck arc lying offshore to the northeast.

Volcanism on Bougainville is similarly associated with subduction of the Solomon Sea plate, but northeastward beneath the Pacific plate [*Bultitude*, 1976]. Estimates of convergence range from 78 to 113 mm yr^{-1} [*Bultitude*, 1976] to 145 mm yr^{-1} [*Tiffin et al.*, 1987].

[7] Of the seventeen historically active volcanoes across the region, five are the focus of this paper and are hence discussed further here: from west to east, Manam, Langila, Ulawun, Rabaul and Bagana (Figure 1).

[8] Manam is a 1807 m-high basaltic stratovolcano on the north coast of mainland New Guinea. One of the most active volcanoes in the region, Manam's longest repose time over the last century was around 9 years, with regular activity being observed since the 1870s [*Palfreyman and Cooke*, 1976]. The characteristic activity comprises series of strombolian paroxysms every few days. The violence of this activity is widely variable: minor ejections of ash are typical, while more vigorous activity may be accompanied by lava flows. Rarer large sub-plinian explosive eruptions also occur, with associated pyroclastic flows sometimes reaching the sea via four evenly spaced avalanche valleys in the edifice flanks [*Palfreyman and Cooke*, 1976]. The erupted products are dominantly low-TiO_2 basalts with minor low-silica (SiO_2 < 53 wt%) andesites [*Johnson et al.*, 1985].

[9] Langila is a 1330 m-high complex volcano in the Cape Gloucester region of New Britain [*Venzke et al.*, 2002–2011]. Activity today is spread over three active craters and comprises strombolian (and occasionally vulcanian) explosive activity, often accompanied by lava flows. Extensive lava flows blanket the area just north of Langila [*Cooke et al.*, 1976]. Eruptive activity at Langila restarted in 1953 after an eighty year repose, and has been constant since; due to the volcano's remoteness however only the most violent activity tends to be reported, and there is frequently uncertainty as to which is the active crater [*Cooke et al.*, 1976]. Only a single petrological analysis has been published for Langila; the lavas are described as low-Si andesites [*Cooke et al.*, 1976].

[10] Ulawun is a 2334 m-high stratovolcano with a history of frequent and violent strombolian eruptions in recent times [*Venzke et al.*, 2002–2011]. Eruptions since the 1970s have been of greater intensity than the preceding 100 years and have involved lava flows and major pyroclastic avalanches [*Melson et al.*, 1972; *Johnson et al.*, 1983]. These avalanches have been linked to instability-driven edifice collapse, which is considered the most significant hazard associated with Ulawun. Ulawun's lavas are uniformly tholeiitic basalts, with minor andesites: average SiO_2 is ~52.75 wt% and total alkalis are <2.8 wt% [*Melson et al.*, 1972; *Cooke et al.*, 1976]. Persistent SO_2 emissions from Ulawun between its eruptions have been attributed to open-vent conditions enabled by the continued absence of any summit lava dome [*Johnson et al.*, 1983].

[11] Rabaul is a large caldera system on the Gazelle Peninsula of eastern New Britain. Activity has comprised large caldera-forming eruptions of rhyolitic to dacitic composition every 2000 years, with smaller-scale basaltic to andesitic composition activity centered on small satellite cones around the caldera rim, with a 30–60 year repeat cycle [*Cunningham et al.*, 2009]. Presently activity is focused on the 688 m-high Tavurvur cone, though Vulcan has been active as recently as 1995. Minor ash explosions are typical, interspersed with persistent passive degassing. In terms of major explosive eruptions, Rabaul has been the most active volcano in Papua New Guinea during historical times [*Venzke et al.*, 2002–2011].

[12] Bagana is a 1750 m-high lava cone, composed of blocky lava flows and minor pyroclastic material [*Bultitude*, 1976]. It is arguably the most active of the volcanoes in the region, with almost-continuous effusion of lava; at the current production rate the entire edifice could have been constructed in only a few hundred years [*Venzke et al.*, 2002–2011; G. Wadge, personal communication, 2011]. Rarer explosive activity has been observed, and emission of thick white vapor from the summit vent is continuous. Bagana lavas are typical low-Si andesites, with limited dacites are also present [*Blake*, 1968; *Bultitude*, 1976].

1.2. Previous Work Quantifying SO_2 Degassing in Papua New Guinea

[13] Several of Papua New Guinea's volcanoes are regarded as major contributors to the global atmospheric SO_2 budget. *Andres and Kasgnoc* [1998] identified Rabaul as a major source via sporadic releases, based on the large SO_2 emission detected via satellite observations of the VEI 4 explosive eruption in September 1994 [*Rose et al.*, 1995; *Roggensack et al.*, 1996]. Bagana, Manam, Langila and Ulawun were all listed among the largest sources of continuous SO_2 emission, with reported SO_2 fluxes of 1204×10^6 kg yr^{-1}, 335×10^6 kg yr^{-1}, 252×10^6 kg yr^{-1} and 175×10^6 kg yr^{-1}, respectively measured by correlation spectroscopy

(COSPEC). Bagana was identified as the second most significant continuous volcanic SO_2 point source globally, after Etna. The overall annual arc production for Papua New Guinea by continuous degassing was estimated at $\sim 2 \times 10^9$ kg yr^{-1}, or ~20% of the overall global SO_2 emission from continuous volcanic sources [*Andres and Kasgnoc*, 1998].

[14] More recent estimates of SO_2 emission rates for six of Papua New Guinea's volcanoes were derived using miniature UV differential optical absorption spectrometers (DOAS) [*McGonigle et al.*, 2004a]. Fluxes calculated from measurement of the volcanic plumes at Bagana, Manam, Langila, Ulawun, Rabaul and Pago were 725×10^6 kg yr^{-1}, 66×10^6 kg yr^{-1}, 91×10^6 kg yr^{-1}, 233×10^6 kg yr^{-1}, 41×10^6 kg yr^{-1} and 44×10^6 kg yr^{-1} respectively. These SO_2 fluxes are generally lower than those reported by *Andres and Kasgnoc* [1998], possibly due to the lower levels of activity noted when the measurements were made in July and August 2003 [*McGonigle et al.*, 2004a]. These fluxes nevertheless suggest the arc still contributes 6–9% of the total global time-integrated volcanic SO_2 emissions to the atmosphere [*Andres and Kasgnoc*, 1998] or 7–12% of emissions to the troposphere [*Halmer et al.*, 2002].

[15] However, these estimates of the SO_2 emission from Papua New Guinea's volcanoes have their limitations. The flux values in the work of *Andres and Kasgnoc* [1998], primarily measured by COSPEC, are sparse and no sense of variations in atmospheric or operational conditions is provided, nor of prevailing volcanic activity. COSPEC measurements are typically subject to errors of ±14–42% [*Stoiber et al.*, 1987]. No specific data sources are detailed for these flux estimates, which prevents any reprocessing or data filtering in this regard. The SO_2 fluxes for sporadically degassing volcanoes in the work of *Andres and Kasgnoc* [1998] are derived from satellite-based Total Ozone Mapping Spectrometer (TOMS) measurements, which have an accuracy of ±10–30% [*Krueger*, 1995]. The extrapolated SO_2 emission cited for Rabaul by *Andres and Kasgnoc* [1998] is based on TOMS detection of the September 1994 eruption, the first day of which saw ice in the volcanic plume significantly hamper accurate retrievals [*Rose et al.*, 1995; *Roggensack et al.*, 1996]. More recent ground-based measurements only provide a snapshot of volcanic degassing in Papua New Guinea [*McGonigle et al.*, 2004a], and therefore extrapolating an arc-scale budget from a very short observation period is subject to obvious uncertainty. In this study we explore the long-term data set offered by OMI to evaluate whether it provides a more rigorous estimate of SO_2 emissions from Papua New Guinea's volcanoes.

2. Data Collection and Methodology

2.1. The Ozone Monitoring Instrument

[16] OMI was launched in July 2004 aboard NASA's Aura satellite, which flies in the "A-train" constellation as part of the Earth Observation System (EOS) program [*Levelt et al.*, 2006; NASA, Introducing the A-train, 2010, http://www.nasa.gov/mission_pages/a-train/a-train.html, accessed June 2011]. OMI is a hyperspectral UV-visible solar backscatter spectrometer, whose unique combination of the optimal features of several earlier instruments offers unprecedented sensitivity to the atmospheric concentrations of various trace gases, including SO_2. Aura flies in a sun-synchronous polar orbit, crossing the Equator at 13:45 local time (ascending node) at an altitude of 705 km. OMI is a non-scanning push-broom instrument; a large 114° field of view results in a 2600 km-wide swath, which together with Aura's 14 daily orbits enables continuous daily global coverage. The swath is binned into pixels measuring 24 × 13 km at nadir, the highest spatial resolution yet for a satellite-based UV/VIS spectrometer. OMI measures top-of-atmosphere (TOA) radiances, resulting from solar backscatter and reflection from the Earth's surface and atmosphere. This radiance spectrum is normalized to the solar irradiance spectrum before being used to derive information about trace gas concentration and other atmospheric parameters [*Levelt et al.*, 2006]. OMI measures radiances in both UV (270–365 nm) and visible (365–500 nm) spectral regions, with an average resolution of ~0.5 nm, the best yet for such an instrument. Consideration of the full UV/VIS spectrum (hyperspectral imaging) enables the simultaneous detection of the trace gases O_3, SO_2, NO_2, BrO, OClO and HCHO [*Ahmad et al.*, 2003].

2.2. The Linear Fit Retrieval Algorithm

[17] Operational SO_2 data is retrieved using the Linear Fit (LF) algorithm which simultaneously derives O_3, SO_2 and effective surface reflectivity using ten discrete wavelength bands exhibiting both strong and weak O_3 and SO_2 absorption [*Yang et al.*, 2007]. The retrieval uses look-up tables generated from TOMS-V8 climatological ozone and temperature profiles, satellite viewing geometry and OMI-measured effective surface/cloud pressures in a forward radiative transfer model and the retrieved

SO_2 columns are adjusted to minimize discrepancy between the measured and calculated radiances. The Linear Fit technique superseded the Band Residual Difference (BRD) algorithm [*Krotkov et al.*, 2006] which underestimated SO_2 columns of >10–20 Dobson Units (DU) due to the breakdown of linear proportionality between absorbance and absorber concentration. The LF algorithm accounts for this by using longer wavelengths for the retrieval, which are less prone to saturation and nonlinearity than those used by the BRD algorithm [*Yang et al.*, 2007]. The LF algorithm has been shown to be internally consistent in its recovery of atmospheric parameters, and includes a sliding median residual correction to reduce along- and cross-track biases.

[18] Accurate retrieval of vertical SO_2 columns requires a realistic Air Mass Factor (AMF) to convert measured slant columns into vertical columns, and the AMF in turn depends on the vertical distribution of SO_2 which is generally unknown. To circumvent this problem, the LF algorithm uses an assumed SO_2 center of mass altitude (CMA). Four different vertical SO_2 columns, each corresponding to a distinct a priori SO_2 CMA, are retrieved: planetary boundary layer (PBL; CMA = 0.9 km), lower troposphere (TRL; CMA = 2.5 km), mid-troposphere (TRM; CMA = 7.5 km) and lower stratosphere (STL; CMA = 17 km). In order to minimize the potential error in the SO_2 retrieval, the most appropriate CMA must be carefully identified. This may be accomplished via comparison between ancillary observations of plume altitude, typically from ground observations, pilot reports [*Tupper and Wunderman*, 2009] or satellite-based data from instruments such as the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) lidar [*Winker et al.*, 2003].

[19] Observations of this kind are sparse in Papua New Guinea, and pilot reports of volcanic plumes tend to refer to infrequent ash-rich plumes originating from short-lived bursts of explosive activity, rather than the passive degassing which is most frequently ongoing. We assume that typical plumes during passive degassing generally rise ≤1 km above the summit of the volcanoes. Summit elevations for the volcanoes observed by OMI range from 688 m to 2334 m. Therefore we use the TRL retrieval throughout; note that it slightly overestimates the mass burden of SO_2 plumes higher than 2.5 km. During the major eruptions of Manam (January 2005) and Rabaul (October 2006), the SO_2 plume reached 21–24 km [*Tupper et al.*, 2007] and 18 km [*Smithsonian Institution*, 2006b] respectively. In both instances, SO_2 mass burdens were calculated interactively offline, using the STL retrieval, which was designed for application to volcanic clouds in the upper troposphere/lower stratosphere (UTLS) and is the most sensitive retrieval to SO_2, due to lower noise in the OMI data. Due to the weak altitude dependence of the retrieval algorithm's averaging kernel in the UTLS, actual cloud height can be represented as 17 km without significant error (~10–15%). We note that maximum reported plume heights may be greater than the height of maximum SO_2 injection.

2.3. Data Selection and Processing

[20] Science-quality OMI SO_2 data gathered from January 2005 to December 2008 was downloaded from the NASA Mirador online repository (http://mirador.gsfc.nasa.gov/) and archived on a local server. From August 2008, an enigmatic pattern of interference known as the 'row anomaly' became evident in OMI data, changing dynamically with latitude and season, and resulting in anomalous radiance data in a number of OMI CCD detector rows. Accurate SO_2 retrievals are not possible in the pixels affected by the row anomaly, which became a noticeable interference from January 2009 onwards. The precise cause of the row anomaly is poorly understood, but its evolution has been monitored (see http://www.knmi.nl/omi/research/product/rowanomaly-background.php). Pending further investigation into this anomaly, data from January 2009 onwards has not been considered for this work. Nonetheless, our 4-year data set enables the first long-term study of the degassing behavior of Papua New Guinea's volcanoes. Data was processed using OMIPLOT [*Carn*, 2011]. Daily total SO_2 burdens across the Papua New Guinea region (Figure 1) were calculated, with accompanying volcanic SO_2 plume distribution maps. OMIPLOT automatically calculates total SO_2 mass, SO_2 plume area, identifies the strongest SO_2 source, and also computes regional average reflectivity values. To identify the strongest source, OMIPLOT selects the active volcano (based on the Smithsonian Global Volcanism Program database) closest to the center coordinates of the OMI pixel containing the maximum retrieved SO_2 column concentration which is generally found in the largest plume present in any given scene [*Carn et al.*, 2008]. Due to the high density of actively degassing volcanoes in Papua New Guinea, and OMI's single daily overpass, plume transport by wind may advect a plume over a downwind volcano that is then misidentified as the source. This data is therefore best considered as a broad overview, and is best interpreted alongside daily OMI images and independent

reports of volcanic activity, such as MODVOLC thermal alerts [*Wright and Flynn*, 2004; *Wright et al.*, 2004]. Daily regional average reflectivity is used as a proxy for meteorological cloud cover, which is important since extensive cloud cover at comparable or greater altitudes may mask or otherwise interfere with OMI's detection of an SO_2 cloud. Monthly mean and cumulative SO_2 distribution maps were also generated by OMIPLOT. Such time-averaging or summing increases signal to noise in the output images, aiding overall plume coherency and enabling improved visualization of degassing patterns at the expense of temporal resolution [*Carn et al.*, 2008]. Such images provide a clear overview of the changing emissions from the region's volcanoes, with daily images then used in more focused studies, generally of shorter duration.

[21] OMI measures total column SO_2, which is converted to SO_2 mass burden, and not emission rate. The region shown in Figure 1 is observed by OMI for ~2 min during each overpass and therefore the burdens measured are near-instantaneous, with the duration of the volcanic event responsible generally unknown. Future studies will consider the precise relationship of OMI SO_2 burdens and source emission fluxes by utilizing accurate plume height estimates, detailed local meteorological information and an appreciation of both atmospheric reactions converting SO_2 into sulphate and various wet/dry deposition processes. It is possible to estimate flux from satellite data by measuring the concentration area of traverse sections through the plume and multiplying by the wind velocity [e.g., *Carn and Bluth*, 2003]. A good correlation has been demonstrated between OMI and ASTER (Advanced Spaceborne Thermal Emission and Reflection radiometer) flux estimates at Etna, Popocatépetl, Nyamuragira and Nyiragongo (R. Campion, personal communication, 2011).

2.4. Data Validation and Error Discussion

[22] A robust comparison between OMI and independently obtained SO_2 data is essential for validation purposes. At present, OMI SO_2 retrievals have only been validated in a limited number of cases. A study of the volcanic plume from the 2008 eruption of Okmok volcano (Aleutian Islands) showed that ground-based DOAS and simultaneous OMI measurements provide consistent SO_2 columns in a mature stratospheric volcanic cloud, under optimal viewing conditions [*Spinei et al.*, 2010]. An opportunistic study comparing OMI and ground-based mobile UV spectrometer (FLYSPEC) measurements of the eruption cloud from the 2009 eruption of Sarychev Peak (Kurile Islands) as it drifted eastward over Alaska also demonstrated good agreement between the data sets [*Carn and Lopez*, 2011]. A comparison of SO_2 retrievals between several UV and IR satellite-based sensors (MODIS, AIRS, OMI) for four recent volcanic eruptions suggested that OMI was the most sensitive to SO_2, and was the only sensor currently capable of resolving low altitude volcanic clouds [*Thomas et al.*, 2011]. However, there has been little attempt at validation of OMI's ability to measure volcanic SO_2 released into the troposphere from lower intensity or passive volcanic degassing. *Carn et al.* [2008] demonstrated OMI's sensitivity to changes in volcanic SO_2 emission from the volcanoes of Ecuador and southern Colombia and OMI observations correlated with independent ground-based records of degassing. Aircraft-based measurements over Ecuador in July 2007 sampled in situ SO_2 columns which, though close to OMI's detection limit, showed reasonable agreement with the average SO_2 column amounts retrieved [*Carn et al.*, 2011].

[23] Further validation is particularly required for OMI detection of SO_2 in the troposphere because the measurement is complex, with many potential sources of systematic and random error. First, compared to stratospheric measurements of SO_2, the SO_2 absorption signal in the retrieved TOA radiance is weaker, owing to the more extensive scattering and attenuation of photons along greater path lengths through the atmosphere. This causes an increase in noise and hence detection limit with decreasing SO_2 altitude. A solution to this problem, utilized here, is to average OMI data over larger areas and longer time intervals, to increase the signal-to-noise ratio and thus lower the detection limit.

[24] Second, the retrieval does not account for the dual effects of volcanic cloud transport and in-plume chemical processing of SO_2. These processes are complex and difficult to isolate. Wind transports plumes away from the volcano and also shears them laterally so that, while the same mass of SO_2 may be present, the vertical column density is reduced, and therefore is harder for OMI to detect. Several chemical reactions may contribute to SO_2 loss from the plume, with oxidation to sulphate aerosol and various wet/dry deposition mechanisms particularly significant [*Eatough et al.*, 1994]. Attempts have been made to characterize the tropospheric lifetime of volcanic SO_2 [*Oppenheimer et al.*, 1998; *McGonigle et al.*, 2004b; *Rodríguez et al.*, 2008] but the results vary widely (e-folding times of 0.28–2778 h). Plume lifetime seems to vary strongly with differing

Table 1. Locations of Three Background Regions Close to Papua New Guinea

Region	Latitude; Longitude	Daily Average SO_2, $\times 10^6$ kg
pngback1 (north of region)	0–10°N; 142–156°E	0.087
pngback2 (northeast of region)	0–10°N; 158–172°E	0.091
pngback3 (east of region)	0–10°S; 158–172°E	0.081

atmospheric conditions in different settings, and therefore without detailed modeling of atmospheric chemistry and transport for the area of interest, supplemented with real measurements of atmospheric constituents and meteorological conditions, we can make no definitive statement on SO_2 loss and dilution rates in plumes over Papua New Guinea. It is, however, possible to make a first order distinction between two end-member scenarios: (1) rapid loss and transport will reduce plume mass greatly during the 24 h separating OMI observations and lead to potential mass underestimates; (2) very slow loss and negligible dispersal will lead to plumes persisting for >24 h and the same SO_2 potentially being measured on successive days.

[25] To estimate whether SO_2 plumes are generally persistent or transient over Papua New Guinea, we analyzed monthly average OMI scenes. Using estimates of regional wind velocity [*McKee et al.*, 1985] and the maximum distances downwind reached by observed detectable plumes, we estimated typical maximum plume ages. We note, from study of several representative monthly average scenes, that the maximum age of detectable volcanic plumes is ~18 h and that the typical ages are considerably lower, <12 h. Therefore, unless SO_2 mass burdens are particularly high or wind velocities are unusually low, it appears that rapid processing and transport will tend to reduce the detected mass of plumes significantly within 24 h. Therefore, in Papua New Guinea it seems likely that SO_2 emitted in the hours following OMI's overpass will not persist for a sufficiently long time to ensure measurement on the following day and that OMI measurements may therefore underestimate the region's SO_2 emissions.

[26] Another potential source of error in the OMI SO_2 retrieval is the use of an incorrect CMA. Detailed error analysis is not possible however in the absence of accurate independent knowledge of SO_2 plume altitudes. Increasing the CMA from TRL to TRM increases the AMF and therefore decreases the retrieved SO_2 mass by ~30% for a general midlatitude case [*Carn et al.*, 2008]. Other complicating factors include the effects of meteorological cloud above the plume, interference by ash and suboptimal observation parameters, such as off-axis swath positions and low solar zenith angles. The most significant of these is likely to be interference by cloud, which we assess using OMI's observed effective reflectivity over the domain, and discuss in further detail below.

[27] Last, OMI data contains a certain element of random noise. To assess the magnitude of this noise, three domains west of Papua New Guinea, over the open Pacific Ocean, containing no actively degassing volcanoes and of comparable size to the study area as defined in Figure 1 were analyzed for their apparent daily SO_2 burdens during 2005–2008. Drifting SO_2 plumes from eruptions at Indonesian volcanoes (February 2005), Anatahan in the Marianas (April, August 2005), Soufrière Hills volcano (June 2006), Rabaul (October 2006) and Alu/Dalaffilla volcano in Ethiopia (November 2008) were removed from the data and replaced with a 7-day mean of the previous days' plume-free data. The daily average SO_2 burden for each area in 2005–2008 is shown in Table 1. The overall mean mass error attributed to noise in a daily scene anticipated for Papua New Guinea is ~0.087 $\times$ 10^6 kg. Hence, if scene-integrated SO_2 burdens are the only source of information, any daily variation in OMI SO_2 lower than this number cannot be considered as significant change in degassing, rather than simply an atmospheric/instrumental artifact.

3. Results

3.1. Major Active Sources of Volcanic SO_2 in Papua New Guinea

[28] We examined monthly mean plots of SO_2 burden over the region throughout the study period and identified degassing volcanoes in each image on the basis of a coherent plume, readily distinguishable from background noise and emanating from a source volcano. Discrete clouds of SO_2 which were not physically connected to a volcano yet which still lay adjacent to one and could not obviously have originated from another source were also regarded as positive evidence for degassing. Ambiguous clouds of SO_2, where significant

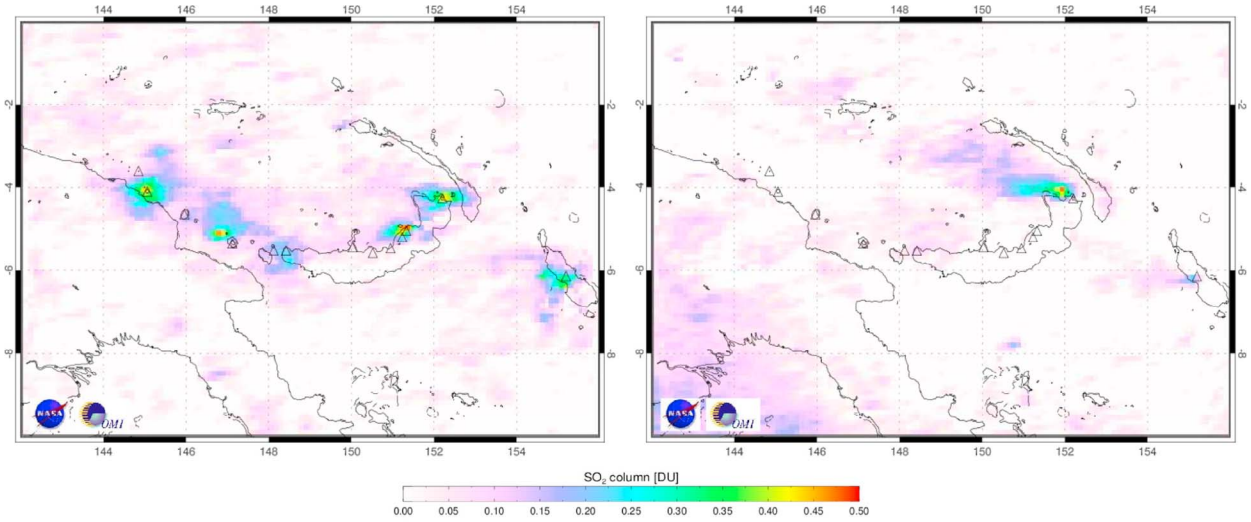

Figure 2. Monthly mean plots of daily SO_2 burden, measured by OMI over Papua New Guinea, during (left) April 2007 and (right) August 2008. In April 2007, SO_2 emission was detected at five volcanoes across the region (from west: Manam, Langila, Ulawun, Rabaul and Bagana). In August 2008, SO_2 emission was only detected at Rabaul and Bagana; the possible plume seen in the vicinity of Langila is not sufficiently coherent to be distinguished from background noise. Color-scale is in Dobson Units (DU), the number of molecules in a square centimeter of atmosphere. If all of the sulphur dioxide in a column of the atmosphere was compressed into a flat layer at standard temperature and pressure, one Dobson Unit would be 0.01 mm thick and would contain 0.0285 g of SO_2 per square meter.

in size or mass, were assigned a source volcano in that month by inspection of daily SO_2 burden plots. Two example monthly mean plots are shown in Figure 2; the complete 2005–2008 sequence is available in the auxiliary material.[1]

[29] OMI detected significant SO_2 emissions from Manam, Langila, Ulawun, Rabaul and Bagana during 2005–2008. These five volcanoes were listed by *Andres and Kasgnoc* [1998] as the major active sources in Papua New Guinea, and were among the targets for the ground-based SO_2 flux measurement campaign of *McGonigle et al.* [2004a] in summer 2003 (see section 1.2.). Activity at Manam, Langila, Ulawun, Rabaul and Bagana was reported regularly throughout 2005–2008 by the Smithsonian Institution's Global Volcanism Program. Activity was also reported at Pago (minor eruption in August 2007 only) and the Garbuna Group (small eruptions in October 2005 and March 2008, with weak vapor release at other times) during the study period [*Venzke et al.*, 2002–2011]. No unambiguous OMI signal for Pago was noted and while emissions from Garbuna were observed by OMI on 17 October 2005 following the small eruption late on 16 October, and possibly on a few other occasions in November 2005, no other significant or persistent emissions were observed throughout 2005–2008.

[30] The presence of a coherent SO_2 plume from any particular volcano in a monthly average scene (Figure 2) was considered evidence of that it was a source of SO_2 throughout or at least for a significant part of the month in question. In Figure 3 we show the frequency and persistence of emission from each volcano month by month on this basis. Volcanic degassing is very persistent across the region in general: in any given month, at least one volcano and generally two or three was found to be emitting detectable levels of SO_2. This reflects OMI's high sensitivity to atmospheric SO_2, as well as confirming the published reports of persistent and significant sources of volcanic SO_2 in Papua New Guinea [*Andres and Kasgnoc*, 1998; *McGonigle et al.*, 2004a]. This persistence of detection by OMI, with detectable levels of SO_2 acting as indicators of continuing volcanic activity, endorses OMI as a robust monitoring tool. Thermal alert systems such as MODVOLC [*Wright et al.*, 2004] do not detect activity with such frequency, at least over Papua New Guinea, while VAAC ash alerts (from either visible satellite images or pilot reports) often occur only in response to short-lived, higher intensity ash venting events, and do not provide any sense of the activity continuing between these.

[1]Auxiliary materials are available in the HTML. doi:10.1029/2011GC003945.

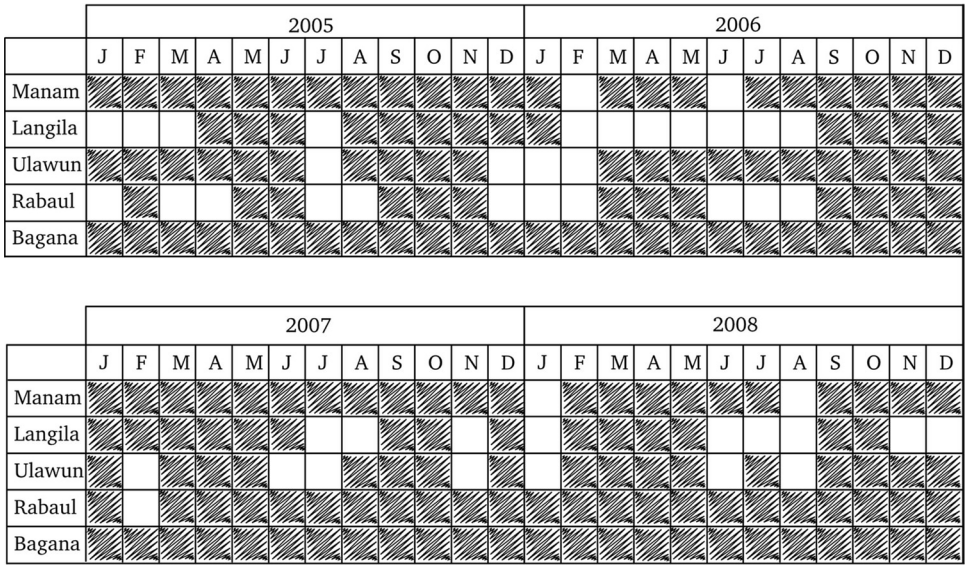

Figure 3. Summary of the frequency and persistence of detection of the five volcanoes in Papua New Guinea where OMI detects coherent SO_2 plumes during 2005–2008.

[31] Bagana is the most frequently detected source volcano, with SO_2 emissions apparent in every month during 2005–2008. The volcano was continuously active during this time, with ongoing effusion of lava from the summit crater and persistent emission of thick white vapor plumes [*Smithsonian Institution*, 2005c, 2008a, 2008b]. Manam's emissions were detected in all but four months, January and June 2006 and February and August 2008. Manam's typical activity is not marked by continuous eruption in the same way as Bagana's; rather it generally exhibits sporadic short-lived cycles of explosive strombolian behavior. The near-continuous release of SO_2 suggested by OMI however, together with the volcano's basaltic composition, suggests the possibility that gas is delivered to the surface via a convecting magma column [*Kazahaya et al.*, 1994]. Ulawun's emissions were detected in 37 of the 48 months analyzed, with emission generally persistent during 2005–2006 and intermittent after January 2007. SO_2 was detected at Rabaul very intermittently until October 2006, when a major explosive eruption took place [*Smithsonian Institution*, 2006b]. Thereafter SO_2 release was almost continuous until the end of 2008. Langila was emitting SO_2 in 28 months, with a generally sporadic pattern – 10 months of continuous emission from September 2006 to June 2007 represents the most persistent period.

[32] Using the automatic source identification capability of OMIPLOT, we identified how many days during 2005–2008 each volcano was the strongest emitter of SO_2 (Figure 4). Manam and Bagana are consistently the strongest sources in the region, together being designated as the maximum daily source on ~700 days during 2005–2008 (the measurement period includes a total of 1461 days). Rabaul is also frequently identified as the maximum source, on over 250 days. Ulawun and Langila are only rarely the strongest SO_2 emitters.

3.2. Linking the OMI SO_2 Data Set With Observed Volcanic Activity

[33] We generated a time series of daily total SO_2 burden measured by OMI over Papua New Guinea during 2005–2008 (Figure 5). The rate of SO_2 degassing in the arc can be estimated using a cumulative plot of SO_2 burden through 2005–2008 (also Figure 5), where the slope is a proxy for SO_2 flux. The total SO_2 burden measured over Papua New Guinea by OMI was 1838×10^6 kg (10^6 kg = 1 kt).

[34] A major release of SO_2 was observed by OMI during a large sub-Plinian explosive eruption of Manam on 27 January 2005. This eruption marked the climax of a period of intense activity that began in October 2004 [*Smithsonian Institution*, 2005a]. The eruption began at 1400 UTC, with the resulting plume penetrating the stratosphere to an altitude of 21–24 km. OMI's first view of the SO_2 cloud was at 0409 UTC on 28 January, when a burden of $\sim 137 \times 10^6$ kg was measured. The main body of the plume was blown rapidly westward during the following days, while a high-altitude component of lower mass persisted over Manam. The mass of

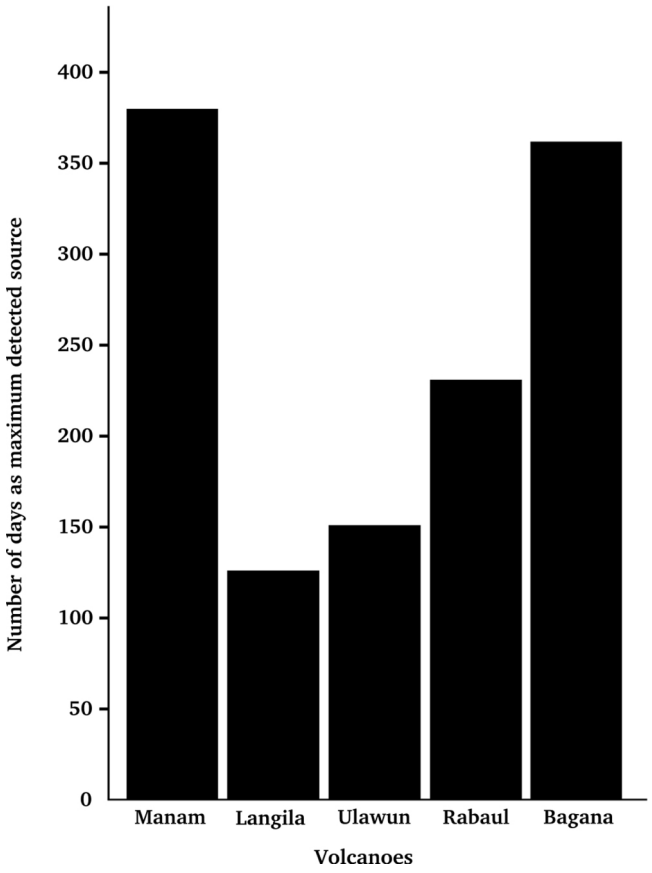

Figure 4. Histogram displaying the number of days where each of the five actively degassing volcanoes was the strongest SO_2 source in the Papua New Guinea region.

SO_2 in the primary plume appeared to decrease rapidly, with OMI measuring $\sim 71 \times 10^6$ kg, $\sim 54 \times 10^6$ kg and $\sim 24 \times 10^6$ kg on 29, 30 and 31 January. This may be an indication of SO_2 scavenging linked to the high ice-content reported in the eruption cloud [*Textor et al.*, 2003; *Tupper et al.*, 2007]. This implies that the total mass of SO_2 released by the eruption may have been greater than the initial mass measured by OMI. A clearer understanding of the rate of chemical processing of the plume SO_2, as well as the extent of dispersal by wind, could enable estimation of the initial mass release but without this constraint, we make no such estimate here. A subsequent large eruption from Manam occurred at 2300 UTC on 28 January [*Smithsonian Institution*, 2005a] with a less significant SO_2 release; OMI measured a burden of $\sim 8 \times 10^6$ kg over the study region (Figure 1) on both 29 and 30 January. Minor degassing was also observed from Bagana and Ulawun in January 2005.

[35] A combination of high emission from Manam in the aftermath of its 2005 eruption and ongoing strong degassing from Bagana (associated with sustained lava effusion) [*Smithsonian Institution*, 2005c] caused a resurgence in arc-scale SO_2 degassing during March 2005, after the total SO_2 burden had fallen through February. The highest daily burden measured at this time was $\sim 4.3 \times 10^6$ kg. Measured burdens decreased through early April before rising in the latter half of the month, with a peak of $\sim 3.4 \times 10^6$ kg recorded; the timing of this increase coincides with the onset of forceful emissions of thick white and gray ash clouds from Langila, representing the onset of a period of more intense activity [*Smithsonian Institution*, 2005b]. This initiation of eruptive activity at Langila is clearly seen by visually inspecting OMI monthly mean scenes. Strong emissions from Manam, Langila and Bagana were observed in May, with a peak daily SO_2 burden of $\sim 6.7 \times 10^6$ kg, before an apparent surge in degassing across the region in June, where for almost two weeks daily SO_2 mass burdens between $\sim 8.7 - 22.0 \times 10^6$ kg were recorded. Ground observations of volcanic activity before June described moderate eruptive activity at Manam and Langila [*Smithsonian Institution*, 2005b], but nothing for Bagana in spite of OMI observations

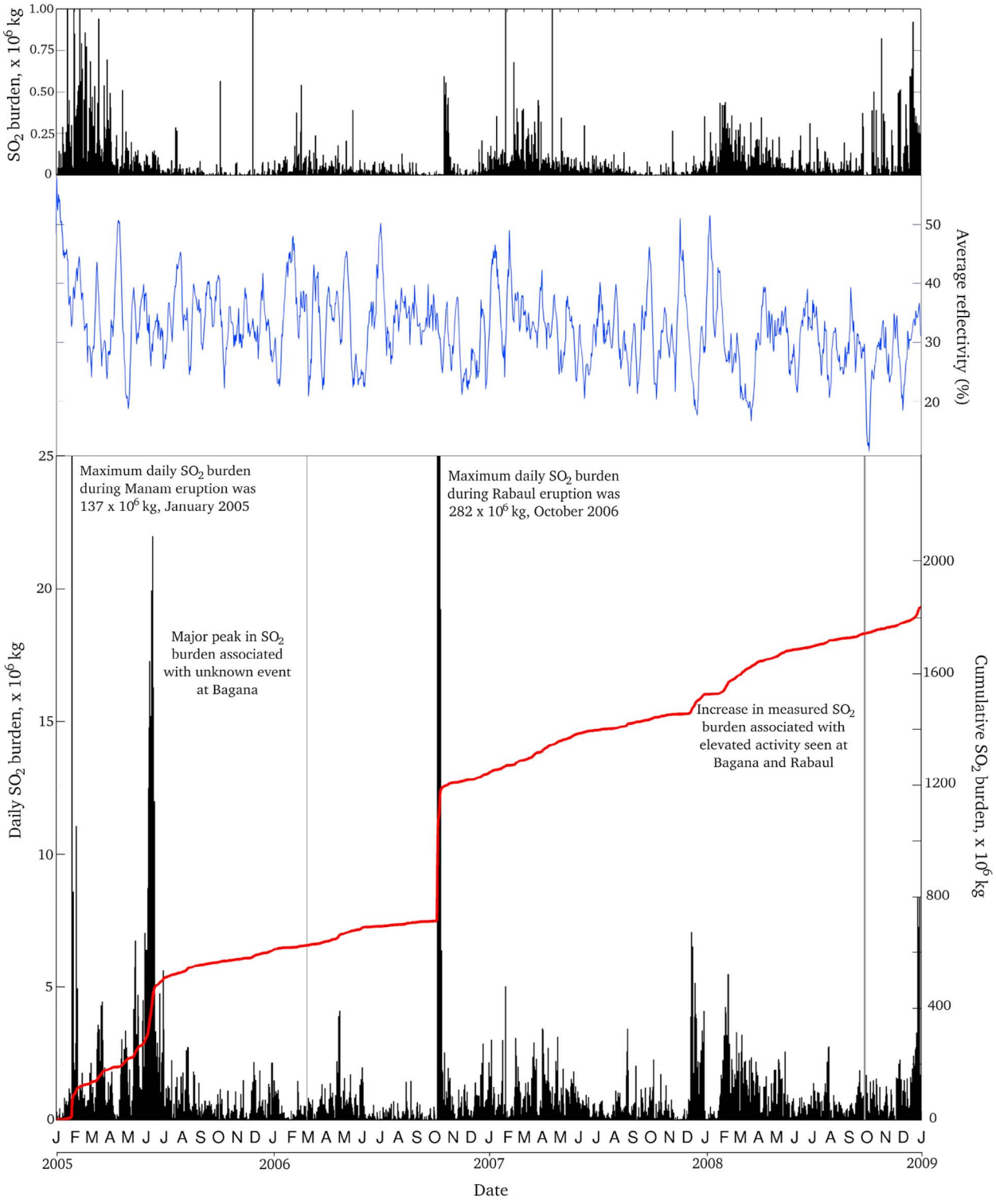

Figure 5. Time series plot of daily total SO_2 burdens measured by OMI over Papua New Guinea during 2005–2008. Also plotted is cumulative time series (red line) of total regional SO_2 and daily average reflectivity (blue curve), which is smoothed with a 7-day moving average. An estimate of background noise in the OMI SO_2 data set is also provided (top panel), calculated from apparent SO_2 mass burden over a nominally SO_2-free area of Pacific Ocean. The vertical gray lines represent gaps in the OMI data of >1 day.

that suggest SO_2 emissions were strongest there (see section 3.3). Overall $\sim 308 \times 10^6$ kg of SO_2 was detected by OMI from 20 April to 30 June 2005.

[36] During July 2005 to October 2006 the average daily burden was $\sim 0.45 \times 10^6$ kg, corresponding to a shallower slope on the cumulative curve than the first half of 2005 (Figure 5). Interpretation of monthly average images (Figure 2) during this interval suggested Bagana remained the major source overall, though not always on a daily timescale. For example, a large explosive event occurred at Manam on 27–28 February 2006 [*Smithsonian Institution*, 2006a]. However, this coincided with a period of OMI downtime due to instrument malfunction, so unfortunately no SO_2 was observed. The eruption was however detected by another space-based instrument, the Atmospheric Infrared Sounder (AIRS) which measured a maximum SO_2 mass of $\sim 27 \times 10^6$ kg (F. Prata, unpublished data available at http://www.bom.gov.au/info/vaac/manam06.shtml, 2006).

[37] On 7 October 2006, a major sub-Plinian eruption occurred at Rabaul, beginning 0845 local time (2245 UTC, 6 October 2006) and continuing until 1730 (0730 UTC); no precursor activity was observed beyond a slight deflation [*Smithsonian Institution*, 2006b]. The eruption resulted in widespread ash deposition over Rabaul and the entire Gazelle Peninsula, as well as the emplacement of lava flows, detected by MODVOLC thermal alerts. The eruption column entered the UTLS, reaching an altitude of ~18 km before dividing into two distinct portions. One was blown rapidly eastward across the Pacific Ocean by the sub-Tropical jet stream, reaching South America on 14 October; the other persisted at higher altitude over Papua New Guinea, finally dispersing northward by 18 October. The first OMI observation of the plume was at 0410 UTC on 7 October when the eruption was still ongoing; the SO_2 mass burden measured was $\sim 226 \times 10^6$ kg. The second OMI observation at 0329 UTC on 8 October reported $\sim 282 \times 10^6$ kg SO_2 while burdens of $\sim 202 \times 10^6$ kg, $\sim 192 \times 10^6$ kg and $\sim 156 \times 10^6$ kg were measured by OMI over the subsequent days (9–12 October). It is difficult to separate the competing contributions of atmospheric SO_2 loss and dispersion and further volcanic SO_2 release from the maximum burdens measured on 7–8 October. We propose from the rapid decrease in total plume mass observed by OMI from 8 October onwards however that some chemical processing of SO_2 occurred between the cessation of eruptive activity and the second OMI observation, a time interval of ~20 h. As with the 27 January 2005 Manam eruption though, we lack the knowledge of the complex local atmospheric processes and so use $\sim 282 \times 10^6$ kg as the best-constrained estimate of total SO_2 mass released by this eruption, acknowledging that it probably represents a minimum value.

[38] The year following the Rabaul eruption (November 2006 – November 2007) showed generally low-level degassing across the region with an average daily burden of $\sim 0.63 \times 10^6$ kg. This is a higher apparent level of emission than was seen during the previous period of sustained passive degassing, from July 2005 to October 2006, which is likely due to the greater persistence of significant degassing from the region's volcanoes (Figure 3). The major consistent emitters between November 2006 and November 2007 were Bagana, where lava effusion accompanied by thick white vapor plumes continued [*Smithsonian Institution*, 2007b, 2008a], and Rabaul, where intermittent, minor explosive activity took place, accompanied by ashfall and occasional roaring, rumbling and incandescence [*Smithsonian Institution*, 2007c, 2008a]. An increase in activity at Langila began in August 2006, with explosions of incandescent lava fragments, regular intervals of roaring and continuous emissions of dark ash plumes [*Smithsonian Institution*, 2007a] and this is reflected in the OMI monthly mean scenes, particularly in October 2006 to January 2007, and April to June 2007. Manam and Ulawun were reasonably active during this time, and their plumes were generally detected by OMI, though the levels of detected SO_2 are low compared to Rabaul, Bagana and Langila.

[39] A steepening of the cumulative SO_2 curve in Figure 5 through December 2007 reflects higher recorded SO_2 burdens, mainly from Rabaul and Bagana (Figure 5). During late 2007, active lava flows were observed on Bagana's southeast flanks [*Smithsonian Institution*, 2008a], which may explain the high levels of SO_2 emission. At Rabaul, no particularly remarkable activity was reported in ground-based reports. Nevertheless, the two volcanoes together led to a total burden of $\sim 70 \times 10^6$ kg during the month.

[40] January 2008 saw very low levels of SO_2 detected by OMI, while February to May 2008 were months of high SO_2 emission, which peaked in mid-February ($\sim 5.5 \times 10^6$ kg on 10 February) and then gradually decreased into mid-April, before rising again for the whole of May (Figure 5). Again, Rabaul and Bagana were the two principal sources, with Bagana dominant initially, before

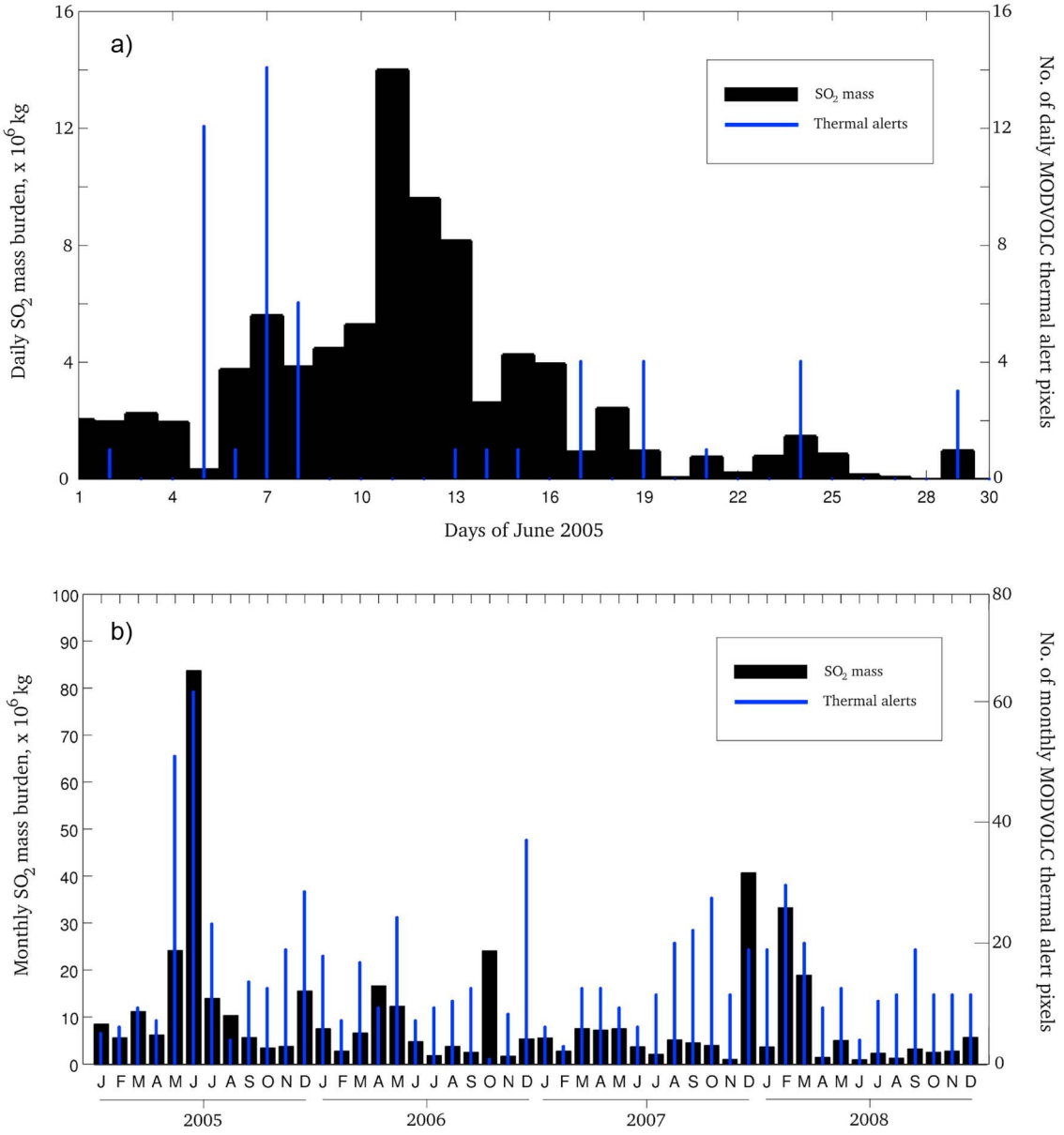

Figure 6. (a) Bagana daily SO_2 during June 2005 (black bars) against MODIS/MODVOLC daily total of thermal anomaly pixels (blue spikes). (b) Monthly total SO_2 burdens measured over Bagana by OMI against monthly total thermal anomaly pixels.

Rabaul's emissions overtook from April onwards. Ground-based reports from May suggested an acceleration in Bagana's lava effusion rate had taken place [*Smithsonian Institution*, 2008b], while those for Rabaul in the middle of 2008 only refer to sporadic ash-rich explosions, set against a background pattern of quiescent degassing [*Smithsonian Institution*, 2008c]. The period from 1 February to 31 May saw a total of $\sim 149 \times 10^6$ kg SO_2 observed over the region and a steeper slope, corresponding to an average daily burden of $\sim 1.25 \times 10^6$ kg, on the cumulative time series (Figure 5).

[41] The remainder of the survey period, until the end of 2008, was largely unremarkable, with OMI SO_2 daily burdens generally $<1 \times 10^6$ kg. The peaks above this value (Figure 5) are almost all associated with Rabaul, which became the dominant SO_2 source during the latter half of 2008. Little change was reported in the observed activity at the volcano, so it is assumed that continuous

degassing was taking place, with periodic violent paroxysmal eruptions resulting in ash-rich and more gas-rich plumes, which also reached higher altitudes [*Smithsonian Institution*, 2008c].

3.3. Case Study on Bagana

[42] We investigated Bagana's SO_2 emissions specifically to evaluate the previous reports that it dominated Papua New Guinea's volcanic SO_2 budget [*Andres and Kasgnoc*, 1998; *McGonigle et al.*, 2004a]. The total SO_2 burden over Bagana during 2005–2008 was $\sim 455 \times 10^6$ kg. Monthly total burdens ranged from $<5 \times 10^6$ kg to $\sim 80 \times 10^6$ kg, with a mean value of $\sim 9.5 \times 10^6$ kg. The peak value of $\sim 84 \times 10^6$ kg was observed in June 2005 and corresponded to a significant SO_2 plume on the monthly OMI image (Figure 2) as well as a major increase in cumulative regional SO_2 burden (Figure 5). However, no reference to elevated activity at Bagana during June 2005 is made in contemporary activity reports; the only reference to any elevated activity near this time is a minor vulcanian eruption on 6 May 2005 in a Rabaul Volcano Observatory report of the time which mentions "ejection of lava fragments [which]… flowed towards Sauva [*sic*] River" (G. Wadge, personal communication, 2011). We examined daily OMI images and generated a time series of daily SO_2 burdens over Bagana during June 2005. The period of elevated emissions occurred from 6 to 16 June and followed a dramatic increase in MODVOLC thermal alerts measured by the MODIS satellite (Figure 6a). The apparent short-duration nature of the thermal anomaly event may be partly due to poor spatial discrimination by the instrument or by meteorological clouds or the volcanic plume obscuring the thermal radiation source. Limited additional satellite data, in the form of ASTER visible-near-infrared (VNIR) imagery, reveals that a pyroclastic flow was emplaced in the Saua River on Bagana's southern flank at some point between 26 June 2003 and 14 March 2006; a fresh lava flow on the southern flank was also observed (G. Wadge, personal communication, 2011). Though the time window is rather wide, we propose that these features may be linked to the elevated degassing. An increase in lava production from the volcano or an edifice collapse, leading to decreased loading, could both result in elevated gas emissions, as could a significant explosive eruption. While such eruptions have occurred at Bagana, they are rare and seem unlikely to have escaped detection on the ground, or by pilots. However, reasonably significant activity at Bagana does seem able to pass unreported in the absence of regular contact with local people [*International Airways Volcano Watch*, 2010]. Further peaks in Bagana's SO_2 output in December 2005, April, May and October 2006, December 2007 and February and March 2008 may reflect similar, if smaller-scale, events. Correlation between monthly SO_2 burdens over Bagana and the number of MODVOLC thermal alerts detected is inconsistent, though peaks in each data set often occur in close time succession (Figure 6b). A lack of thermal alerts can either indicate the absence of a strong heat source or its obstruction by extensive cloud cover over the volcano or the volcanic plume [*Wright et al.*, 2004].

4. Discussion

4.1. Relative Significance of Major Explosive Activity Versus All Other Activity

[43] The previous section demonstrates that OMI is sensitive to changes in degassing at individual volcanoes and across Papua New Guinea, and correlates well with variations in volcanic activity described by contemporary observations. However, with only an incomplete independent record of volcanism available, and pending robust and thorough validation of OMI's measurement of low-level SO_2 emissions into the troposphere, we are hesitant to divide our total SO_2 burden purely between eruptive and non-eruptive degassing components. Therefore we distinguish only between major explosive eruptions and all other forms of activity, with such eruptions defined as those accompanied by major SO_2 release into the UTLS. In the last $\sim$100 years (1919–2011) there have been seven VEI 4 explosive eruptions recorded in Papua New Guinea, at Manam (1919, 2005), Ulawun (2000), Rabaul (1937, 1994, 2006) and Bagana (1952). The SO_2 release associated with these historical eruptions is poorly constrained. However, we can make an estimate of the contribution of major explosive events to Papua New Guinea's long-term SO_2 emission budget, using the two OMI observations of VEI 4 eruptions during 2005–2008. The Manam and Rabaul eruptions together released a total of $\sim 420 \times 10^6$ kg SO_2, though of course the SO_2 masses reported by OMI for these eruptions must be considered as minimum estimates. The total mass release of seven similar-sized eruptions is therefore estimated as $\sim 1.5 \times 10^9$ kg over $\sim$100 years, corresponding to an average annual

contribution of $\sim 15 \times 10^6$ kg. The total SO_2 burden detected by OMI over Papua New Guinea in 2005–2008 was $\sim 1.8 \times 10^9$ kg, corresponding to an annual mean emission of $\sim 350 \times 10^6$ kg after removal of the major explosive contribution. Assuming that this four-year period is representative, averaging over a 100-year period suggests that explosive volcanism contributes only ~4% of total annual SO_2 emissions from Papua New Guinea's volcanoes. This estimate agrees with previous studies, which suggest that the explosive contribution to the global volcanic SO_2 budget is far smaller than that from non-explosive and non-eruptive degassing combined [*Berresheim and Jaeschke*, 1983; *Bluth et al.*, 1993; *Andres and Kasgnoc*, 1998]. *Berresheim and Jaeschke* [1983] concluded that non-eruptive activity is always dominant over eruptive activity as the major source of volcanic SO_2, with eruptive release accounting for ~7% of total annual volcanic SO_2 emission, despite significant uncertainty in the data available for SO_2 released by major eruptions. *Bluth et al.* [1993] used TOMS data to provide improved constraint on the contribution by major explosive eruptions and showed that they accounted for ~31% of total annual volcanic SO_2 emissions. This higher estimate is probably heavily skewed in favor of explosive volcanism due to the low value used for the SO_2 output associated with non-eruptive volcanism, which was compiled from only a 2-year period of ground-based observations and COSPEC measurements and assumed that global degassing rates are largely constant [*Stoiber et al.*, 1987]. The more extensive time-averaged inventory of volcanic SO_2 emissions of *Andres and Kasgnoc* [1998], which combined TOMS estimates of explosive release and COSPEC measurements of non-eruptive degassing, suggested that explosive volcanism contributes <1% to total annual global SO_2 emissions.

4.2. Comparison of OMI SO_2 Burden to Ground-Based Estimates

[44] In order to assess OMI's sensitivity to tropospheric SO_2 released by passive degassing in Papua New Guinea more fully, we compared our four-year OMI SO_2 burden time series to ground-based estimates of degassing. Two studies estimate annual volcanic SO_2 emission from the region by extrapolating from limited ground-based measurements of SO_2 flux [*Andres and Kasgnoc*, 1998; *McGonigle et al.*, 2004a]. Our OMI record includes major explosive eruptions, while the other two estimates are based entirely on SO_2 released by non-eruptive degassing; without the explosive component, our mean OMI-measured annual release is $\sim 350 \times 10^6$ kg. *McGonigle et al.* [2004a] estimate the annual emission to be $\sim 1160 \times 10^6$ kg; *Andres and Kasgnoc* [1998] estimate it to be $\sim 1970 \times 10^6$ kg. Our OMI record therefore suggests an annual non-eruptive SO_2 emission that is ~18–30% of the two estimates published previously.

[45] We also compared OMI estimates of Bagana's degassing with ground-based flux measurements for the volcano [*McGonigle et al.*, 2004a]. The total SO_2 burden measured by OMI over Bagana in 2005–2008 was $\sim 455 \times 10^6$ kg, or ~25% of the total regional emission. Excluding the contribution by the large Manam and Rabaul eruptions, Bagana contributed ~33% of the non-eruptive degassing to the arc. *McGonigle et al.* [2004a] estimated that Bagana's emissions constituted ~61% of the arc budget; this discrepancy may be explained by the elevated activity at Manam and Rabaul relative to Bagana during 2005–2008, particularly in terms of higher SO_2 emission following their respective major explosive eruptions in January 2005 and October 2006. The daily SO_2 release reported for Bagana by *McGonigle et al.* [2004a] was $\sim 2 \times 10^6$ kg, which equates to $\sim 2900 \times 10^6$ kg over 4 years compared to our OMI estimate of $\sim 455 \times 10^6$ kg. Again, OMI estimates a much lower total SO_2 release, seeing ~16% of the total emissions estimated from ground-based measurements. This is comparable to the difference in arc-wide estimates of SO_2 degassing.

[46] Comparison of OMI and ground-based data at other volcanoes suggests this phenomenon may be a universal one. OMI estimated that degassing from Tungurahua during 2004–2006 released $\sim 234 \times 10^6$ kg per year, not counting major explosive eruptions [*Carn et al.*, 2008]. Ground-based DOAS measurements at Tungurahua recorded an average SO_2 flux of $1.458 \pm 2.026 \times 10^6$ kg per day during 1999–2006 [*Arellano et al.*, 2008]. The large uncertainty on this value is due to the alternation of long periods of passive degassing and short periods of intense explosive activity with a much higher SO_2 flux. Estimated annual emissions therefore vary widely, from $\sim 532 \times 10^6$ kg to $\sim 1272 \times 10^6$ kg; the higher intensity of activity at Tungurahua during 2004–2006 implies that the true value is probably toward the upper bound of this range. If this is the case, the OMI total is only ~18% of the DOAS; if the lower bound DOAS value is used, the discrepancy is ~44%.

[47] Comparisons between OMI and ground-based estimates of SO_2 passive degassing have also been

published for Ambrym volcano, in the Vanuatu archipelago situated to the southeast of Papua New Guinea and the Solomon Islands [*Bani et al.*, 2009, 2011]. Ambrym volcano is a prodigious source of volcanic SO_2, with an output comparable to Etna; other active sources in the archipelago are Yasur, Lopevi, Ambae and Vanua Lava. DOAS measurements made on over thirty days between 2004 and 2009 give an estimated mean SO_2 emission rate of 5.4 ($\pm$1.6) $\times$ 10^6 kg d^{-1} from Ambrym, which contributes ~63% of the total arc output, estimated from DOAS as 3.1 ($\pm$0.8) $\times$ 10^9 kg yr^{-1} without contribution from sporadic phases of extreme degassing [*Bani et al.*, 2011]. OMI data covers a far longer period, from 2004 to 2009, and indicates a total burden measured of ~4.3 $\times$ 10^9 kg across the arc. This would correspond to an annual average of ~0.9 $\times$ 10^9 kg, which is considerably lower than the annual total estimated from DOAS measurements. Direct comparison between OMI and DOAS measurements of Ambrym's degassing on particular days also highlighted this discrepancy; OMI SO_2 burdens amounted to only ~23% of the corresponding emission rates measured by DOAS [*Bani et al.*, 2011].

[48] The discrepancy apparent between OMI and DOAS SO_2 estimates suggests that there are limitations to OMI's sensitivity to passive degassing of SO_2, although in many cases these may be correctable e.g., with some knowledge of the SO_2 loss rate. First however, we note that the available ground-based data sets may not be totally robust or representative estimates of a volcano's degassing behavior. Frequently, due to remoteness, inaccessibility or other logistical factors, ground-based estimates of degassing are accomplished following short campaigns rather than long-duration continuous monitoring; the DOAS surveys of Papua New Guinea [*McGonigle et al.*, 2004a] and Vanuatu [*Bani et al.*, 2011] typify this. Such an approach only provides a snapshot of the degassing regime and particularly if the campaign is in response to reports of elevated activity, the observed SO_2 fluxes may not be representative of the long-term typical release. With the recent increase in long-term surveying initiatives such as the Network for Observation of Volcanic and Atmospheric Change (NOVAC [*Galle et al.*, 2010]), this practice may become less prevalent, but for many remote volcanoes, isolated measurements may continue to be all that are available. Furthermore, even long-duration ground-based campaigns may be subject to significant uncertainties in the generated data sets. The uncertain combination of two strongly different degassing regimes at Tungurahua and consequently wide-ranging DOAS estimates of typical SO_2 flux typify this [*Arellano et al.*, 2008]. Errors associated with observational conditions such as variable wind speed and plume ash-content may also be significant in the DOAS retrieval. Again, implementation of newer instruments via projects like NOVAC, use of accurate meteorological data and increased operational expertise will work to reduce these uncertainties.

[49] Despite the uncertainties in ground-based measurements though, it is clear that OMI tends to consistently return lower estimates of total SO_2 emission. This is likely due to a number of factors, many of which were outlined in section 2.4. We suggest that the principal reasons for the low masses reported are the dual effects of dispersion and chemical processing of the volcanic plume. OMI's daily revisit time is unprecedented for an instrument of its sensitivity but we suggest that the 24-h interval between overpasses is easily sufficient for potentially dramatic reductions in plume mass due to dispersal and chemical loss processes. In a humid tropical atmosphere, rapid SO_2 processing is expected [*Oppenheimer et al.*, 1998; *Carn et al.*, 2011] and is likely to have had a greater impact on the relatively mature plumes seen by OMI compared to the fresh plumes observed by DOAS instruments which are typically only a few kilometers downwind of the source.

[50] Local atmospheric and meteorological conditions are expected to limit OMI's sensitivity in other ways too. We suggest that the impact of ash on the OMI SO_2 retrievals has little significance to the study of frequently ash-poor passive degassing plumes. Meteorological clouds however do have the potential to significantly affect SO_2 detection and retrieval, though as yet this is not rigorously quantified. Nevertheless, we make a few remarks here, since Papua New Guinea's tropical location gives the potential for widespread cloud cover, both high level cirrus clouds and lower, thicker cumulus and cumulonimbus, and particularly in the wet season (approximately December to March). The greatest interference is likely to come from thick, optically reflective cumulonimbus and cumulus clouds, and we assess the extent of their coverage using OMI's measurement of effective reflectivity across the observed scene. From the curve plotted in Figure 5, we note that reflectivity fluctuates around ~30% (mean = 32.3 $\pm$ 6.3%), and although there are possibly periods where higher cloud cover seems to coincide with low detected SO_2 (e.g., January 2008), we do not judge this to be sufficient

cloud cover for major interference in OMI's retrieval during the majority of our study. We also note that OMI's good spatial resolution should enable it to see through gaps in broken cloud cover [*Levelt et al.*, 2006]. We acknowledge that a more robust assessment of cloud interference with OMI is necessary; one option would be coincident use of higher spatial resolution MODIS data to map cloud locations within an OMI pixel, but such a study falls beyond the scope of this paper.

[51] Last, we anticipate potentially significant error inherent to the measurement of passive degassing specifically. Plumes from low-elevation vents released passively may become entrained in the well-mixed PBL rather than the free troposphere if they rise to less than ~2 km. The OMI PBL SO_2 retrieval is limited to cases of optimal viewing conditions (clear skies, near-nadir viewing angles, high solar zenith angle) or particularly strong SO_2 sources, such as Ambrym volcano, Vanuatu, which nevertheless exhibits the same apparent OMI underestimate as weaker SO_2 sources. In Papua New Guinea, the 688 m-high vent of Rabaul may mean that much of its passively released SO_2 is emplaced in the boundary layer and so is not fully incorporated into the overall OMI SO_2 budget.

[52] Future work must tackle the uncertainties outlined above in order to constrain the limitations of the OMI data set better. Of major importance is to understand the nature of processes impacting tropospheric SO_2. Direct measurement approaches to constrain loss rates of volcanic SO_2 have resulted in a range of estimates covering four orders of magnitude, suggesting that isolated snapshots can never be totally representative of rapidly or nonlinearly varying atmospheric conditions. However, detailed atmospheric chemistry/transport modeling of volcanic emissions [e.g., *Langmann et al.*, 2009] has been shown to be capable of refining our understanding of volcanic plume behavior, and may present a suitably synoptic approach to simulate the complex atmospheric processes. Such models also offer great potential for sensitivity studies of the impact of variations in altitude and amount of SO_2 on OMI SO_2 retrievals.

5. Conclusion

[53] This study provides the first synoptic, long-term analysis of the volcanic degassing regime in Papua New Guinea. The area is remote and several of the volcanoes inaccessible; satellite-based remote sensing by OMI is therefore an essential tool in terms of long-term monitoring endeavors. Consequently this study not only provides a new and better-informed understanding of Papua New Guinea's volcanoes, it also serves as a valuable appraisal of OMI's capabilities in the detection and measurement of volcanic SO_2 emissions, particularly via passive degassing.

[54] We have produced a new four-year (2005–2008) time series of SO_2 mass burdens in the atmosphere over Papua New Guinea. SO_2 emissions were detected by OMI from each of the region's persistently active volcanoes, namely Manam, Langila, Ulawun, Rabaul and Bagana. No significant SO_2 release was observed from the intermittently active Pago or Garbuna Group volcanoes. The total SO_2 burden for 2005–2008 as measured by OMI was $\sim 1.8 \times 10^9$ kg with Bagana, Manam and Rabaul the most significant sources by persistence and volume of emissions. Major explosive eruptions at Manam (27 January 2005) and Rabaul (7 October 2006) releasing $\sim 137 \times 10^6$ kg and $\sim 282 \times 10^6$ kg SO_2 respectively were observed by OMI. Patterns in the OMI SO_2 record were noted and shown to correlate well with independent contemporary observations and reports of changing volcanic activity in the region. OMI also identified a previously unknown but apparently significant SO_2 emission event at Bagana in June 2005. We made an assessment of the relative significance of large yet infrequent SO_2 releases versus the release from continuous passive degassing to the overall regional SO_2 budget and showed that on timescales of ~100 years, major explosive volcanism contributes <5% of the total emitted SO_2. This agrees well with several previously published studies, though we note that due to OMI's apparent underestimate of the total SO_2 mass released by passive degassing, the contribution by major explosive eruptions is likely to be even less significant over long timescales.

[55] Our understanding of OMI's application to volcanic gas remote sensing has also greatly developed. The identification of and distinction between five active sources in close proximity, as well as the coherent visualization of their plumes is testament to OMI's excellent spatial resolution. The persistence of observed degassing illustrates the remarkable robustness of detection by OMI, particularly compared to other metrics of activity used in monitoring such as MODVOLC thermal anomalies and VAAC ash alerts. The major eruptions which occurred during the observation period resulted in large SO_2 clouds which were detected, measured and tracked by OMI, further demonstrating the instrument's potential for aviation hazard mitigation. The

major advances came in the study of passive degassing however. This study joins a growing body of work that demonstrates that OMI is sensitive to passive volcanic SO_2 release into the lower troposphere and can observe patterns in emission that correlate well with observed activity. This suggests that OMI may be used with some confidence to monitor volcanic regions where ground-based gas measurement is sparse or non-existent. However, uncertainty does remain. In the absence of robust validation of the OMI SO_2 data set the accuracy of OMI's measurement of passive degassing remains unclear. Comparison between OMI and ground-based estimates of total SO_2 release in Papua New Guinea, Ecuador and Vanuatu all suggest that OMI measures ~20–30% of the total emissions. While we stress that comparing OMI and DOAS is inherently problematic (due to different measurement techniques and scales) and also that DOAS data sets may not be entirely robust (large wind field uncertainties, isolated measurements not representative of typical degassing), there are several uncertainties in the OMI measurement. The impact of simultaneous in-plume chemical reaction and wind dispersal on the volcanic cloud, the interference of meteorological cloud and the uncertainty which remains over SO_2 emissions into the PBL all require detailed further study in order to refine OMI estimates of the full range of volcanic degassing exhibited in arc-scale settings around the world.

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