

## Prodigious sulfur dioxide emissions from Nyamuragira volcano, D.R. Congo

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[1] Nyamuragira (Virunga chain, D.R. Congo) is among Africa's most active volcanoes, but direct observations of its eruptions are rare. From 1978–2002 the Total Ozone Mapping Spectrometer (TOMS) instruments measured SO<sub>2</sub> emissions during each of Nyamuragira's 13 most recent eruptions. Due to continuous co-eruptive SO<sub>2</sub> degassing, estimating the total atmospheric SO<sub>2</sub> loading resulting from eruptions of Nyamuragira is not straightforward. Using TOMS SO<sub>2</sub> plume cross-sections and model wind profiles we derive eruptive SO<sub>2</sub> fluxes and SO<sub>2</sub> loss rates for emissions on 19 October 1998 and 6 February 2001, giving peak SO<sub>2</sub> fluxes of 0.21 and 0.74 Tg day<sup>-1</sup> respectively and loss rates of  $\sim 10^{-6}$ – $10^{-5}$  s<sup>-1</sup>. Based on current data, time-averaged SO<sub>2</sub> emission rates at Nyamuragira are comparable to persistent emitters (Etna, Kilauea), but eruptive fluxes at Nyamuragira are much higher. Considering the alkaline composition of Virunga lavas, eruptive CO<sub>2</sub> fluxes could be seven times the SO<sub>2</sub> flux. **INDEX TERMS:** 0370 Atmospheric Composition and Structure: Volcanic effects (8409); 8419 Volcanology: Eruption monitoring (7280); 8409 Volcanology: Atmospheric effects (0370); 8494 Volcanology: Instruments and techniques; 9305 Information Related to Geographic Region: Africa. **Citation:** Carn, S. A., and G. J. S. Bluth, Prodigious sulfur dioxide emissions from Nyamuragira volcano, D.R. Congo, *Geophys. Res. Lett.*, 30(23), 2211, doi:10.1029/2003GL018465, 2003.

### 1. Introduction

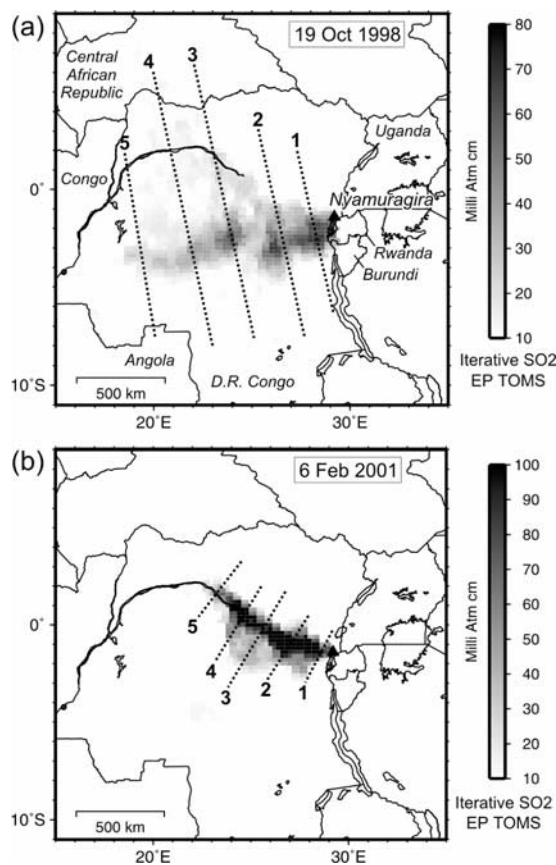
[2] Global volcanic SO<sub>2</sub> degassing (13–28 Tg yr<sup>-1</sup>;  $\sim 6$ –14% of SO<sub>2</sub> from all sources) [Bluth *et al.*, 1993; Graf *et al.*, 1997; Andres and Kasgnoc, 1998] is subordinate to anthropogenic SO<sub>2</sub> emissions (134 Tg yr<sup>-1</sup>), but differences in source distributions and higher emission altitude result in a disproportionately large volcanic contribution to the global atmospheric SO<sub>4</sub><sup>2-</sup> burden (36%) [Graf *et al.*, 1997]. Quantification of the magnitude and variability of volcanic SO<sub>2</sub> output therefore plays a key role in assessing the natural radiative forcing of climate due to atmospheric sulfate aerosol. In particular, whilst the flux of volcanic SO<sub>2</sub> to the stratosphere is well established by ultraviolet (UV) Total Ozone Mapping Spectrometer (TOMS) satellite data

[e.g., Bluth *et al.*, 1993; Carn *et al.*, 2003], emissions in the free troposphere are poorly constrained.

[3] Here we document exceptional tropospheric SO<sub>2</sub> emissions from Nyamuragira (1.41°S, 29.2°E, altitude 3058 m), a K-basaltic (potassic) shield volcano situated in the Virunga region of the Western Rift Valley in eastern Democratic Republic of Congo (D.R.C.). It is one of Africa's most active volcanoes, with  $\sim 40$  recorded eruptions since 1865 and 13 between 1980 and 2002 [Simkin and Siebert, 1994; recent eruption reports are available at [http://www.volcano.si.edu/gvp/world/region02/africa\\_c/nyamura/var.htm](http://www.volcano.si.edu/gvp/world/region02/africa_c/nyamura/var.htm)]. The 1978–2003 TOMS volcanic SO<sub>2</sub> database covering Nyamuragira's 13 most recent eruptions reveal it as the most prolific point source of sulfur emissions in Africa, and its cumulative total ( $\sim 9$  Tg) is only surpassed by the  $\sim 20$  Tg released by Pinatubo in 1991 [Bluth and Oppenheimer, 2001].

[4] Nyamuragira's SO<sub>2</sub> emissions are significant not only from a volcanological standpoint, but also as a prodigious natural supply of SO<sub>2</sub> and sulfate aerosol in a region where anthropogenic and oceanic sources are scarce. Furthermore its 1981–82 eruption, which produced  $\sim 3$  Tg of SO<sub>2</sub> in clouds that reached the tropopause [Krueger *et al.*, 1996], resulted in a sulfate aerosol loading deemed responsible for a cooling signal in climate records [Mao and Robock, 1998]. Recent investigations of lidar data from Mauna Loa also indicate a contribution to the variability in the background stratospheric aerosol from Nyamuragira's eruptions [W.B. Grant, pers. comm., 2003]. Quantification of its SO<sub>2</sub> output is therefore necessary to understand the potential environmental, atmospheric and climatic impacts of these emissions.

[5] Eruptions of Nyamuragira involve effusion of lava flows with concomitant near-continuous SO<sub>2</sub> emissions. Emitted gases are lofted to upper tropospheric altitudes by the intense heat over erupting fissure vents and fire fountains [e.g., Stothers *et al.*, 1986], analogous to the production of stratospheric aerosols by forest fires [Fromm *et al.*, 2000]. Determining the atmospheric SO<sub>2</sub> loading using polar-orbiting TOMS data, available once per day at low latitudes, is more complicated than for an explosive eruption [e.g., Krueger *et al.*, 1996]. Data gaps that occur intermittently in equatorial Earth Probe (EP) TOMS data increase uncertainty by obscuring swathes of volcanic clouds. A procedure for determining the SO<sub>2</sub> flux at the source is therefore desirable.



**Figure 1.** Column  $\text{SO}_2$  (milli atm cm or Dobson Units [DU]) from EP TOMS for Nyamuragira emissions on (a) 19 October 1998 at 0953 UT, and (b) 6 February 2001 at 0935 UT. TOMS data were interpolated onto a  $0.2 \times 0.2^\circ$  grid to create these maps. Plume transects used for  $\text{SO}_2$  flux analysis are indicated.

[6] In this paper we test a method of determining  $\text{SO}_2$  production from satellite data using a technique used to reduce field-based  $\text{SO}_2$  column measurements (e.g., via correlation spectroscopy or COSPEC), i.e., by calculating the product of plume cross-section and plume velocity, assumed to equal the wind speed at the plume altitude [Stoiber *et al.*, 1983]. We derive  $\text{SO}_2$  fluxes for plumes produced by eruptions of Nyamuragira that began on 17 October 1998 [GVN, 1998] and 6 February 2001 [GVN, 2001a], and compare the results to  $\text{SO}_2$  loadings determined using the TOMS plume ‘snapshot’ mass calculation described by Krueger *et al.* [1995], and to  $\text{SO}_2$  fluxes from other major volcanic sources.

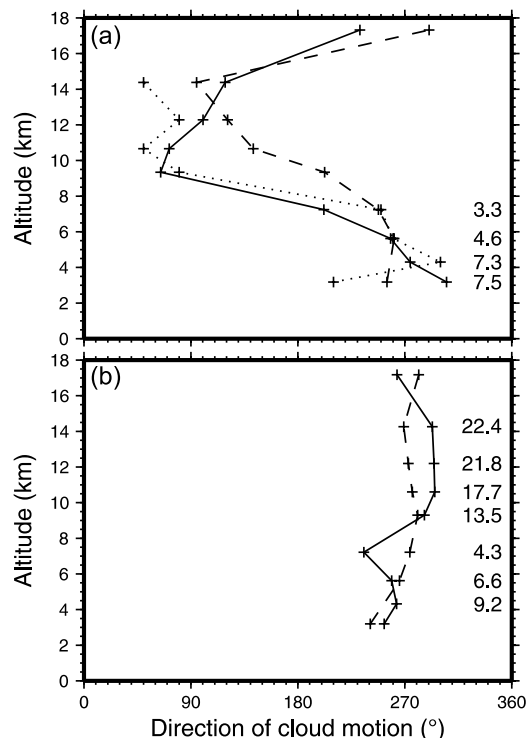
## 2. Plume Analysis

[7] The EP TOMS data used for this analysis were collected over Nyamuragira on 19 October 1998 and 6 February 2001. EP TOMS  $\text{SO}_2$  maps of the volcanic clouds are shown in Figure 1. The  $\text{SO}_2$  data were generated using an iterative 4-band inversion technique [Krueger *et al.*, 1995] assuming a mid- to upper-tropospheric cloud height.

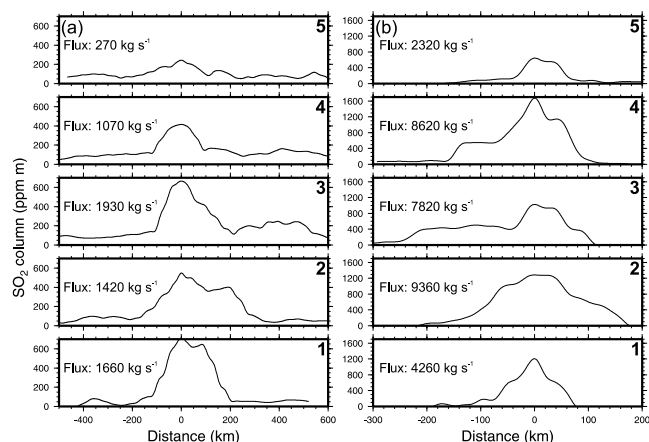
[8] We mimic the COSPEC  $\text{SO}_2$  flux method using upper air wind profiles to estimate plume velocity (Figure 2) and plume transects from TOMS (Figure 3). Radiosonde data

are sparse across central Africa, hence wind profiles over Nyamuragira (Figure 2) were derived from two meteorological analysis datasets, the NCEP/NCAR Reanalysis (NNR) product [Kalnay *et al.*, 1996] and the UARS/UK Meteorological Office (UKMO) Assimilation product [Swinbank and O’Neill, 1994], interpolated to the location of the volcano. The NNR data produced the best fit with the observed plume distributions and with the closest available radiosonde data (from Nairobi, Kenya;  $\sim 840$  km E of Nyamuragira; Figure 2). For both the 1998 and 2001 plumes, radiosonde data from Nairobi gave a thermal tropopause height of 16–17 km.

[9] We are unable to assess the contribution to each TOMS  $\text{SO}_2$  column from gas at different altitudes and hence some assumptions are required. For each plume segment we assume that all the measured  $\text{SO}_2$  is moving in the same direction with the same velocity, with the direction corresponding to the azimuth from Nyamuragira and the velocity obtained from the wind profiles at the altitude matching the direction of cloud motion (Figure 2). We also assume that all emitted  $\text{SO}_2$  remains above the summit height of Nyamuragira ( $\sim 3$  km) when assessing cloud motion and velocity. To calculate  $\text{SO}_2$  fluxes for each plume transect, an azimuth was calculated for each 1-km profile segment and used to correct for obliquity to the transect and to obtain a plume velocity from the profiles in



**Figure 2.** Model wind profiles and cloud motion (North =  $0^\circ$ ) for Nyamuragira on (a) 19 October 1998 at 0953 UT, and (b) 6 February 2001 at 0935 UT. Data are derived from NCEP/NCAR Reanalyses (solid line) and the UARS/UKMO Assimilation product (dashed line). Model-derived wind speeds ( $\text{m s}^{-1}$ ) at selected altitudes (used in  $\text{SO}_2$  flux calculations) are plotted to the right of the profiles. Also shown in (a) are radiosonde data from Nairobi on 19 October 1998 at 1100 UT (dotted line).



**Figure 3.** SO<sub>2</sub> plume cross-sections (in ppm m; assuming 1 DU = 10 ppm m) and SO<sub>2</sub> fluxes for (a) 19 October 1998, and (b) 6 February 2001. Profiles are numbered with reference to the transects in Figure 1. The smoothed profiles were generated by sampling the data in Figure 1 at 1 km intervals along each transect using bicubic interpolation. Plume cross-sections were evaluated by integrating under the profiles using an appropriate baseline to screen out EP TOMS background noise (typically  $\leq 100$  ppm m).

Figure 2. The products of plume cross-section and plume velocity for each segment were then summed to obtain SO<sub>2</sub> flux.

[10] The SO<sub>2</sub> cloud detected on 19 October 1998 (Figure 1) was one of the largest mapped during the 1998 eruption. Wind profiles indicate that the bulk of the plume resided at altitudes of 3–8 km (Figure 2). The 6 February 2001 cloud was detected  $\sim 11$  hours after an earthquake swarm at 2232 UTC on 5 February that heralded the start of the 2001 eruption [GVN, 2001a], and we assume that SO<sub>2</sub> emission began no earlier than this. Using this constraint, the minimum plume velocity derived from the TOMS image is  $\sim 20$  m s<sup>-1</sup> with an azimuth of 290–300°, in good agreement with the NNR wind profile at 10–14 km altitude (Figure 2).

[11] Cloud altitude and underlying surface reflectivity affect tropospheric SO<sub>2</sub> retrievals in the UV [Krueger *et al.*, 1995]. Average reflectivities for the 19 October 1998 and 6 February 2001 EP TOMS scenes were  $\sim 33\%$  and  $50\%$ , respectively, due to meteorological cloud though with considerable variability over each SO<sub>2</sub> cloud region. For the assumed SO<sub>2</sub> cloud altitudes, this implies  $\sim 10$ – $30\%$  and

20–40% overestimation of SO<sub>2</sub>, respectively [Krueger *et al.*, 1995], with the largest excess (i.e., highest reflectivity) in transect 3 on 19 October 1998 (Figures 1a and 3a) and transects 3 and 4 on 6 February 2001 (Figures 1b and 3b). Since eruptions of Nyamuragira do not typically produce large amounts of fine ash that can sequester SO<sub>2</sub> through adsorption processes [e.g., Rose, 1977] we expect minimal SO<sub>2</sub> loss via this route, and negligible ash effects on TOMS SO<sub>2</sub> retrievals.

### 3. Results and Discussion

[12] Results of SO<sub>2</sub> flux calculations are shown in Figure 3 and summarized in Table 1, where they are compared with ‘snapshot’ loading estimates (hereafter, SSL = ‘snapshot’ SO<sub>2</sub> loading) with a typical 30% uncertainty. Also shown are the total cumulative SO<sub>2</sub> mass observed by EP TOMS and the estimated total SO<sub>2</sub> production (after Krueger *et al.* [1996]) for the 1998 and 2001 eruptions. Predicted peak SO<sub>2</sub> fluxes (i.e. source strengths) and SO<sub>2</sub> loss rates (Table 1) were derived by fitting exponential curves to plots of SO<sub>2</sub> flux against plume age [e.g., Oppenheimer *et al.*, 1998] with reasonable allowances for possible overestimation of SO<sub>2</sub>. ‘Average’ plume velocities were used to derive plume ages due to probable variations in plume height and hence velocity along each transect.

[13] Eruptive SO<sub>2</sub> fluxes from Nyamuragira are likely to vary in tandem with parameters such as lava effusion rate (which is unknown), and this is most apparent in the 2001 data. We therefore used a subset of the 6 February 2001 transects to derive a peak SO<sub>2</sub> emission rate ( $\sim 8570$  kg s<sup>-1</sup> or  $\sim 0.74$  Tg day<sup>-1</sup>) that may have been sustained for a minimum of  $\sim 4$  hours. The 19 October 1998 data show a more regular decrease in SO<sub>2</sub> mass with distance from the volcano, but due to the low density of transects we cannot rule out variable fluxes. Correspondence between peak SO<sub>2</sub> fluxes and SSL is best on 19 October 1998, when the SO<sub>2</sub> emissions evaluated using the two methods narrowly overlap within error (Table 1). The low peak SO<sub>2</sub> flux relative to the SSL, and the extensive plume (Figure 1a), probably reflect rather low SO<sub>2</sub> loss rates (Table 1). On 6 February 2001 the peak SO<sub>2</sub> flux is considerably larger than the total mass in the scene, but this peak flux only persisted for a few hours. Integrating estimated SO<sub>2</sub> fluxes over the life of the plume we obtain  $\sim 0.26$ – $0.33$  Tg SO<sub>2</sub>, which is roughly consistent with the SSL (Table 1).

[14] SO<sub>2</sub> loss rates for the two datasets are comparable, although the 19 October 1998 data indicate a lower mini-

**Table 1.** Summary of EP TOMS SO<sub>2</sub> Plume Analysis Results for Nyamuragira

	19 Oct 1998	6 Feb 2001
Peak SO <sub>2</sub> flux (Tg day <sup>-1</sup> )	$0.213 \pm 0.05^a$	$0.737 \pm 0.13^b$
Snapshot SO <sub>2</sub> loading (Tg)	$0.342 \pm 0.10$	$0.390 \pm 0.12$
SO <sub>2</sub> loss rate (s <sup>-1</sup> )	$3 - 9 \times 10^{-6a}$	$6 \times 10^{-6} - 1 \times 10^{-5b}$
Total eruption SO <sub>2</sub> (Tg) – sum of daily totals	$2.94 \pm 0.88^c$	$1.51 \pm 0.45^d$
Total eruption SO <sub>2</sub> (Tg) – Krueger <i>et al.</i> [1996] method	$1.39 \pm 0.42^c$	$0.958 \pm 0.29^f$

<sup>a</sup>Derived using transects 1–5 in Figure 3a.

<sup>b</sup>Derived using transects 2–4 in Figure 3b.

<sup>c</sup>SO<sub>2</sub> detected 17–31 Oct inclusive.

<sup>d</sup>SO<sub>2</sub> detected 6–17, 19, 21–26, 28 Feb, 2, 6–7 and 10 Mar inclusive.

<sup>e</sup>26% day<sup>-1</sup> loss rate used throughout.

<sup>f</sup>50% day<sup>-1</sup> loss rate used 6–12 Feb inclusive, 100% day<sup>-1</sup> used thereafter.



loss rate despite a lower cloud height (Table 1). In general the loss rates appear similar to or greater than those calculated for the 1981 Nyamuragira eruption, which is consistent with the higher altitude of the 1981 cloud [Krueger *et al.*, 1996]. Loss rates were input into the Krueger *et al.* [1996] model to estimate total SO<sub>2</sub> output for each eruption (Table 1). The Krueger *et al.* [1996] technique seeks the best fit between measurements of apparent 'new' SO<sub>2</sub> production (usually the SO<sub>2</sub> mass in the plume attached to the volcano) and production estimates derived from application of a reasonable daily loss rate to the total observed SO<sub>2</sub> mass on each day. A total SO<sub>2</sub> yield is then obtained by summing the new SO<sub>2</sub> amounts over the duration of the eruption. In this case actual durations for either eruption are not accurately known, but in 1998 the last EP TOMS observation of SO<sub>2</sub> from Nyamuragira occurred on 31 October and due to the paucity of local reports [GVN, 1998] this is the best indicator of the end of significant eruptive activity. In 2001 SO<sub>2</sub> was last observed on 10 March when cessation of lava flows was also reported [GVN, 2001b].

[15] Our results (Table 1) demonstrate the importance of constraining SO<sub>2</sub> output on days with the highest loadings as accurately as possible, since these represent a significant proportion (here at least ~25–41%) of the total production. For the cases presented here the SSL technique emulates calculations using SO<sub>2</sub> fluxes derived using reasonable assumptions of cloud height and motion to an acceptable degree, if short-term variability of SO<sub>2</sub> fluxes is taken into account. A better approach would be to derive SO<sub>2</sub> fluxes for each day of an eruption using a higher density of plume transects. However, the spatial resolution and noise level of EP TOMS data are not sufficient for accurate analysis of the small plumes emitted during the waning stages of Nyamuragira's eruptions. Such work will be more feasible when data from the Ozone Monitoring Instrument (OMI) become available in 2004 [Carn *et al.*, 2003].

#### 4. Tropospheric SO<sub>2</sub> Emissions From Basaltic Volcanoes

[16] Eruptive SO<sub>2</sub> flux measurements are rare but those determined here for Nyamuragira (Table 1) are clearly exceptional compared to existing data. Regular COSPEC measurements from 1975–2003 at Mt. Etna (Italy) show co-eruptive peaks of ~0.03 Tg day<sup>-1</sup> during paroxysms and a mean SO<sub>2</sub> output of ~1.8–2.0 Tg yr<sup>-1</sup> [Allard, 1997; Caltabiano *et al.*, 2003]. At Kilauea (Hawai'i) SO<sub>2</sub> fluxes recorded in the early stages of the Pu'u 'O'o-Kupaianaha eruption (1983–present) also reached a minimum of ~0.03 Tg day<sup>-1</sup>, and the 1979–1997 average release was ~0.5 Tg yr<sup>-1</sup> [Sutton *et al.*, 2001]. An accurate assessment of Nyamuragira's long-term sulfur budget is underway, but using the current TOMS inventory gives a mean SO<sub>2</sub> flux of ~0.4 Tg yr<sup>-1</sup> since 1978. However, we stress that this only accounts for eruptive emissions, and at present we lack constraints on whether Nyamuragira emits significant SO<sub>2</sub> between eruptions. Eruptive emissions attain 8.5–10.6 Tg of SO<sub>2</sub> per km<sup>3</sup> of erupted magma (compared to 5–9 Tg of SO<sub>2</sub> per km<sup>3</sup> magma erupted for Icelandic flood basalt events [Thordarson and Self, 2003]), so a high volume,

sustained eruption from the volcano could have a severe environmental impact.

#### 5. Conclusions

[17] Basaltic effusive eruptions produce vast quantities of tropospheric SO<sub>2</sub>, and dominate the eruptive SO<sub>2</sub> flux in periods lacking major explosive eruptions [e.g., Carn *et al.*, 2003]. Nyamuragira has been among the most prolific sources of volcanic SO<sub>2</sub> for at least the last 25 years. The chemical composition of eruptive gas has never been measured at Nyamuragira, but sampled gases from nearby Nyiragongo [Gerlach, 1980] suggest that CO<sub>2</sub> fluxes from Nyamuragira could be 7 times greater than the SO<sub>2</sub> flux. Considering the global volcanic CO<sub>2</sub> emission rate estimates of Williams *et al.* [1992] and our SO<sub>2</sub> fluxes from this study, peak Nyamuragira emissions represent ~10–70% of the annual eruptive CO<sub>2</sub> flux or ~5–30% of the total annual volcanic CO<sub>2</sub> output, and as much as ~10% of an average daily anthropogenic CO<sub>2</sub> flux. However, we note that volcanic CO<sub>2</sub> flux estimates [e.g., Williams *et al.*, 1992] are subject to considerable uncertainty and do not include the full contribution of Nyamuragira's emissions.

[18] Quantification of SO<sub>2</sub> degassed by effusive eruptions using polar-orbiting instruments will always pose problems due to inadequate temporal sampling of the plume. This will also apply to passive volcanic plumes detected from space by instruments more sensitive than TOMS. Use of the techniques documented here with UV OMI data from 2004, in conjunction with geostationary data from the Spinning Enhanced Visible and Infrared Imager on the Meteorat Second Generation platform, which images Africa every 15 minutes, should improve estimates of SO<sub>2</sub> degassing from African volcanoes.

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