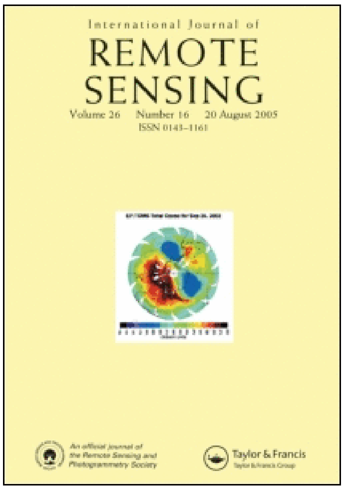

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Exceptional sulfur degassing from Nyamuragira volcano, 1979–2005

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The sulfur dioxide (SO_2) output from Nyamuragira volcano has been monitored by the Total Ozone Mapping Spectrometer (TOMS) since 1979, and is evaluated here to quantify the emissions from this highly productive volcano. The majority of Nyamuragira's emissions were emplaced in the lower to middle troposphere, with SO_2 removal rates of 30–90% per day (k=4.13×10^{-6} to $2.66\times10^{-5}\,s^{-1}$). We have tested a new method of back-calculating persistent, effusive emission fluxes from once-daily observations, which accounts for this rapid daily removal of SO_2 that cannot be measured using satellite 'snapshots'. Twelve of the 14 eruptions during this period each produced $\geqslant$0.8 teragrams (Tg) of SO_2. Nyamuragira erupted nearly 25 Tg of SO_2 during these eruptions, and probably emitted significantly more than we could measure by TOMS. Nyamuragira may be the largest volcanic source of sulfur to the atmosphere for the past few decades.

1. Introduction

The motivation for this study is an ongoing effort to constrain the volcanogenic contribution to the global sulfur (S) cycle. This has been elusive because of both the difficulty in global monitoring and the sporadic nature of volcanic activity (e.g. Bluth *et al.* 1993, Graf *et al.* 1997, Andres and Kasgnoc 1998). Andres and Kasgnoc (1998) estimated that volcanic activity in the period 1970–97 produced approximately 13 teragrams (Tg) of sulfur dioxide (SO_2) annually; man-made contributions during this period (1990) have been estimated at 75 Tg S/year (150 Tg SO_2/year equivalent; Lefohn *et al.* 1999). However, discoveries of 'new' volcanic emissions through new, more sensitive technology or methodologies, more intensive examination of known eruptions, as well as discoveries of previously unknown or unquantified activity, have underscored the need for continuing study of large-scale volcanic gas fluxes.

Nyamuragira (1.41 °S, 29.20 °E) is a massive basaltic shield volcano within the western branch of the East African Rift Valley in DR Congo, 14 km northwest of Nyiragongo volcano. The city of Goma (population ~500 000) lies only 18 km south of Nyiragongo volcano, in a region subject to constant threat of lava flows and gas emissions. Nyamuragira's summit rises approximately 3 km above sea level, with a 2 km × 2.3 km caldera. Historical lava flows extend down the volcano's flanks more than 30 km from the summit, and ashfalls have been reported with occasionally significant accumulation in nearby villages. Its activity typically involves either individually, or combinations of, lava and gas emissions and lesser ash emissions from its summit (central vent), radial and flank vents, and in the last century a lava lake. Associated seismic activity includes swarms of long-period events, signalling

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the movement of magma and impending eruptive activity. Between main eruptions, weak degassing occurs from the main fissures and fumaroles, although gas emissions are not monitored. From 1865 to 2005, there were 40 confirmed eruptive periods of Nyamuragira (Smithsonian GVN 2006), with an average repose time of only 3.5 years. The period of activity 1979–2005, with eruptions roughly every 2 years, represents its highest known sustained period of activity.

Nyamuragira is arguably the most active volcano in the world, yet relatively unstudied due to persistent political strife. Therefore, its contribution to the global terrestrial sulfur flux is considerable but largely undetermined. Satellite-based methods for monitoring global-scale volcanic activity, particularly the main contributors, are needed to add to the growing knowledge of global-scale volcanogenic gas fluxes.

Eruptions from Nyamuragira between 1979 and 2005 were observed and measured by the satellite-based Total Ozone Mapping Spectrometer (TOMS). TOMS sensors have produced an invaluable, near-continuous record of volcanic SO_2 emission since 1978, and TOMS-based studies have been a mainstay of satellite volcano gas monitoring, particularly for large, explosive eruptions (Carn *et al.* 2003). Data from the last TOMS sensor, Earth Probe, are currently only available up to the end of 2005; the sensor may have collected data until May 2007 but contact with the spacecraft was lost in 2006 and no date were transmitted thereafter. The Ozone Monitoring Instrument (OMI) records began in September 2004 with much finer spatial and spectral resolution (Krotkov *et al.* 2006). OMI's greater sensitivity to SO_2 and daily global contiguous coverage will allow routine studies of effusive activity that were possible only in a few cases with the TOMS sensors.

Nyamuragira's effusive activity over the past 25 years represents a significant problem for analysis with the TOMS sensors, which collect data over the region in a single daily overpass. Most significant explosive events produce a discrete cloud that can be tracked over the course of days, and an emitted mass can be calculated by analysing the daily mass total of the decaying cloud (e.g. Krueger *et al.* 1995, 2000, Guo *et al.* 2004). By contrast, Nyamuragira typically erupts for days to weeks at a time, producing a near-continuous emission of SO_2. A single daily image is capable of capturing the emitted plume; however, a difficulty arises from trying to determine how much of the SO_2 has been newly produced each day.

After emission, SO_2 is converted to sulfate aerosol at an altitude-dependent rate, and also scavenged from the plume by wet and dry deposition. It is not possible to determine directly from TOMS data which SO_2 in the image is 'new' ($<$24 h) and which is 'old' ($>$24 h). Analyses of single images of a continuously fed plume can systematically underestimate the total erupted mass because every image will contain an age range of SO_2 from fresh up to 24 h or older. Oppenheimer *et al.* (1998) compiled measured SO_2 decay rates for tropospheric plumes that range from 25% daily mass removal to 100% loss in less than 1 day. Thus, to accurately evaluate SO_2 release by continuously emitting volcanoes, it is necessary to account for SO_2 loss in their eruption plumes.

Krueger *et al.* (1996) evaluated the 1981 Nyamuragira eruption using TOMS data, and used several methods to account for the continuous emissions: (1) assuming a threshold level of SO_2, and assuming all SO_2 'attached' to the volcano and above that level was produced from within the previous 24 h; (2) visually estimating daily emissions, primarily based upon cloud geometry; and (3) measuring only the total daily amount in the TOMS images, calculating a set of decay curves, and fitting the TOMS mass retrievals to estimate a daily 'new' SO_2 production. These results suggested decay rates of about 10–40% per day [k=(1.22−5.91)$\times 10^{-6}\,s^{-1}$], for clouds emplaced into the middle to upper troposphere.

Carn and Bluth (2003) used a different approach to analyse the persistent Nyamuragira emissions, taking cross-sectional analyses of the synoptic TOMS images, and using modelled wind profiles to calculate emission fluxes during the 1998 and 2001 eruptions. This produced plume profiles similar to those used in ground-based spectroscopic methods (e.g. correlation spectrometry (COSPEC) and differential optical absorption spectroscopy (DOAS)), with the assumption that the plume height and emission rate remained constant throughout the 24-h period between TOMS emissions. Their results indicated that calculated SO_2 fluxes could significantly exceed daily TOMS 'snapshot' SO_2 loads for continuously fed plumes. Decay rates calculated using plumes emitted in 1998 and 2001 ranged from 25% to 60% loss per day ($k=3\times 10^{-6}$ to $1\times 10^{-5}\,s^{-1}$, respectively; Carn and Bluth 2003).

For this work we build upon the Carn and Bluth (2003) approach, using independent wind-trajectory model results to delineate the rate of cloud movement and make regular cross-sectional 'slices' through the plume to determine the change in mass as the plume drifts away from the volcano during the course of the day. Here we continuously sample the complete cloud to produce a record of emission characteristics over the 24-h period between TOMS overpasses. We summarize SO_2 cloud and meteorological characteristics of the 14 Nyamuragira eruptions between 1980 and 2004, and attempt to evaluate the total SO_2 release by this extremely productive system.

2. Analysis methods

This work combines the standard method of TOMS SO_2 analyses for emitted volcanic clouds (detailed in Krueger *et al.* 1995, 2000) and an important modification to retrieve SO_2 masses produced by continuously emitting volcanoes.

Data from four different TOMS sensors (table 1) were used in this study; although each sensor is slightly different (e.g. different spatial resolutions), the procedures for deriving SO_2 masses from the data are essentially identical. The main concern is with changes in spatial resolution because the TOMS sensors produce daily maps of SO_2; the Nimbus-7 and Meteor-3 TOMS cover the sunlit earth with no major gaps in coverage. However, the Earth Probe TOMS traded increased resolution for decreased surface coverage at low latitudes, and therefore the SO_2 clouds may not be observed completely by this sensor if they intersect a gap between instrument orbits. ADEOS included additional scans so that full coverage was attained, even at

Table 1. TOMS sensors used in this study.

Sensor	Dates of operation	Nadir resolution	Reference
Nimbus-7	1 November 1978 to 6 May 1993	50 km	McPeters *et al.* (1993)
Meteor-3	22 August 1991 to 24 December 1994	62 km	Herman *et al.* (1996)
ADEOS	11 September 1996 to 30 June 1997	42 km	Krueger *et al.* (1998)
Earth Probe	17 July 1996 to present	24 km until September 1997 39 km to present	McPeters *et al.* (1998)

the higher resolution. Nimbus-7, ADEOS and Earth Probe TOMS were aboard sun-synchronous platforms, producing images at local solar noon each day; however, the Meteor-3 orbit was precessing and therefore the overpass time varied slightly (typically by <1 h) each day. We use the most recent versions of the TOMS pre-processed data: version 8 for Nimbus 7 and Earth Probe, and version 7 for Meteor-3 and ADEOS (e.g. McPeters *et al.* 1998). In this paper, we use the general-case uncertainty for TOMS retrievals of 30%, as suggested by Krueger *et al.* (1995). This is certainly not a robust error analysis; however, no systematic study of TOMS errors has been attempted, largely because the meteorological and volcanological conditions surrounding each eruption are unique and often poorly constrained.

The standard TOMS SO_2 retrieval produces a mass value for each pixel within a scan line (i.e. 35 steps, or pixels, per scan). We then use a resampling scheme to interpolate the TOMS data to a regular latitude–longitude grid. This allows SO_2 mass to be correlated to distance from the volcano, and therefore for a continuously erupting volcano, the daily TOMS snapshot becomes a proxy of the emission history during the previous 24 h. Assuming exponential decay of SO_2, we back-calculate the emission rate(s) throughout the 24-h period that would be necessary to produce the observed pattern of mass distribution. We make several additional assumptions for this method:

- For each day on which at least a portion of a TOMS-observed SO_2 cloud is located physically over the volcano's geometric coordinates, the volcano emitted SO_2 continuously throughout the previous 24-h period, with the following exception: for all non-consecutive days of emission, such as the first and last days of an eruption, we rely on wind trajectory modelling to determine the length of emission. In other words, based upon on how far the cloud has drifted from the volcano, we calculate how long the emission has been occurring.
- The emission rate can vary within each 24-h period, and from one day to the next; however, the rate of SO_2 removal is exponential. The removal processes of SO_2 from TOMS-based observation include chemical conversion, physical dispersion below detection, and wet and/or dry deposition. Deposition is not necessarily exponential, but could be faster. This, and obscuration of SO_2 by meteorological clouds, could cause underestimation of the actual SO_2 flux.
- The cloud altitude and drift speed and direction (i.e. wind fields) can vary from one day to the next, but are assumed constant throughout each 24-h period.

The general procedure for deriving a daily SO_2 flux from TOMS data is presented graphically in figure 1. The first step is to complete a standard TOMS SO_2 analysis, including creating an image using the iterative SO_2 retrieval (figure 1(*a*)) and a tonnage analysis. This involves defining the region containing the SO_2 cloud and calculating a total mass for that region, determining SO_2 noise levels, and then subtracting a noise-equivalent SO_2 amount from the total to determine the net SO_2 mass. Figure 1(*a*) shows a simple case for a cloud over Nyamuragira, and drifting directly west, containing approximately 0.1 Tg SO_2 using the 'snapshot' analysis.

The next step is to perform a trajectory analysis, visually matching an online, modelled trajectory (HYbrid Single-Particle Lagrangian Integrated Trajectory, HYSPLIT; Draxler and Rolph 2003) with the TOMS-derived SO_2 cloud location, to estimate the 24-h trajectory of the cloud (figure 1(*b*)). Using the Global Reanalysis

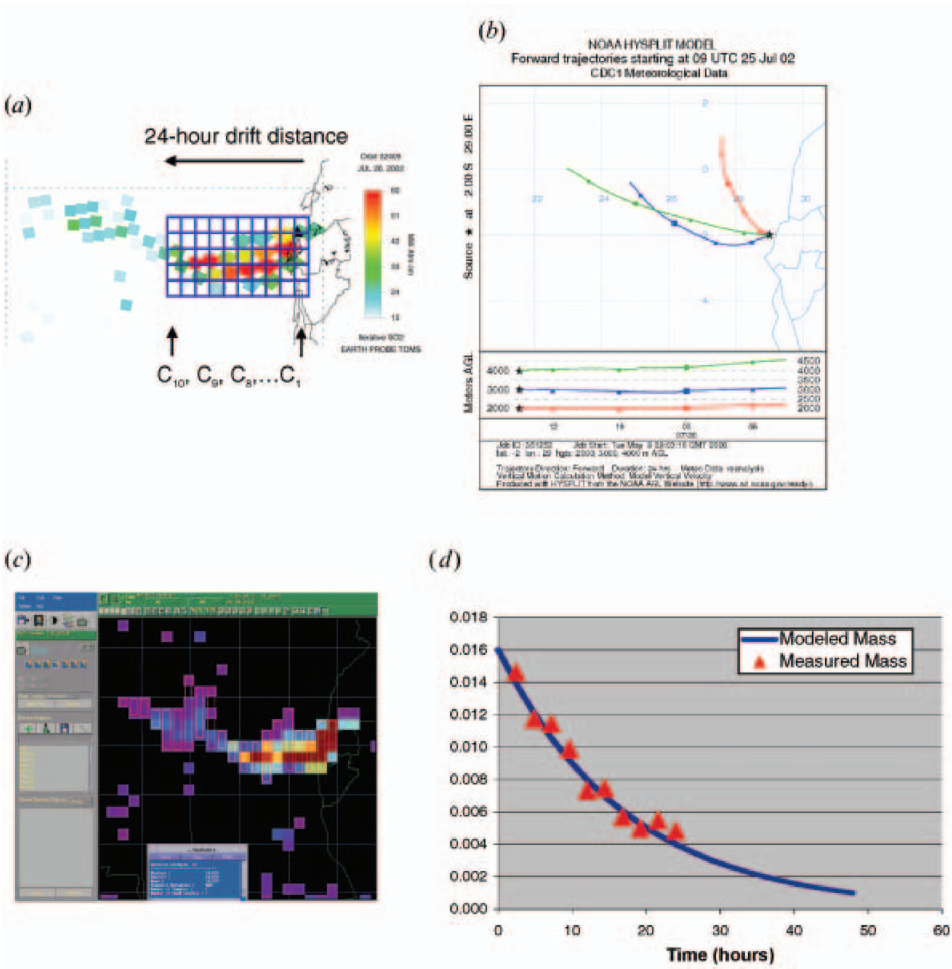

Figure 1. Calculation of daily SO_2 fluxes. (*a*) The original, false-colour TOMS data are displayed with true footprint geometries. (*b*) The HYSPLIT wind trajectory model is used to determine the distance the cloud drifted during the previous 24 h. (*c*) The TOMS SO_2 (in milli atm cm, or Dobson Units, a measure of the gas concentration in terms of the pure gas thickness at STP) data are resampled into a 50 km square grid aligned along latitude–longitude, and the total mass in each column ($C_1 \ldots C_{10}$) is calculated (or row, if for example the cloud drifted to the north or south); (*d*) using the trajectory model to relate distance from the volcano to time since emission, the mass data are fit to exponential decay curves, to back-calculate the emission rate that would have produced the observed pattern of SO_2 mass with distance from the volcano. In this simplified example, the data correspond to a single emission rate of 0.016 Tg/2.4 h; thus over a 24 h period, the daily flux is estimated to be 0.160 Tg/day. The drift distance is covered by 10 columns (*a*); the total mass within each column therefore corresponds to 2.4 h emission time; the distance of the mass from the volcano corresponds to the time elapsed since emission. See text for details.

archived meteorology dataset, the best match in this example was the 3000 m above ground level trajectory, which allows us to constrain the distance the SO_2 may have travelled from the volcano during the past 24 h to roughly 500 km (figure 1(*a*)). Note that a portion of the contiguous cloud (furthest west) is not included, as based on the trajectory analysis this portion is now defined as having been emitted more than 24 h earlier. The trajectory model results are initially defined above ground level; however, because of the coarse resolution of the embedded terrain model, the volcano itself is only estimated at 1.5 km altitude. All trajectory height results reported here are given with respect to mean sea level (see Appendix), thus producing (in three cases) the obviously faulty result where the modelled cloud height is less than the true volcano height. However, this discrepancy has little effect on the overall analysis because the total surface area around the volcano that is represented inaccurately is small compared to the daily drift distance of the SO_2 clouds.

The TOMS SO_2 data are then resampled to a 50 km grid, oriented along latitude–longitude, using a nearest-neighbour interpolation scheme in Terascan^{TM} image processing software (figure 1(*c*)). The resampling procedure is only used if the cloud area and instantaneous mass of the resampled cloud are within 10% of the original, to ensure that the resampling process does not significantly modify the original data. The grid is shown overlain on the TOMS data in figure 1(*a*). Because the SO_2 clouds may drift in any direction (or change direction in response to shifting wind directions), a grid aligned in a north–south or east–west direction will not always allow perpendicular traverses of the cloud for all drift directions (e.g. for clouds that are drifting northwest from the volcano). As a worst case scenario, the maximum offset of 45° could produce an overestimate of cloud width of up to 30% (because traverse methods use the sine of the observation angle to calculate projected plume width; $\sin(45)=0.71$), similar in magnitude to the general-case TOMS uncertainty. However, although maintaining a perpendicular cross-section sampling of the drifting cloud would be ideal, this could also introduce problems of over- and undersampling portions of the cloud. In our method, the full cloud is sampled completely, and only once.

The resampled TOMS data are used to model the previous day's emission history of the volcano, in this case by calculating the total mass in each evenly spaced column and plotting versus distance from the volcano (figure 1(*d*)). In this example, the cloud travelled 500 km in 24 h (based upon modelled wind trajectories), which is divided into 10 columns, each representing 2.4 h of emission. Each of the 10 mass values are then fit to a set of exponential decay curves that represent the expected relationships of mass with distance from the volcano (i.e. time) for a given emission rate. In the simple case depicted in figure 1, only a single decay curve is used for clarity; typically three or more curves are used to best match the data. The mass of SO_2 measured in each of the 10 cross-sectional areas of the cloud is thereby matched to a characteristic emission rate (mass emitted per 2.4 h). To calculate the 24-h SO_2 emission (that produced the original snapshot satellite image of the cloud), the emitted masses produced during each 2.4-h time segment are summed. In our example, the SO_2 emission rate is constant over the 24 h, at 0.016 Tg/2.4 h, and therefore the 24-h flux is calculated as 0.160 Tg/day ($=10\times0.016$ Tg).

Recall that the standard TOMS analysis of the same data produced a tonnage of only 0.100 Tg. Assuming that the general-case 30% retrieval error affects both methods equally, this 60% difference presumably represents the loss due to chemical conversion to sulfate, scavenging, and physical dispersion of SO_2 during a 24-h emission period, which cannot be recognized in a standard TOMS analysis. Although each event will vary in terms of environmental and emission characteristics, we believe that the underestimation of SO_2 emission by a snapshot analysis can be significant, and the calculation of daily fluxes more closely replicates the true emission characteristics of continuously emitting volcanoes.

3. Results

TOMS observed each of the 14 eruptions of Nyamuragira between 1980 and 2004. For each event, we searched the TOMS database for the duration of reported activity in the online database maintained by the Smithsonian's Global Volcanism Program (www.volcano.si.edu). In addition to the Smithsonian database, TOMS data were examined until no more SO_2 emissions could be detected, which sometimes extended the activity beyond the reported duration. Table 2 and figure 2 summarize the TOMS-observed activity for these events, which include 225 individual days (daily data are included in the Appendix). The total SO_2 mass released is estimated as nearly 25 Tg, roughly 50% higher than that calculated by simply totalling the cloud masses from once-daily 'snapshot' images. The majority of the clouds were estimated from wind trajectory modelling to have been emplaced in the lower and middle troposphere, between roughly 4–12 km above sea level. The average calculated removal rates for these clouds ranged from 60% to 85% per day [$k=(1.06-1.86)\times10^{-5}\,s^{-1}$], although individual daily removal rates ranged from 30% to 90% per day ($k=4.13\times10^{-6}$ to $2.66\times10^{-5}\,s^{-1}$).

Despite the large increase in estimated SO_2 release from Nyamuragira using this new method of evaluation, it is likely that this study still underestimates the emitted SO_2. For example, our flux estimation method can only be used when the observed plume is observed drifting away from the volcano; in cases where the cloud was centred over the volcano it was not possible to assign a wind direction, or make cross-sectional slices through the cloud. Other examples of poor viewing conditions include non-contiguous plumes, or when the cloud could not be observed over the volcano due to gaps in coverage. Overall, emission fluxes were calculated for 185 days (82%) of the total TOMS-observed clouds. For the 40 days when the flux method was not possible, the standard snapshot method (Krueger *et al.* 1995) of evaluating the mass from the observed cloud was used.

For clouds that could be matched directly to a wind trajectory, a 24-h drift constraint was determined directly from the model results. A wind trajectory 'match' was defined here as $\pm10°$ of the model direction, and this occurred for 73% of the possible cases (135 days). For days where a wind trajectory could not be matched to the cloud, an estimate of the 24-h emission boundary had to be determined visually, using the geometry of the cloud 'attached' to the volcano, and other trajectory matches as guides to remain consistent with the observed plume behaviour as much as possible.

A different type of evaluation of SO_2 emissions was undertaken to determine the daily 'instantaneous' emissions. For this we assume that TOMS pixels including or immediately adjacent to the volcano contain the most recently emitted SO_2 and are a relatively objective measure of the emission rate at the time of the daily TOMS overpass. The daily SO_2 flux and instantaneous emission results over time for each eruption, except for the 2-day 1987–88 event, are presented in figure 3. The most striking result in the overall pattern of emissions shows the skewness in peak SO_2 production. The majority of SO_2 emission occurs during the first few days in eight out of the 14 events (1980, 1984, 1986, 1994, 1998, 2000, 2001 and 2004). During most of the eruptions, more than 50% of the total emitted SO_2 was produced in 3 days or less. A notable exception was the 1991 eruption, in which there was not a single day that produced more than 10% of the total. The changes in magnitude of the instantaneous emissions typically track with the daily mass estimates, which suggests that our method of deriving daily fluxes is accurately reflecting at least the

Table 2. Summary of Nyamuragira SO_2 eruption characteristics.

Eruption year	TOMS analysis dates	Total days observed	TOMS sensor used	Average cloud height (km)	Average removal rate (%/day)	Total snapshot SO_2 mass (Tg)	Total flux SO_2 mass (Tg)
1980	1–13 Feb	13	Nimbus-7	5.4	66	0.58	0.88
1981	26 Feb 1981–11 Jan 1982	16	Nimbus-7	9.4	62	3.64	4.11
1984	24 Feb–4 Mar	10	Nimbus-7	3.7	64	0.75	1.00
1986	17 Jul–3 Aug	14	Nimbus-7	4.5	68	0.66	0.98
1987	31 Dec 1987–1 Jan 1988	2	Nimbus-7	2.0	85	0.02	0.03
1989	24 Apr–18 May	24	Nimbus-7	4.5	69	1.37	2.10
1991	22 Sep 1991–27 Jun 1992	32	Nimbus-7	2.8	71	0.41	0.85
1994	5–16 Jul	11	Meteor-3	3.8	74	1.12	1.47
1996	1–16 Dec	14	ADEOS	3.8	72	0.49	0.86
1998	17–31 Oct	15	Earth Probe	4.0	72	1.42	2.54
2000	28 Jan–11 Feb	12	Earth Probe	4.7	74	0.14	0.31
2001	6 Feb–7 Mar	22	Earth Probe	4.0	70	0.95	1.73
2002	25 Jul–2 Aug	9	Earth Probe	4.8	81	1.34	2.32
2004	8 May–13 Jun	31	Earth Probe	3.3	75	1.62	2.60
Totals		225		4.3	72	16.21	24.51

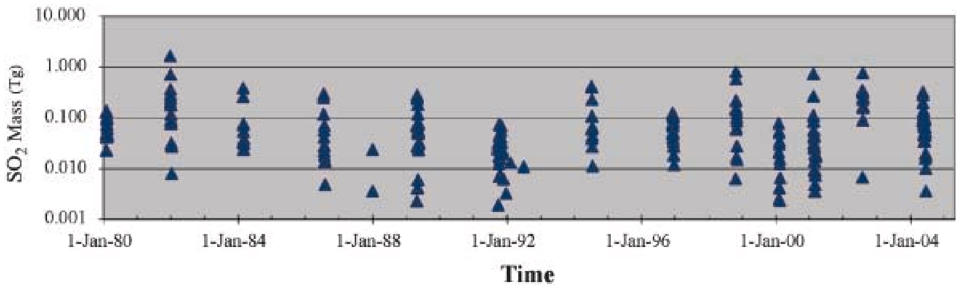

Figure 2. TOMS daily SO_2 flux measurements of Nyamuragira activity, 1980–2004. The data include 225 individual days of observations, for 14 separate eruptions. The eruptions have occurred with remarkable frequency, and productivity, with numerous daily emissions exceeding 0.1 Tg of SO_2.

major trends in SO_2 emissions. An apparent exception may be the July 2002 eruption, but unfortunately the end of this event was not observed because TOMS data were not available for 3–9 August.

The length of TOMS-observed activity ranges from as little as 2 days in the 1987–88 eruption, up to more than 9 months of intermittent activity for the 1991 eruption. In some cases, especially during the country's civil war years (1994–2000), when ground-based monitoring was rare, the TOMS SO_2 data gave the best available estimates of the durations of each eruption. Most of the events produced 10–20 days of observable activity (minimum of 2 and a maximum of 32 days). Daily emissions could be fairly large; peak emissions greater than 0.1 Tg occurred in 11 out of the 14 events; and the highest 24-h emission, 1.7 Tg, occurred during the 1981 eruption as part of a total release of over 4 Tg SO_2.

The SO_2 release by Nyamuragira totals 25 Tg from 1980 to 2004. Overall, Nyamuragira's eruptive cycles consistently released high SO_2 amounts, averaging almost 2 Tg per event: only two events (1987, 2000) released less than 0.8 Tg of SO_2.

4. Discussion

The 25 Tg of SO_2 emitted by Nyamuragira during its 14 eruptive periods makes a significant impact on evaluations of global volcanic sulfur production (table 3). The most comprehensive evaluation of global volcanic emissions has been generated by Andres and Kasgnoc (1998), who estimated SO_2 emissions of 13.4 Tg per year during the period 1970–97 from both explosive and non-explosive volcanism (which includes earlier, lower estimates of Nyamuragira emissions). Our current work suggests that Nyamuragira alone produces at least 5–10% of the total annual volcanic SO_2 flux. The volcano has produced more than 0.8 Tg of SO_2 a total of 12 times between 1980 and 2004 (table 2). During the same time period, only 10 other eruptions in the world have produced this same level (TOMS online research website, http://toms.umbc.edu/); the greatest emission from an individual event was Mount Pinatubo in 1991, with an explosive release of approximately 18 Tg of SO_2 (Guo *et al.* 2004). Taken on an average annual basis, Nyamuragira's output is of the same order as the passive emissions of Mount Etna. Few volcanoes in the world are monitored as intensely as Etna, whose prolific emissions during a relatively active period between 1987 and 1995 averaged 2 Tg per year (Bruno *et al.* 1999). We emphasize that, because of unknown low-level and intra-eruption emissions, loss of

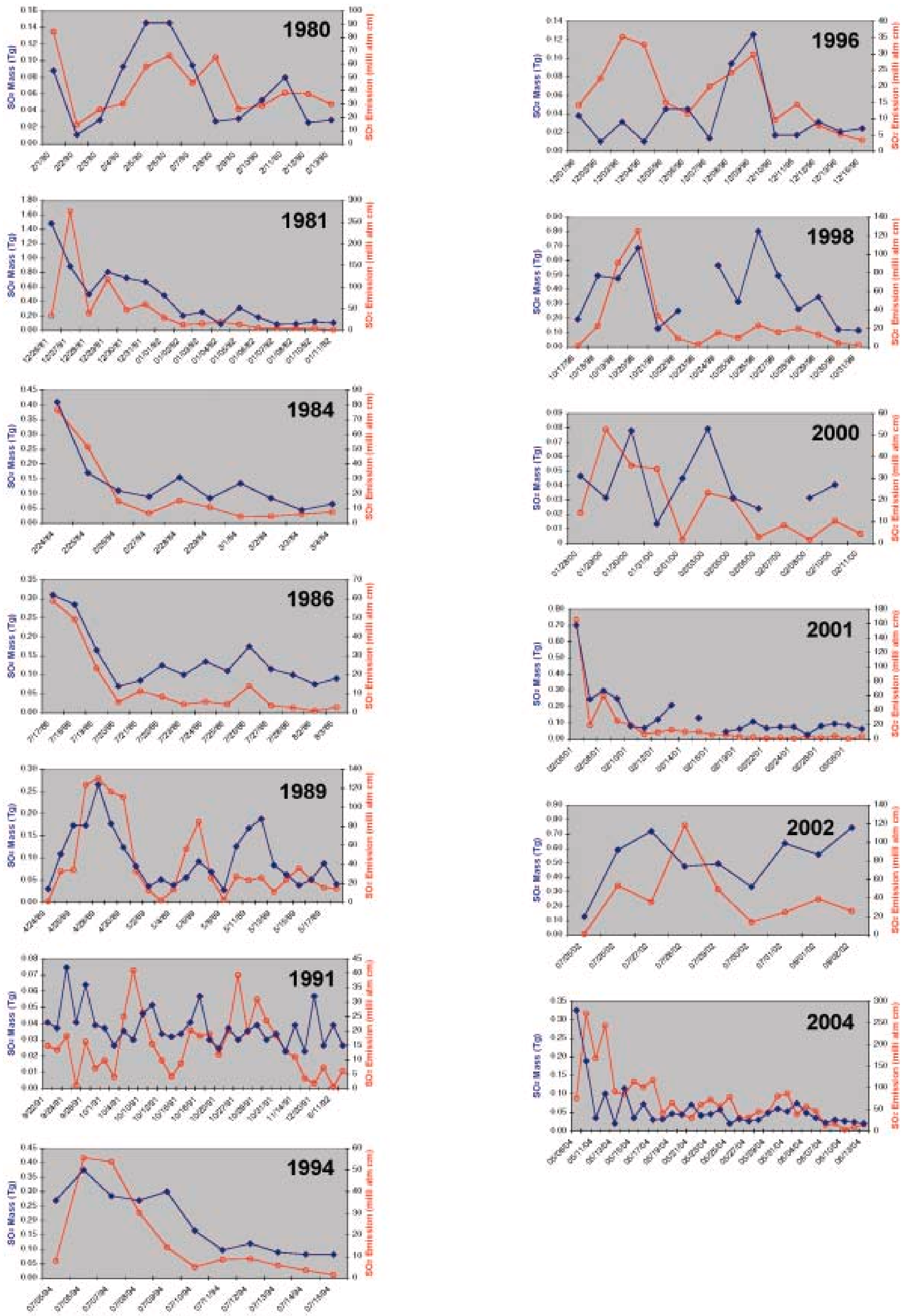

Figure 3. Daily SO_2 fluxes (solid diamonds) and 'instantaneous' emission levels (open circles) for the Nyamuragira eruptions, 1980–2004. The 1987–88 event is not shown because there were only 2 days of observed activity. Instantaneous emissions represent the SO_2 release at the time of the TOMS overpass, determined from the TOMS pixel over or adjacent to the volcano. Gaps occur where there was no TOMS coverage over the volcano. Both of these datasets give an indication of Nyamuragira's activity levels over the course of each eruption period.

Table 3. Comparative summary of Nyamuragira SO_2 emissions.

Nyamuragira activity (this work)	
Largest emission, total eruption	4.11 Tg (1981–82)
Largest emission, 1-day total	1.66 Tg (27 December 1981)
Largest average emission rate	0.26 Tg/day over 9 days (2002)
Total emissions, 14 eruptions between 1979 and 2005	24.51 Tg
Average annual emissions, 1979–2005	0.94 Tg/year
Global volcanic emissions	13.4 Tg/year*
Total emission, single eruption event, 1991 Mt Pinatubo†	18 Tg
Passive degassing, 1987–1995 Mt Etna‡	2.01 Tg/year

*Andres and Kasgnoc (1998); †Guo *et al.* (2004); ‡Bruno *et al.* (1999).

SO_2 by wet/dry deposition and observation interferences by meteorological clouds, there is certainly even more SO_2 released by Nyamuragira that was not measured or could be extrapolated from our TOMS observations.

Nyamuragira's productivity also reflects a relatively high efficiency of sulfur gas release per solid emissions. Figure 4 (after Blake 2003) shows the relationship of Nyamuragira's eruptions for which both associated lava/tephra and SO_2 emissions are known (1980, 1981, 1984, 1986, 1987 and 1989), in comparison to other tectonic settings. Non-arc settings include the Galapagos islands, Iceland and Hawaii; arc settings include Pacific Ring of fire volcanoes in Alaska, Indonesia, mainland USA and Mexico. The 1% gas : solid mass ratio is plotted for reference. With relatively few data points, it is difficult to speculate on complex trends, but note that for the most productive (e.g. >1 Tg SO_2 release) events Nyamuragira has a significantly higher gas : solid release ratio than either arc or other non-arc volcanoes.

Our results indicate that the majority of Nyamuragira's gas production tends to occur within 1–3 days, and it would be interesting to determine similar information regarding the lava/tephra production of these events. For example, Wadge (1981) found that lava emission tends to peak early in effusive basaltic eruptions (Mauna

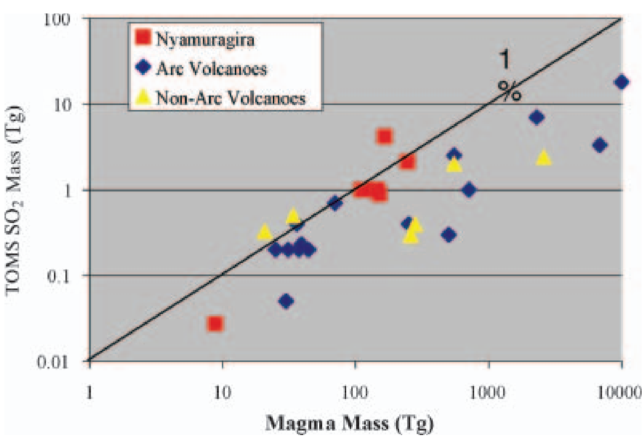

Figure 4. SO_2 emission data versus equivalent magma mass. Emission volumes of lava and/or tephra were converted to magma equivalent mass using densities of 2.6 and 1.0 g/cm^3, respectively. The 1% gas : magma mass ratio line is plotted for reference. Data sources: Blake (2003) and references cited therein; Head (2006); Smithsonian GVN; this work.

Loa, Kilauea and Etna), then gradually wanes as magma chamber overpressure decreases. It is beyond the scope of this gas-analysis work to examine the sulfur source, but future studies could focus on the feeder system for Nyamuragira as an indicator of the magnitude and longevity of future events. Burt *et al.* (1994) noted that, since 1980, the magma supply rate at Nyamuragira increased significantly over the previous 80 years of activity, primarily due to a higher eruption frequency. They suggest a 'pressure-cooker' model of activity, whereby eruptions follow when the magma exerts a pressure exceeding the physical strength of the confining reservoir rocks. Burt *et al.* (1994) suggest that the volume of lava erupted during each event by this mechanism is random (which is consistent with our observations of gas release), governed primarily by the pressure build-up and release. There is certainly no evidence at this point to suggest any limitations on Nyamuragira's current activity cycle; the past 25 years of SO_2 emissions have been remarkably consistent as well as productive (figure 2).

Despite the very high productivity, there is little known about the influence of Nyamuragira's emissions on atmospheric chemistry or climate (e.g. Bluth *et al.* 1997), although Mao and Robock (1998) suggest that the 1981 eruption produced a measurable cooling. The majority of emissions have been emplaced into the lower to middle troposphere, except for the 1981 eruption, which reached the tropopause (Krueger *et al.* 1996). The impacts of Nyamuragira activity on tropospheric sulfate loading have not been examined but could produce significant meteorological changes because sulfate aerosols can act as cloud condensation nuclei.

Based on 147 images showing progressive SO_2 decrease in drifting cloud masses, emissions in the low to mid-troposphere (5–10 km) exhibited SO_2 removal rates of 50–85% per day ($k=8.02\times10^{-6}$ to $2.20\times10^{-5}\,s^{-1}$). During the 1981 eruption, we found somewhat slower decay rates of 30% and 35% per day ($k=4.13\times10^{-6}$ and $4.99\times10^{-6}\,s^{-1}$) for clouds at 14–15 km. These rates are consistent with those found by Oppenheimer *et al.* (1998), based on a limited survey of volcanogenic emissions into the troposphere, who suggested that the dominant SO_2 removal processes involved multiphase reactions of chemical oxidation to sulfate and particle scavenging.

Although Nyamuragira's near-continuous emissions make the identification of a discrete cloud segment difficult, it was possible to compare our resampling method with the decay of a discrete cloud emitted on 27 April 1989. On the following 2 days the cloud drifted slowly to the east at an altitude of at least 10 km, while an ongoing emission at roughly 3–5 km altitude was observed over the volcano and consistently drifting to the north and northwest. Based upon the SO_2 mass evolution in the E-drifting cloud, a removal rate of 40% per day ($k=5.91\times10^{-6}\,s^{-1}$) was determined; by contrast, decay of the low-altitude cloud suggested a faster removal rate of 70% per day ($1.39\times10^{-5}\,s^{-1}$), as would be expected due to higher removal rates in the lower troposphere and boundary layer (Oppenheimer *et al.* 1998).

5. Conclusions

We have developed and tested a new method of extracting daily SO_2 fluxes from TOMS satellite data, for more accurately quantifying emissions from continuously erupting volcanoes. This method accounts for the rapid loss of SO_2 following emission, which cannot be evaluated using standard 'snapshot' analyses from once-daily images. We re-evaluated the SO_2 emissions from Nyamuragira volcano (DR Congo) to assess its productivity during 1979–2005.

Based upon our reanalysis of the TOMS data, we find that Nyamuragira produced a minimum of 25 Tg of SO_2 between 1979 and 2005, from 14 separate eruptive periods. The majority of these emissions were tropospheric, at altitudes between 5 and 10 km, and calculated removal rates ranged between 60% and 85% per day [k=(1.06−1.86)×10^{-5} s^{-1}]. High-altitude plumes at 14–15 km had significantly slower removal rates, up to 30% per day (k=4.13×10^{-6} s^{-1}).

Comparing our new method to previous evaluations of Nyamuragira emissions, the largest eruption in terms of gas release occurred in 1981, producing over 4 Tg (this is an increase over Krueger *et al.*'s 1996 estimate of 3 Tg using an earlier version of TOMS data). Our flux estimate for 6 February 2001 (~0.74 Tg/day) is in agreement with that of Carn and Bluth (2003), using a similar methodology, but again using earlier versions of the TOMS SO_2 algorithm and data.

Despite the large amount of SO_2 we calculate to be emitted from Nyamuragira, we strongly suspect that we have underestimated the emitted SO_2. We were able to use our 'flux' method for 85% of the possible days of TOMS-observed emissions; nearly every comparison between the flux and snapshot methods produces higher SO_2 estimates using the former. Furthermore, this study only evaluates SO_2 emitted during major eruptive activity. As noted by Carn and Bluth (2003), there is almost no information on passive degassing characteristics of Nyamuragira; future work with the more sensitive OMI satellite sensor should provide important new information on Nyamuragira's overall productivity.

Based upon our evaluation of the past 25 years of activity, Nyamuragira volcano is the world's largest volcanic source of SO_2. However, the effects of its eruptions on the surrounding population and the atmosphere have not been well documented. Its persistent and voluminous activity over the past two decades features characteristically S-rich emissions, and a tendency to focus production of these emissions over relatively short periods (table 3). Thus, Nyamuragira volcano holds the potential to create serious short-term and ongoing atmospheric and environmental impacts at both regional and global scales, and future work could focus on assessing these impacts.

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Appendix: Analysis results for all TOMS-observed Nyamuragira SO_2 clouds

Date	Overpass time (GMT)*	Cloud height (km amsl)†	Removal rate (%/day)	Snapshot SO_2 mass (Tg)	Calculated 24-h SO_2 flux (Tg/day)
01-Feb-80	1025	8	65	0.0925	0.1350
02-Feb-80	1045	7	60	0.0618	0.0230
03-Feb-80	0920	8	65	0.0366	0.0415
04-Feb-80	0939	10	65	0.0336	0.0485
05-Feb-80	0957	8	65	0.0496	0.0930
06-Feb-80	1016	8	80	0.0559	0.1060
07-Feb-80	1035	na	80	0.0449	0.0735
08-Feb-80	1054	5	60	0.0622	0.1040
09-Feb-80	0929	4	65	0.0138	0.0420
10-Feb-80	0948	na	65	0.0241	0.0460
11-Feb-80	1007	na	65	0.0383	0.0615
12-Feb-80	1025	10	65	0.0617	0.0600
13-Feb-80	1044	6	65	0.0085	0.0480
26-Dec-81	1013	8	80	0.1392	0.2000
27-Dec-81	1032	14	80	0.9843	1.6550
28-Dec-81	0908–1051	15	30	0.7867	0.2350
29-Dec-81	0926	na	60	0.5738	0.7060
30-Dec-81	0944	14	35	0.2746	0.2840
31-Dec-81	1002	na	75	0.3096	0.3610
01-Jan-82	1020	12	60	0.2336	0.1755
02-Jan-82	1039	12	65	0.0617	0.0775
03-Jan-82	0914	14	65	0.0634	0.0935

Date	Overpass time (GMT)*	Cloud height (km amsl)†	Removal rate (%/day)	Snapshot SO_2 mass (Tg)	Calculated 24-h SO_2 flux (Tg/day)
04-Jan-82	0933	15	65	0.0566	0.1170
05-Jan-82	0951	14	65	0.0948	0.0840
06-Jan-82	1009	11	65	0.0146	0.0320
07-Jan-82	1028	10	65	0.0220	0.0295
08-Jan-82	1046	7	65	0.0148	0.0290
10-Jan-82	0940	6	65	0.0048	0.0275
11-Jan-82	0958	8	50	0.0030	0.0080
24-Feb-84	1013	5	70	0.2254	0.3840
25-Feb-84	1031	6	65	0.3039	0.2580
26-Feb-84	1026	6	65	0.0741	0.0740
27-Feb-84	0925	5	65	0.0122	0.0340
28-Feb-84	0943	6	65	0.0342	0.0770
29-Feb-84	1001	5	65	0.0163	0.0540
01-Mar-84	1019	7	55	0.0130	0.0235
02-Mar-84	1037	na	na	0.0239	na
03-Mar-84	0912	6	65	0.0058	0.0315
04-Mar-84	0931	5	65	0.0385	0.0385
17-Jul-86	1003	9	70	0.1944	0.2950
18-Jul-86	1022	5	65	0.1541	0.2460
19-Jul-86	1040	6	65	0.0882	0.1170
20-Jul-86	0915	9	85	0.0140	0.0280
21-Jul-86	0932	5	55	0.0767	0.0570
22-Jul-86	0951	6	70	0.0272	0.0420
23-Jul-86	1009	6	70	0.0088	0.0220
24-Jul-86	1027	8	75	0.0135	0.0290
25-Jul-86	0903	na	na	0.0226	na
26-Jul-86	0921	na	75	0.0497	0.0705
27-Jul-86	0939	5	70	0.0049	0.0185
28-Jul-86	0957	7	55	0.0010	0.0135
02-Aug-86	0945	na	na	0.0049	na
03-Aug-86	1002	6	65	0.0040	0.0145
31-Dec-87	0914	4	85	0.0126	0.0235
01-Jan-88	0932	na	na	0.0036	na
24-Apr-89	0931	na	na	0.0023	na
25-Apr-89	0949	6	75	0.0485	0.0700
26-Apr-89	1007	na	na	0.0727	na
27-Apr-89	0841	na	70	0.1830	0.2650
28-Apr-89	0859	na	65	0.1532	0.2800
29-Apr-89	0918	na	70	0.1872	0.2500
30-Apr-89	0936	7	70	0.1599	0.2370
01-May-89	0953	na	65	0.0488	0.0700
02-May-89	1011	7	75	0.0159	0.0270
03-May-89	0846	na	na	0.0042	na
04-May-89	0904	8	70	0.0130	0.0285
05-May-89	0922	6	70	0.0643	0.1200
06-May-89	0940	6	75	0.1134	0.1820
07-May-89	0958	4	75	0.0375	0.0545
08-May-89	1016	6	60	0.0054	0.0060
10-May-89	0909	8	70	0.0340	0.0580
11-May-89	0927	na	75	0.0310	0.0495
12-May-89	0945	na	65	0.0366	0.0555
13-May-89	1003	na	na	0.0233	na
14-May-89	0838	na	65	0.0400	0.0510

Date	Overpass time (GMT)*	Cloud height (km amsl)†	Removal rate (%/day)	Snapshot SO_2 mass (Tg)	Calculated 24-h SO_2 flux (Tg/day)
15-May-89	0856	na	65	0.0380	0.0770
16-May-89	0914	7	70	0.0352	0.0500
17-May-89	0932	8	65	0.0181	0.0330
18-May-89	0950	5	65	0.0074	0.0310
22-Sep-91	0834	4	50	0.0158	0.0265
23-Sep-91	0852	4	55	0.0090	0.0240
24-Sep-91	0910	3	90	0.0133	0.0325
25-Sep-91	0927	na	na	0.0019	na
28-Sep-91	0847	na	60	0.0228	0.0290
29-Sep-91	0855	4	65	0.0056	0.0125
01-Oct-91	0930	5	65	0.0045	0.0175
03-Oct-91	0833	na	na	0.0071	na
04-Oct-91	0840	4	70	0.0173	0.0445
06-Oct-91	0916	5	65	0.0261	0.0730
10-Oct-91	0843	8	85	0.0225	0.0470
11-Oct-91	0901	5	70	0.0086	0.0275
12-Oct-91	0918	4	85	0.0095	0.0175
15-Oct-91	0828	4	70	0.0011	0.0075
16-Oct-91	0846	na	70	0.0070	0.0155
17-Oct-91	0904	na	70	0.0310	0.0360
18-Oct-91	0922	3	85	0.0146	0.0325
19-Oct-91	0939	9	70	0.0228	0.0335
20-Oct-91	0814	na	70	0.0068	0.0210
24-Oct-91	0924	na	65	0.0102	0.0355
27-Oct-91	0834	5	85	0.0241	0.0700
28-Oct-91	0852	7	75	0.0093	0.0350
29-Oct-91	0910	5	85	0.0234	0.0550
30-Oct-91	0927	5	75	0.0353	0.0420
31-Oct-91	0945	4	65	0.0174	0.0325
13-Nov-91	0825	na	75	0.0094	0.0230
14-Nov-91	0843	4	65	0.0015	0.0195
20-Nov-91	0846	na	na	0.0063	na
20-Dec-91	0900	na	na	0.0033	na
04-Feb-92	0847	na	na	0.0129	na
11-Jun-92	0833	na	na	0.0009	na
27-Jun-92	0805	na	na	0.0109	na
05-Jul-94	1039	4	80	0.0579	0.0600
06-Jul-94	1021	na	70	0.3752	0.4180
07-Jul-94	1003	na	65	0.2064	0.4040
08-Jul-94	0946	na	85	0.1950	0.2280
09-Jul-94	0928	4	75	0.0699	0.1075
10-Jul-94	0910	7	75	0.0649	0.0390
11-Jul-94	0853	8	70	0.0388	0.0645
12-Jul-94	0833	na	na	0.0670	na
13-Jul-94	0815	na	70	0.0111	0.0445
14-Jul-94	0950	na	na	0.0272	na
16-Jul-94	0914	na	na	0.0112	na
01-Dec-96	0848	na	na	0.0496	na
02-Dec-96	0822	6	65	0.0583	0.0785
03-Dec-96	0935	6	70	0.0788	0.1235
04-Dec-96	0908	6	70	0.0600	0.1150
05-Dec-96	0841	7	70	0.0297	0.0525
06-Dec-96	0814	7	85	0.0141	0.0405
07-Dec-96	0927	na	85	0.0331	0.0695

Date	Overpass time (GMT)*	Cloud height (km amsl)†	Removal rate (%/day)	Snapshot SO_2 mass (Tg)	Calculated 24-h SO_2 flux (Tg/day)
08-Dec-96	0901	6	70	0.0431	0.0845
09-Dec-96	0834	5	80	0.0435	0.1040
10-Dec-96	0807	6	70	0.0187	0.0340
11-Dec-96	0920	6	70	0.0296	0.0505
12-Dec-96	0854	5	70	0.0171	0.0275
13-Dec-96	0826	6	70	0.0104	0.0183
16-Dec-96	0846	4	60	0.0056	0.0120
17-Oct-98	0942	na	na	0.0064	na
18-Oct-98	0858	8	70	0.1037	0.1480
19-Oct-98	0952	7	65	0.3576	0.5840
20-Oct-98	0908	na	80	0.3089	0.8060
21-Oct-98	1003	7	70	0.1194	0.2160
22-Oct-98	0920	na	70	0.0304	0.0590
23-Oct-98	1015	5	85	na	0.0170
24-Oct-98	0931	na	70	0.0616	0.1005
25-Oct-98	0847	na	na	0.0642	na
26-Oct-98	0942	6	70	0.1116	0.1505
27-Oct-98	0858	na	na	0.1032	na
28-Oct-98	0953	5	80	0.0624	0.1280
29-Oct-98	0909	na	65	0.0514	0.0865
30-Oct-98	1004	na	65	0.0237	0.0280
31-Oct-98	0920	na	na	0.0150	na
28-Jan-00	0857	6	75	0.0177	0.0210
29-Jan-00	0951	7	85	0.0234	0.0790
30-Jan-00	0907	na	75	0.0320	0.0540
31-Jan-00	1002	na	75	0.0141	0.0515
01-Feb-00	0918	na	na	0.0026	na
03-Feb-00	0929	na	65	0.0133	0.0350
05-Feb-00	0939	na	70	0.0159	0.0305
06-Feb-00	0855	na	na	0.0042	na
07-Feb-00	0906	na	70	0.0018	0.0125
08-Feb-00	0906	na	na	0.0023	na
10-Feb-00	0916	7	75	0.0059	0.0155
11-Feb-00	1011	na	na	0.0065	na
06-Feb-01	0935	10	70	0.3925	0.7350
07-Feb-01	0849	6	70	0.0514	0.0850
08-Feb-01	0942	8	70	0.1326	0.2660
09-Feb-01	0856	9	65	0.0603	0.1110
10-Feb-01	0949	5	65	0.0633	0.0865
11-Feb-01	0903	4	70	0.0155	0.0315
12-Feb-01	0956	na	75	0.0332	0.0420
13-Feb-01	0911	5	85	0.0178	0.0575
14-Feb-01	1003	na	na	0.0442	na
15-Feb-01	0918	6	65	0.0254	0.0455
16-Feb-01	1010	na	na	0.0252	na
17-Feb-01	0925	na	65	0.0105	0.0265
19-Feb-01	0932	6	75	0.0121	0.0140
21-Feb-01	0939	na	na	0.0097	na
22-Feb-01	0853	na	na	0.0049	na
23-Feb-01	0946	na	na	0.0089	na
24-Feb-01	0900	na	na	0.0035	na
25-Feb-01	0954	4	70	0.0096	0.0195
28-Feb-01	0915	na	na	0.0074	na
02-Mar-01	0922	7	65	0.0120	0.0195

Date	Overpass time (GMT)*	Cloud height (km amsl)†	Removal rate (%/day)	Snapshot SO_2 mass (Tg)	Calculated 24-h SO_2 flux (Tg/day)
06-Mar-01	0936	na	na	0.0009	na
07-Mar-01	0850	6	65	0.0070	0.0175
25-Jul-02	0848	na	na	0.0068	na
26-Jul-02	0938	5	85	0.1621	0.3430
27-Jul-02	0849	6	70	0.1771	0.2300
28-Jul-02	0939	10	80	0.3151	0.7620
29-Jul-02	0851	na	90	0.4070	0.3140
30-Jul-02	0940	5	85	0.0362	0.0900
31-Jul-02	0852	5	85	0.0669	0.1590
01-Aug-02	0943	na	85	0.0710	0.2490
02-Aug-02	0854	na	70	0.0986	0.1670
08-May-04	0856	4	70	0.0481	0.0885
10-May-04	0855	6	80	0.1834	0.3170
11-May-04	0943	5	85	0.1087	0.1960
12-May-04	0853	6	85	0.1763	0.2850
13-May-04	0942	na	na	0.1067	na
14-May-04	0852	na	70	0.0536	0.0980
15-May-04	0941	na	85	0.2111	0.1330
16-May-04	0851	na	70	0.0721	0.1180
17-May-04	0940	na	85	0.0604	0.1375
18-May-04	0850	5	70	0.0255	0.0470
19-May-04	0938	na	85	0.0222	0.0755
20-May-04	0849	6	75	0.0157	0.0455
21-May-04	0937	4	85	0.0692	0.0345
22-May-04	0848	4	85	0.0557	0.0710
23-May-04	0936	6	75	0.0384	0.0850
24-May-04	0846	5	70	0.0432	0.0660
25-May-04	0935	na	70	0.0387	0.0915
26-May-04	0845	5	75	0.0138	0.0350
27-May-04	0933	na	na	0.0359	na
28-May-04	0844	8	70	0.0296	0.0525
29-May-04	0932	4	65	0.0271	0.0500
30-May-04	0843	6	80	0.0568	0.0940
31-May-04	0931	7	75	0.0223	0.1015
03-Jun-04	0840	na	80	0.0150	0.0450
04-Jun-04	0928	na	70	0.0376	0.0660
06-Jun-04	0927	6	65	0.0202	0.0535
07-Jun-04	0838	4	65	0.0079	0.0160
08-Jun-04	0926	3	65	0.0063	0.0200
10-Jun-04	0925	na	na	0.0036	na
12-Jun-04	0924	na	na	0.0101	na
13-Jun-04	0834	6	65	0.0027	0.00180

*Approximate time of the TOMS overpass of the volcano.
†Above mean sea level, as determined by HYSPLIT modelling.