

Exceptional sulphur degassing from Nyamuragira volcano, 1979-2005

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The sulphur dioxide (SO₂) output from Nyamuragira volcano has been monitored by the Total Ozone Mapping Spectrometer (TOMS) since 1979, and is evaluated here in order to quantify the emissions from this highly productive volcano. The majority of Nyamuragira's emissions were emplaced in the lower to middle troposphere, with SO₂ removal rates of 30-90 %/day ($k = 4.128 \times 10^{-6} \text{ s}^{-1} - 1.863 \times 10^{-5} \text{ s}^{-1}$). We test a new method of back-calculating persistent, effusive emission fluxes from once-daily observations, which accounts for this rapid daily removal of SO₂ that cannot be measured using satellite 'snapshots'. Twelve of the 14 eruptions during this period each produced ≥ 0.8 million tonnes (Mt) of SO₂. Nyamuragira erupted nearly 25 Mt of SO₂ during these eruptions, and likely emitted significantly more than we could measure by TOMS. Nyamuragira may be the largest volcanic source of sulphur to the atmosphere for the past few decades.

Keywords: Nyamuragira; Volcanoes; Sulphur dioxide; Satellite monitoring

Introduction

The motivation for this study is an ongoing effort to constrain the volcanogenic contribution to the global sulphur 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). However, discoveries of 'new' volcanic emissions, through new technology or methodologies, more intensive

examination of known eruptions, as well as discoveries of previously unknown or unquantified activity, have underscored the need for ongoing 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 NW of Nyiragongo volcano. The city of Goma (population ~500,000) lies only 18 km S Nyiragongo volcano, in a region under ongoing threat of lava flows and gas emissions. Nyamuragira's summit rises approximately 3 km above sea level, with a 2 x 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.

It's 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 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 2004, there have been 40 confirmed eruptive periods of Nyamuragira (Smithsonian Global Volcanism Network 2006), with an average repose time of only 3.5 years. The period of activity since 1980, with eruptions roughly every 2 years, represents it's highest known sustained period (25 years) of activity.

Nyamuragira is arguably the most active volcano in the world, yet relatively unstudied due to on-going political strife. Therefore it's contribution to the global terrestrial sulphur 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 1980 and 2004 were observed and measured by the satellite-based Total Ozone Mapping Spectrometer (TOMS). TOMS has produced an invaluable, near-continuous record of volcanic sulphur dioxide (SO₂) emission since 1978, and TOMS-based studies are a mainstay of satellite volcano gas monitoring, particularly for large, explosive eruptions (Carn *et al.* 2003). The current TOMS sensor, Earth Probe, has continued the dataset to the beginning of 2006, and a new sensor (OMI - Ozone Monitoring Instrument) will continue the record, with much finer spatial and spectral resolution (Krotkov *et al.* 2006). OMI's greater sensitivity to SO₂ and daily global contiguous coverage will allow routine studies of effusive activity, which 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 which can be tracked over the course of days, and an emitted mass can be calculated from analysing the daily mass total of the decaying cloud (e.g. Krueger *et al.* 1995; 2000; Guo *et al.* 2004). In contrast, Nyamuragira typically erupts for days to weeks at a time, producing a near-continuous emission of SO₂. A single daily image is capable of capturing the emitted plume; however, a difficulty arises from trying to determine how much of the SO₂ has been newly produced each day.

After emission, SO₂ is converted to sulphate aerosol at an altitude-dependent rate, and also scavenged from the plume by wet and dry deposition. It is not possible to directly

determine from TOMS data which SO₂ in the image is ‘new’ (<24 hours) and which is ‘old’ (>24 hours). 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 SO₂ from fresh up to 24 hours or more old. Oppenheimer *et al.* (1998) compiled measured SO₂ decay rates for tropospheric plumes, which range from 25% daily mass removal to 100% loss in less than one day. Thus to accurately evaluate SO₂ release by continuously emitting volcanoes it is necessary to account for SO₂ 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 SO₂, and assuming all SO₂ both ‘attached’ to the volcano and above that level was produced from within the previous 24 hours; (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’ SO₂ production. These results suggested decay rates of about 10-40% per day ($k = 1.219 - 5.912 \times 10^{-6} \text{ s}^{-1}$), 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. COSPEC, DOAS), with the assumption that the plume height and emission remained constant throughout the 24-hour period between TOMS emissions. Their results indicated

that calculated SO₂ fluxes could significantly exceed daily TOMS ‘snapshot’ SO₂ loads for continuously fed plumes. Decay rates calculated using plumes emitted in 1998 and 2001 ranged from 25-60% loss per day ($k = 3 \times 10^{-6}$ to $1 \times 10^{-5} \text{ s}^{-1}$, 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-hour period between TOMS overpasses. We summarise SO₂ cloud and meteorological characteristics of the 14 Nyamuragira eruptions between 1980 and 2004, and attempt to evaluate the total SO₂ release by this extremely productive system.

Analysis Methods

This work combines the standard method of TOMS SO₂ analyses for emitted volcanic clouds, and an important modification to retrieve SO₂ 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 SO₂ masses from the data are essentially identical (described by Krueger *et al.* 1995; 2000). The main concern with changes in spatial resolution occur because the TOMS sensors produce daily maps of SO₂; 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 SO₂ 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 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 <1 hr) 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.

[Insert table 1 about here]

Our method combines the standard TOMS SO₂ retrievals, with a resampling scheme to interpolate the TOMS data to a regular grid. This allows cloud mass to be correlated to distance from the volcano, thereby recording the daily emission history of an eruption. Assuming exponential decay of SO₂, we back-calculate the emission rate throughout the 24 hour period. We make several additional assumptions for this method:

-for each day that at least a portion of a TOMS-observed SO₂ cloud is located physically over the volcano's geometric coordinates, the volcano emitted SO₂ continuously throughout the previous 24-hour 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 the wind trajectory model to determine the length of emission (that is, depending 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-hour period, and from one day to the next; however, exponential SO₂ removal can be observed in the data during periods when the emission rate is constant. This removal of SO₂ from TOMS-based observation consists of both 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 SO₂ by meteorological clouds are possible causes of underestimation of the actual SO₂ flux.

-the cloud altitude, drift rate, and removal rate can vary from one day to the next, but are assumed constant throughout each 24-hour period.

The general procedure for deriving a daily SO₂ flux from TOMS data is presented graphically in Figure 1. The first step is to complete a standard TOMS SO₂ analysis, including creating an image using the iterative SO₂ retrieval (Figure 1a) and a tonnage analysis. This involves defining the region containing the SO₂ cloud and calculating a total mass for that region, determining SO₂ noise levels, and then subtracting a noise-equivalent SO₂ amount from the total to determine the net SO₂ mass. Figure 1a shows a simple case for a cloud over Nyamuragira, and drifting directly W, containing approximately 100

kilotonnes (kt) SO₂ using the ‘snapshot’ analysis.

[Insert figure 1 about here]

The next step is to perform a trajectory analysis, visually matching an on-line, modeled trajectory (HYSPLIT, HYbrid Single-Particle Lagrangian Integrated Trajectory; Draxler and Rolph 2003) with the TOMS-derived SO₂ cloud location, in order to estimate the 24-hour trajectory of the cloud (Figure 1b). Using the Global Reanalysis archived meteorology dataset, the best match in this example was the 5,000 m above ground level trajectory, which allows us to constrain the distance the SO₂ may have travelled from the volcano during the past 24 hours to roughly 500 km (Figure 1a). 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 hours earlier. The trajectory model results are initially defined above ground level; however, due to 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 3 cases) the obviously faulty result where the modelled cloud height is actually less than the true volcano height. However, this discrepancy has little effect on the overall analysis, because the total surface area around the volcano which is represented inaccurately is small compared to the daily drift distance of the SO₂ clouds.

The TOMS SO₂ data are then resampled to a 50 km grid using a nearest neighbour interpolation scheme in Terascan™ image processing software (Figure 1c). The cloud area

and instantaneous mass of the resampled cloud are compared to 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 1a. Because the SO₂ clouds may drift in any direction (or change direction in response to shifting wind directions), an orthogonal sampling scheme will not always sample the cloud perpendicular to the drift direction. 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, while maintaining a perpendicular sampling would be ideal, this would also introduce problems of over- and under-sampling portions of the cloud. In our method, the full cloud is sampled completely, and only once.

The resampled TOMS data are used to slice the plume into evenly spaced columns with distance from the volcano, to determine the mass change with distance (Figure 1d). In this example, the cloud travels 500 km in 24 hours (based upon modelled wind trajectories) which is covered by 10 columns. Therefore, cloud cross-sectional mass is calculated every 2.4 hours (Figure 1b). The data are then fit to a set of exponential decay curves. In the simple case depicted in Figure 1, a single decay rate is used; typically 3 or more curves are used to best match the data. Each data point then corresponds to a decay curve with its characteristic emission rate. To calculate the 24-hour flux, each individual (e.g. 2.4 hour) flux is summed. In this case, the total 24-hour flux is calculated as 160 kt (=10 x 16 000 t).

Recall that the standard TOMS analysis of the same data produced a tonnage of only 100 kt. This 60% difference represents the loss due to chemical conversion to sulphate, scavenging, and physical dispersion of SO₂ during a 24-hour emission period, which

cannot be recognised in a standard TOMS analysis. While each event will vary in terms of environmental and emission characteristics, we believe that the underestimation of SO₂ emission by a snapshot analysis can be quite significant, and the calculation of daily fluxes more closely replicates the true emission characteristics of continuously-emitting volcanoes.

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 on-line 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 SO₂ emissions could be detected, which sometimes extended the activity beyond the reported duration. Table 2 and Figure 2 summarise the TOMS-observed activity for these events, which include 225 individual days (daily data are included in the Appendix). The total SO₂ flux is estimated as nearly 25 Mt (megatonnes), roughly 50% higher than that calculated by simply totalling the cloudmasses 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 60-85% per day ($k = 1.061-1.863 \times 10^{-5} \text{ s}^{-1}$), although individual daily removal rates ranged from 30 - 90% per day ($k = 4.128 \times 10^{-6}$ to $2.665 \times 10^{-5} \text{ s}^{-1}$).

[Insert table 2 about here]

[Insert figure 2 about here]

Despite the large increase in estimated SO₂ release from Nyamuragira using this new method of evaluation, it is likely that this study still underestimates the emitted SO₂. For example, the flux 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 plume, 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-hour drift constraint was determined directly from the model results. A wind trajectory ‘match’ was defined here as +/- 10° 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-hour 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 SO₂ emissions was undertaken to determine the daily ‘instantaneous’ emissions. For this we assume that TOMS pixels over or immediately adjacent to the volcano contain the SO₂ most recently emitted and are a relatively objective measure of the emission rate at the time of the daily TOMS overpass. The daily SO₂ 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 SO₂ production. The majority of SO₂ emission occurs during the first few days of 8 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 SO₂ was produced in 3 days or less. A notable exception was the 1991 eruption, in which there was not a single day which produced more than 10% of the total. The instantaneous emission rates typically track with the daily flux estimates, which suggests that the fluxes are accurately reflecting at least the major trends in SO₂ emissions. An apparent exception is the July 2002 eruption, but unfortunately the end of this event was not observed, because TOMS data were unavailable for August 3-9.

[Insert figure 3 about here]

The length of TOMS-observed activity ranges from as little as 2 days in the 1987-88 eruption, up to over 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 SO₂ 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 quite large; peak emissions greater than 100 kt occurred in 11 out of the 14 events; the highest 24-hour emission, 1.7 Mt, occurred during the 1981 eruption as part of a total release of over 4 Mt SO₂.

The SO₂ release by Nyamuragira totals 25 Mt from 1980 to 2004. Overall, Nyamuragira's eruptive cycles consistently released high SO₂ amounts, averaging almost 2 Mt per event: only two events (1987, 2000) released *less* than 0.8 Mt of SO₂.

Discussion

The 25 Mt SO₂ emitted by Nyamuragira during its 14 eruptive periods makes a significant impact on evaluations of global volcanic sulphur production (Table 3). The most comprehensive evaluation of global volcanic emissions has been generated by Andres and Kasgnoc (1998), who estimated SO₂ emissions of 13.4 Mt per year 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 SO₂ flux. The volcano has produced more than 0.8 Mt of SO₂ a total of 12 times (Table 2). During the same time period, only 10 other eruptions in the world have achieved this same level (TOMS on-line research website, <http://toms.umbc.edu/>); the greatest emission from an individual event was Mt. Pinatubo in 1991 with an explosive release of approximately 18 Mt SO₂ (Guo *et al.* 2004). Taken on an average annual basis, Nyamuragira's output is of the same order as the passive emissions of Mt. 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 Mt per year (Bruno *et al.* 1999). We emphasise that, because of unknown low-level and intra-eruption emissions, loss of SO₂ by wet/dry deposition and observation interferences by meteorological clouds, there is certainly even more SO₂ released by Nyamuragira that was not measured or could be extrapolated from TOMS observations.

[Insert table 3 about here]

Nyamuragira's productivity also reflects a relatively high efficiency of sulphur gas release per solid emissions. Figure 4 (after Blake 2003) shows the relationship of Nyamuragira's eruptions for which both associated lava/tephra and SO₂ 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 U.S. and Mexico. The 1% gas:solid mass ratio is plotted for reference. It is difficult with relatively few data points to speculate on complex trends, but note that for the most productive (e.g. >1 Mt SO₂ 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 tend 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 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 sulphur 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 last 25 years of SO₂ emissions have been remarkably consistent as well as productive (Figure 2).

[Insert figure 4 about here]

Despite the very high productivity, Nyamuragira has had few noticeable impacts on atmospheric chemistry or climate (Bluth *et al.* 1997), although Mao and Robock (1998) suggest 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 sulphate loading have not been examined, but could produce significant meteorological changes as sulphate aerosols can act as cloud condensation nuclei.

Based on 147 images showing progressive SO₂ decrease in drifting cloud masses, emissions in the low to mid troposphere (5-10 km) exhibited SO₂ removal rates of 50-85%. During the 1981 eruption, we found decay rates of 30 and 35% for clouds at 14-15 km. These conditions are consistent with those of Oppenheimer *et al.* (1998), based on a limited survey of volcanogenic emissions into the troposphere, who suggested that the

dominant SO₂ removal processes involved multiphase reactions of chemical oxidation to sulphate 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 April 27, 1989. On the following two days the cloud drifted slowly to the E 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 N and NW. Based upon the SO₂ mass evolution in the E-drifting cloud, a removal rate of 40% per day ($k = 6.008 \times 10^{-6} \text{ s}^{-1}$) was determined; in contrast, the low altitude emission was estimated with a somewhat larger removal rate of 70% per day ($1.393 \times 10^{-5} \text{ s}^{-1}$), as would be expected due to higher removal rates in the lower troposphere and boundary layer (Oppenheimer *et al.* 1998).

Conclusions

We have developed and tested a new method of extracting daily SO₂ fluxes from TOMS satellite data, for more accurately quantifying emissions from continuously erupting volcanoes. This method accounts for the rapid loss of SO₂ following emission, which cannot be evaluated using standard 'snapshot' analyses from once-daily images. We re-evaluated the SO₂ emissions from Nyamuragira volcano (DR Congo) in order to assess its productivity and potential impacts on the atmosphere.

Based upon our reanalysis of the TOMS data, we find that Nyamuragira has produced a total of >25 Mt of SO₂ from 14 separate eruptive periods between 1980 and 2004. The majority of these emissions were tropospheric, at altitudes between 5 and 10 km, and

calculated removal rates ranged between 60-85% per day ($k = 1.061-1.863 \times 10^{-5} \text{ s}^{-1}$). High altitude plumes at 14-15 km had significantly lower removal rates, to 30% per day ($k = 4.128 \times 10^{-6} \text{ s}^{-1}$).

The largest eruption in terms of gas release occurred in 1981, producing over 4 Mt (this is an increase over Krueger *et al.*'s 1996 estimate of 3 Mt using an earlier version of TOMS data). Our flux estimate for February 6, 2001 ($\sim 0.74 \text{ Mt/day}$) is in agreement with that of Carn and Bluth (2003), using a similar methodology, but again using an earlier version of the TOMS SO_2 algorithm.

Despite the large amount of SO_2 we calculate to be emitted from Nyamuragira, we strongly suspect that we underestimate the emitted SO_2 . We were able to calculate the 24-hour flux 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. Further, this study only evaluates SO_2 emitted during major eruptive activity. As noted by Carn and Bluth (2003), there is virtually no information on passive degassing characteristics of Nyamuragira.

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. It's 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.

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APPENDIX: Analysis results for all TOMS-observed Nyamuragira SO₂ clouds

Date	Overpass Time (GMT) ¹	Cloud Height (km amsl) ²	Removal Rate (%/day)	Snapshot SO ₂ Mass (kt)	Calculated 24-Hour SO ₂ Flux (kt)
01-Feb-80	1025	8	65	92.5	135.0
02-Feb-80	1045	7	60	61.8	23.0
03-Feb-80	0920	8	65	36.6	41.5
04-Feb-80	0939	10	65	33.6	48.5
05-Feb-80	0957	8	65	49.6	93.0
06-Feb-80	1016	8	80	55.9	106.0
07-Feb-80	1035	na	80	44.9	73.5
08-Feb-80	1054	5	60	62.2	104.0
09-Feb-80	0929	4	65	13.8	42.0
10-Feb-80	0948	na	65	24.1	46.0
11-Feb-80	1007	na	65	38.3	61.5
12-Feb-80	1025	10	65	61.7	60.0
13-Feb-80	1044	6	65	8.5	48.0
26-Dec-81	1013	8	80	139.2	200.0
27-Dec-81	1032	14	80	984.3	1655.0
28-Dec-81	0908-1051	15	30	786.7	235.0
29-Dec-81	0926	na	60	573.8	706.0
30-Dec-81	0944	14	35	274.6	284.0
31-Dec-81	1002	na	75	309.6	361.0
01-Jan-82	1020	12	60	233.6	175.5
02-Jan-82	1039	12	65	61.7	77.5
03-Jan-82	0914	14	65	63.4	93.5
04-Jan-82	0933	15	65	56.6	117.0
05-Jan-82	0951	14	65	94.8	84.0
06-Jan-82	1009	11	65	14.6	32.0
07-Jan-82	1028	10	65	22.0	29.5
08-Jan-82	1046	7	65	14.8	29.0
10-Jan-82	0940	6	65	4.8	27.5
11-Jan-82	0958	8	50	3.0	8.0
24-Feb-84	1013	5	70	225.4	384.0
25-Feb-84	1031	6	65	303.9	258.0
26-Feb-84	1026	6	65	74.1	74.0
27-Feb-84	0925	5	65	12.2	34.0
28-Feb-84	0943	6	65	34.2	77.0
29-Feb-84	1001	5	65	16.3	54.0
01-Mar-84	1019	7	55	13.0	23.5
02-Mar-84	1037	na	na	23.9	na

03-Mar-84	0912	6	65	5.8	31.5
04-Mar-84	0931	5	65	38.5	38.5
17-Jul-86	1003	9	70	194.4	295.0
18-Jul-86	1022	5	65	154.1	246.0
19-Jul-86	1040	6	65	88.2	117.0
20-Jul-86	0915	9	85	14.0	28.0
21-Jul-86	0932	5	55	76.7	57.0
22-Jul-86	0951	6	70	27.2	42.0
23-Jul-86	1009	6	70	8.8	22.0
24-Jul-86	1027	8	75	13.5	29.0
25-Jul-86	0903	na	na	22.6	na
26-Jul-86	0921	na	75	49.7	70.5
27-Jul-86	0939	5	70	4.9	18.5
28-Jul-86	0957	7	55	1.0	13.5
02-Aug-86	0945	na	na	4.9	na
03-Aug-86	1002	6	65	4.0	14.5
31-Dec-87	0914	4	85	12.6	23.5
01-Jan-88	0932	na	na	3.6	na
24-Apr-89	0931	na	na	2.3	na
25-Apr-89	0949	6	75	48.5	70.0
26-Apr-89	1007	na	na	72.7	na
27-Apr-89	0841	na	70	183.0	265.0
28-Apr-89	0859	na	65	153.2	280.0
29-Apr-89	0918	na	70	187.2	250.0
30-Apr-89	0936	7	70	159.9	237.0
01-May-89	0953	na	65	48.8	70.0
02-May-89	1011	7	75	15.9	27.0
03-May-89	0846	na	na	4.2	na
04-May-89	0904	8	70	13.0	28.5
05-May-89	0922	6	70	64.3	120.0
06-May-89	0940	6	75	113.4	182.0
07-May-89	0958	4	75	37.5	54.5
08-May-89	1016	6	60	5.4	6.0
10-May-89	0909	8	70	34.0	58.0
11-May-89	0927	na	75	31.0	49.5
12-May-89	0945	na	65	36.6	55.5
13-May-89	1003	na	na	23.3	na
14-May-89	0838	na	65	40.0	51.0
15-May-89	0856	na	65	38.0	77.0
16-May-89	0914	7	70	35.2	50.0
17-May-89	0932	8	65	18.1	33.0
18-May-89	0950	5	65	7.4	31.0

22-Sep-91	0834	4	50	15.8	26.5
23-Sep-91	0852	4	55	9.0	24.0
24-Sep-91	0910	3	90	13.3	32.5
25-Sep-91	0927	na	na	1.9	na
28-Sep-91	0847	na	60	22.8	29.0
29-Sep-91	0855	4	65	5.6	12.5
01-Oct-91	0930	5	65	4.5	17.5
03-Oct-91	0833	na	na	7.1	na
04-Oct-91	0840	4	70	17.3	44.5
06-Oct-91	0916	5	65	26.1	73.0
10-Oct-91	0843	8	85	22.5	47.0
11-Oct-91	0901	5	70	8.6	27.5
12-Oct-91	0918	4	85	9.5	17.5
15-Oct-91	0828	4	70	1.1	7.5
16-Oct-91	0846	na	70	7.0	15.5
17-Oct-91	0904	na	70	31.0	36.0
18-Oct-91	0922	3	85	14.6	32.5
19-Oct-91	0939	9	70	22.8	33.5
20-Oct-91	0814	na	70	6.8	21.0
24-Oct-91	0924	na	65	10.2	35.5
27-Oct-91	0834	5	85	24.1	70.0
28-Oct-91	0852	7	75	9.3	35.0
29-Oct-91	0910	5	85	23.4	55.0
30-Oct-91	0927	5	75	35.3	42.0
31-Oct-91	0945	4	65	17.4	32.5
13-Nov-91	0825	na	75	9.4	23.0
14-Nov-91	0843	4	65	1.5	19.5
20-Nov-91	0846	na	na	6.3	na
20-Dec-91	0900	na	na	3.3	na
04-Feb-92	0847	na	na	12.9	na
11-Jun-92	0833	na	na	0.9	na
27-Jun-92	0805	na	na	10.9	na
05-Jul-94	1039	4	80	57.9	60.0
06-Jul-94	1021	na	70	375.2	418.0
07-Jul-94	1003	na	65	206.4	404.0
08-Jul-94	0946	na	85	195.0	228.0
09-Jul-94	0928	4	75	69.9	107.5
10-Jul-94	0910	7	75	64.9	39.0
11-Jul-94	0853	8	70	38.8	64.5
12-Jul-94	0833	na	na	67.0	na
13-Jul-94	0815	na	70	11.1	44.5
14-Jul-94	0950	na	na	27.2	na
16-Jul-94	0914	na	na	11.2	na

01-Dec-96	0848	na	na	49.6	na
02-Dec-96	0822	6	65	58.3	78.5
03-Dec-96	0935	6	70	78.8	123.5
04-Dec-96	0908	6	70	60.0	115.0
05-Dec-96	0841	7	70	29.7	52.5
06-Dec-96	0814	7	85	14.1	40.5
07-Dec-96	0927	na	85	33.1	69.5
08-Dec-96	0901	6	70	43.1	84.5
09-Dec-96	0834	5	80	43.5	104.0
10-Dec-96	0807	6	70	18.7	34.0
11-Dec-96	0920	6	70	29.6	50.5
12-Dec-96	0854	5	70	17.1	27.5
13-Dec-96	0826	6	70	10.4	18.3
16-Dec-96	0846	4	60	5.6	12.0
17-Oct-98	0942	na	na	6.4	na
18-Oct-98	0858	8	70	103.7	148.0
19-Oct-98	0952	7	65	357.6	584.0
20-Oct-98	0908	na	80	308.9	806.0
21-Oct-98	1003	7	70	119.4	216.0
22-Oct-98	0920	na	70	30.4	59.0
23-Oct-98	1015	5	85	na	17.0
24-Oct-98	0931	na	70	61.6	100.5
25-Oct-98	0847	na	na	64.2	na
26-Oct-98	0942	6	70	111.6	150.5
27-Oct-98	0858	na	na	103.2	na
28-Oct-98	0953	5	80	62.4	128.0
29-Oct-98	0909	na	65	51.4	86.5
30-Oct-98	1004	na	65	23.7	28.0
31-Oct-98	0920	na	na	15.0	na
28-Jan-00	0857	6	75	17.7	21.0
29-Jan-00	0951	7	85	23.4	79.0
30-Jan-00	0907	na	75	32.0	54.0
31-Jan-00	1002	na	75	14.1	51.5
01-Feb-00	0918	na	na	2.6	na
03-Feb-00	0929	na	65	13.3	35.0
05-Feb-00	0939	na	70	15.9	30.5
06-Feb-00	0855	na	na	4.2	na
07-Feb-00	0906	na	70	1.8	12.5
08-Feb-00	0906	na	na	2.3	na
10-Feb-00	0916	7	75	5.9	15.5
11-Feb-00	1011	na	na	6.5	na

06-Feb-01	0935	10	70	392.5	735.0
07-Feb-01	0849	6	70	51.4	85.0
08-Feb-01	0942	8	70	132.6	266.0
09-Feb-01	0856	9	65	60.3	111.0
10-Feb-01	0949	5	65	63.3	86.5
11-Feb-01	0903	4	70	15.5	31.5
12-Feb-01	0956	na	75	33.2	42.0
13-Feb-01	0911	5	85	17.8	57.5
14-Feb-01	1003	na	na	44.2	na
15-Feb-01	0918	6	65	25.4	45.5
16-Feb-01	1010	na	na	25.2	na
17-Feb-01	0925	na	65	10.5	26.5
19-Feb-01	0932	6	75	12.1	14.0
21-Feb-01	0939	na	na	9.7	na
22-Feb-01	0853	na	na	4.9	na
23-Feb-01	0946	na	na	8.9	na
24-Feb-01	0900	na	na	3.5	na
25-Feb-01	0954	4	70	9.6	19.5
28-Feb-01	0915	na	na	7.4	na
02-Mar-01	0922	7	65	12.0	19.5
06-Mar-01	0936	na	na	0.9	na
07-Mar-01	0850	6	65	7.0	17.5
25-Jul-02	0848	na	na	6.8	na
26-Jul-02	0938	5	85	162.1	343.0
27-Jul-02	0849	6	70	177.1	230.0
28-Jul-02	0939	10	80	315.1	762.0
29-Jul-02	0851	na	90	407.0	314.0
30-Jul-02	0940	5	85	36.2	90.0
31-Jul-02	0852	5	85	66.9	159.0
01-Aug-02	0943	na	85	71.0	249.0
02-Aug-02	0854	na	70	98.6	167.0
08-May-04	0856	4	70	48.1	88.5
10-May-04	0855	6	80	183.4	317.0
11-May-04	0943	5	85	108.7	196.0
12-May-04	0853	6	85	176.3	285.0
13-May-04	0942	na	na	106.7	na
14-May-04	0852	na	70	53.6	98.0
15-May-04	0941	na	85	211.1	133.0
16-May-04	0851	na	70	72.1	118.0
17-May-04	0940	na	85	60.4	137.5
18-May-04	0850	5	70	25.5	47.0
19-May-04	0938	na	85	22.2	75.5
20-May-04	0849	6	75	15.7	45.5

21-May-04	0937	4	85	69.2	34.5
22-May-04	0848	4	85	55.7	71.0
23-May-04	0936	6	75	38.4	85.0
24-May-04	0846	5	70	43.2	66.0
25-May-04	0935	na	70	38.7	91.5
26-May-04	0845	5	75	13.8	35.0
27-May-04	0933	na	na	35.9	na
28-May-04	0844	8	70	29.6	52.5
29-May-04	0932	4	65	27.1	50.0
30-May-04	0843	6	80	56.8	94.0
31-May-04	0931	7	75	22.3	101.5
03-Jun-04	0840	na	80	15.0	45.0
04-Jun-04	0928	na	70	37.6	66.0
06-Jun-04	0927	6	65	20.2	53.5
07-Jun-04	0838	4	65	7.9	16.0
08-Jun-04	0926	3	65	6.3	20.0
10-Jun-04	0925	na	na	3.6	na
12-Jun-04	0924	na	na	10.1	na
13-Jun-04	0834	6	65	2.7	18.0

¹approximate time of the sensor over the volcano

²above mean sea level, as determined by HYSPLIT trajectory modelling

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Table 1. TOMS sensors used in this study

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

Table 2. Summary of Nyamuragira SO₂ 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 (Mt)	Total SO ₂ Flux (Mt)
1980	2/1 – 2/13	13	Nimbus-7	5.4	66	0.58	0.88
1981	2/26 – 1/11/82	16	Nimbus-7	9.4	62	3.64	4.11
1984	2/24 – 3/4	10	Nimbus-7	3.7	64	0.75	1.00
1986	7/17 – 8/3	14	Nimbus-7	4.5	68	0.66	0.98
1987	12/31 – 1/1/88	2	Nimbus-7	2.0	85	0.02	0.03
1989	4/24 – 5/18	24	Nimbus-7	4.5	69	1.37	2.10
1991	9/22 – 6/27/92	32	Nimbus-7	2.8	71	0.41	0.85
1994	7/5 – 7/16	11	Meteor-3	3.8	74	1.12	1.47
1996	12/1 – 12/16	14	ADEOS	3.8	72	0.49	0.86
1998	10/17 – 10/31	15	Earth Probe	4.0	72	1.42	2.54
2000	1/28 – 2/11	12	Earth Probe	4.7	74	0.14	0.31
2001	2/6 – 3/7	22	Earth Probe	4.0	70	0.95	1.73
2002	7/25 – 8/2	9	Earth Probe	4.8	81	1.34	2.32
2004	5/8 – 6/13	31	Earth Probe	3.3	75	1.62	2.60
TOTALS		225		4.3	72	16.21	24.51

Table 3. Comparative summary of Nyamuragira SO₂ emissions

Nyamuragira activity (this work)

Largest emission, total eruption: 4.11 Mt (1981-82)

Largest emission, one-day total: 1.66 Mt (Dec. 27, 1981)

Largest average emission: 0.26 Mt/day over 9 days (2002)

Total emissions, 14 eruptions between 1979-2005: 24.51 Mt

Average annual emissions, 1979-2005: 0.94 Mt/yr

Global volcanic emissions: 13.4 Mt/yr¹

Total emission, single eruption event: 18 Mt, 1991 Mt. Pinatubo²

Passive degassing: 2.01 Mt/yr, 1987-1995 Mt. Etna³

¹Andres and Kasgnoc (1998); ²Guo *et al.* (2004); ³Bruno *et al.* (1999)

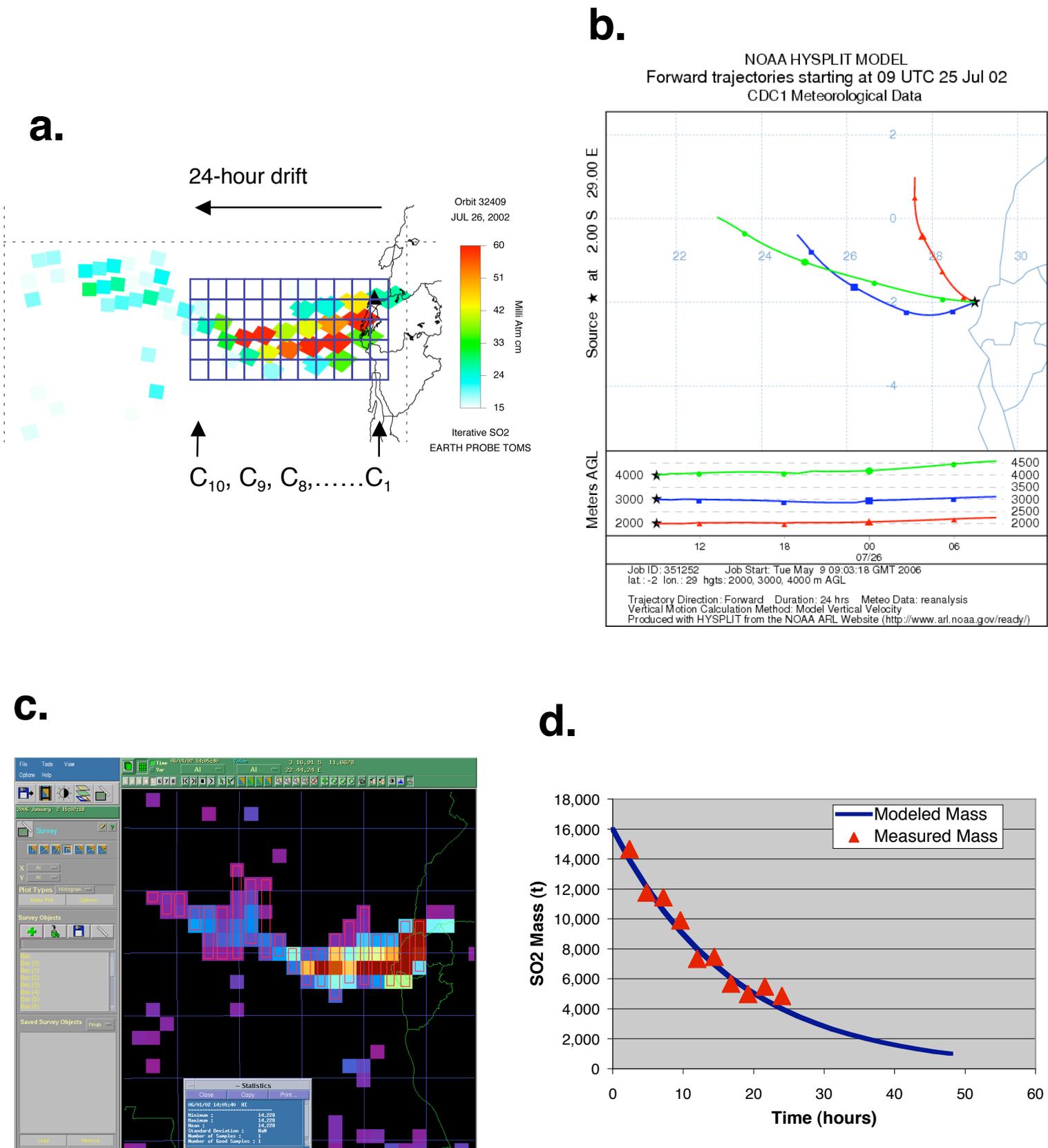


Figure 1

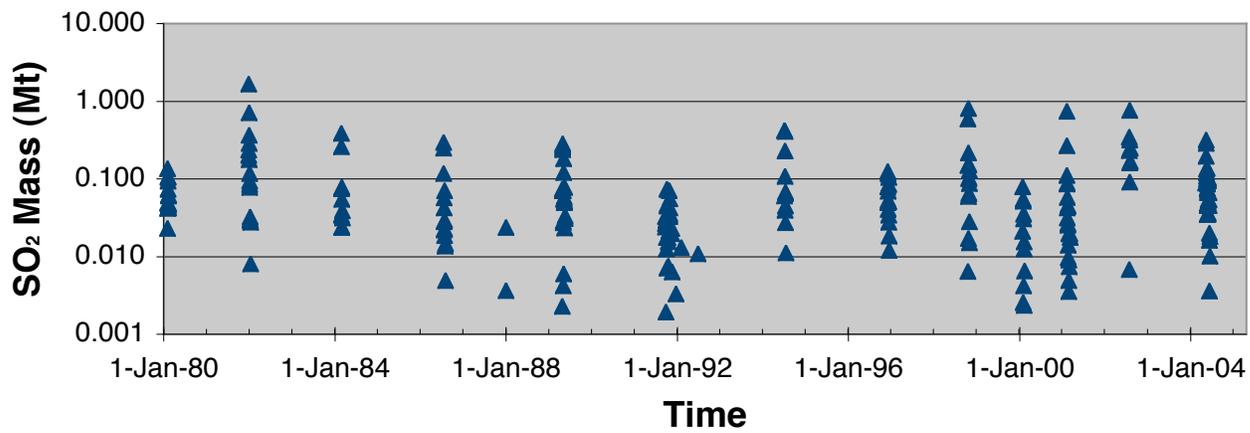
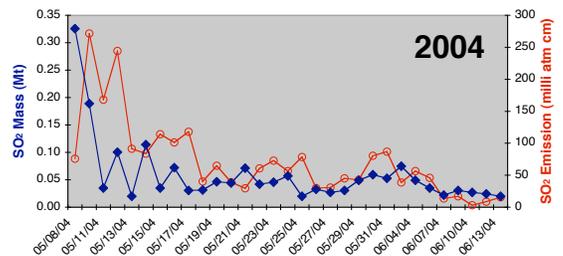
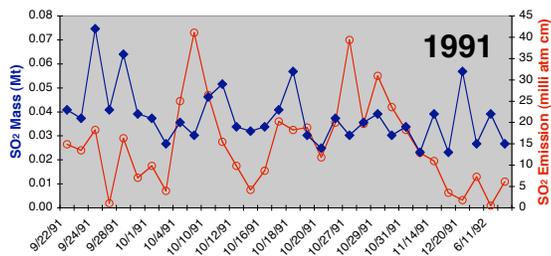
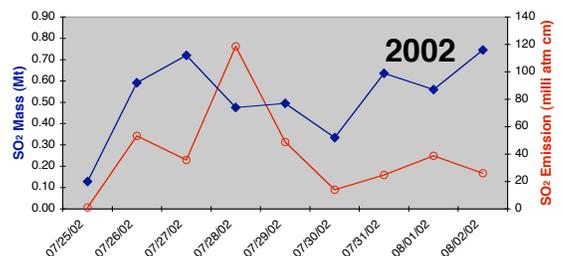
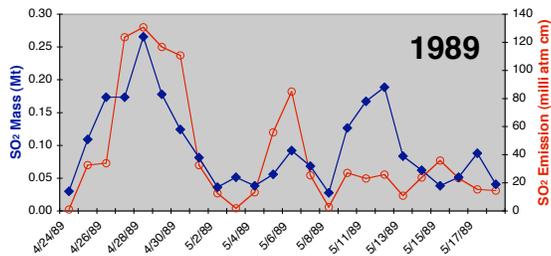
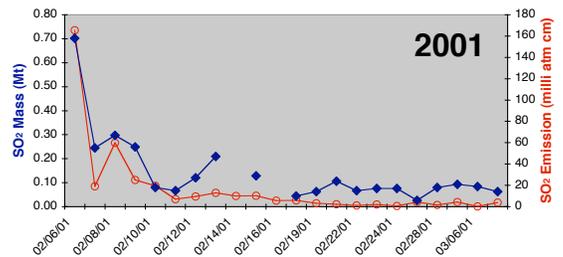
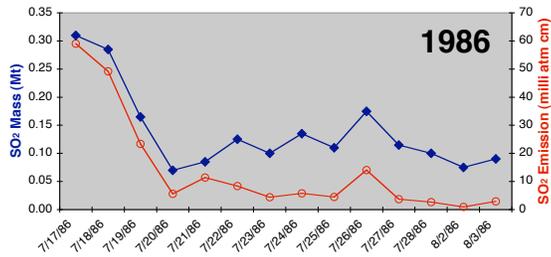
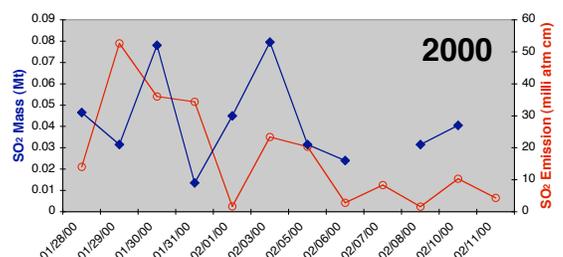
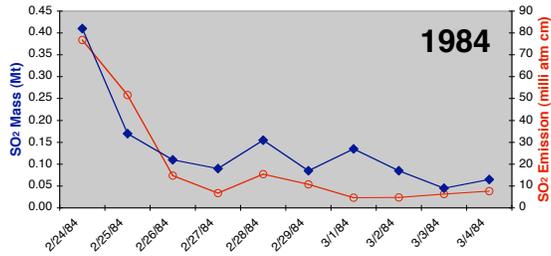
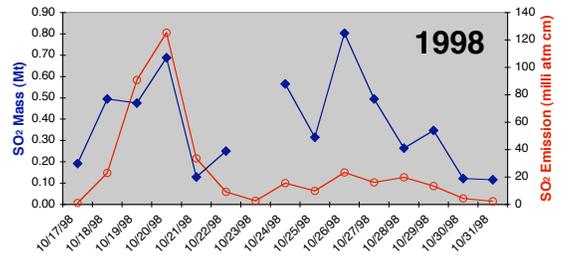
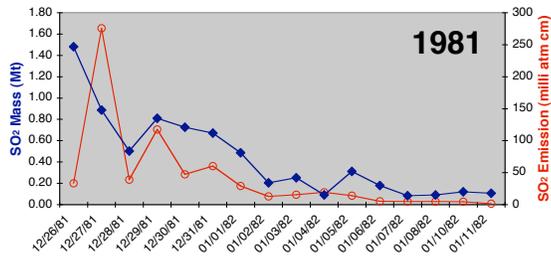
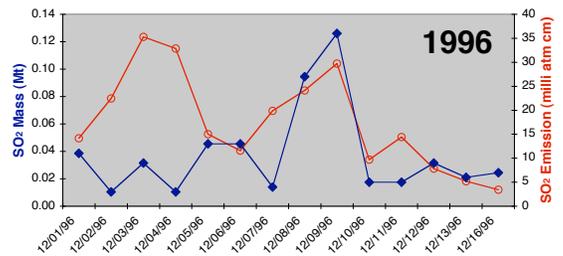
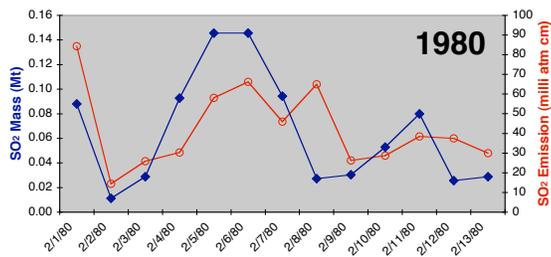


Figure 2



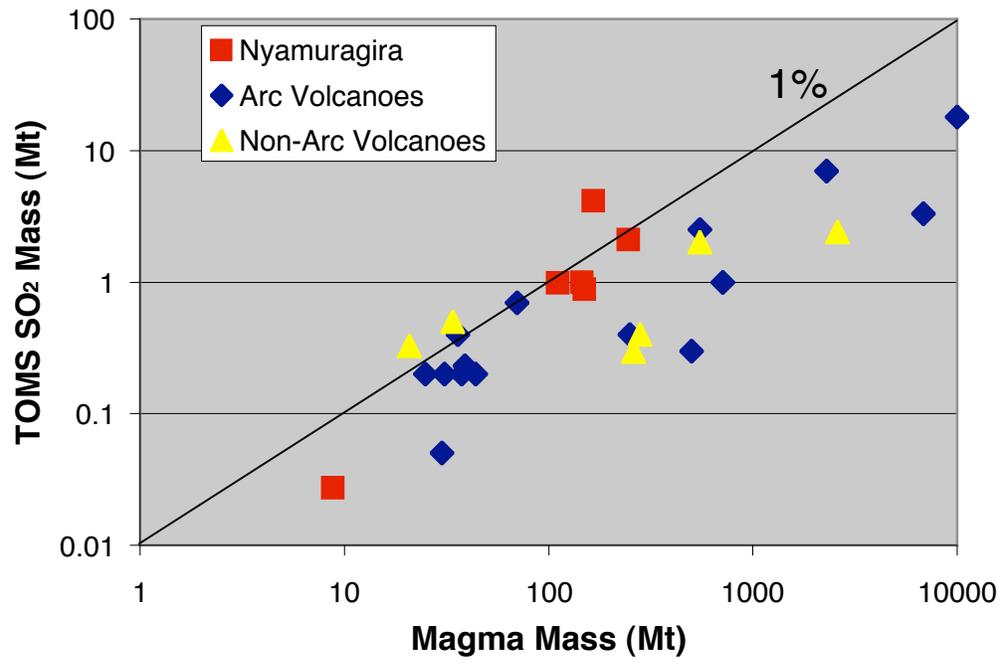


Figure 4

Figure Captions

Figure 1. Calculation of daily SO₂ 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 hours. (c) The TOMS SO₂ (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, and (d) the average SO₂ concentrations with distance are then calculated for each grid column (C₁, C₂, C₃...), or row, if for example the cloud drifted to the north or south. In this example, the drift distance is covered by 10 columns (1a); each column therefore corresponds to 2.4 hours emission time. Grid column masses are calculated from the average column concentrations multiplied by the total grid column areas. See text for details.

Figure 2. TOMS daily SO₂ 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 Mt of SO₂.

Figure 3. Daily SO₂ fluxes (solid blue diamonds) and “instantaneous” emission levels (open red circles) for the Nyamuragira eruptions, 1980-2004. The 1987-88 event is not shown, because there were only two days of observed activity. Instantaneous emissions represent the SO₂ 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 overage over the volcano. Both of these datasets give an indication of Nyamuragira’s activity levels over the course of each eruption period.

Figure 4. SO₂ 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/cm³, respectively. The 1% gas:magma mass ratio line is plotted for reference. Data sources: Blake (2003) and references within; Head (2006); Smithsonian GVN; this work.