Long-term monitoring of SO_2 quiescent degassing from Nyiragongo's lava lake

Arellano S.[1]*, Yalire M.[2], Galle B.[1], Bobrowski N.[3], Dingwell A.[4], Johansson M.[1], Norman P.[1]

[1] Department of Earth and Space Sciences, Chalmers University of Technology, Gothenburg, Sweden

[2] Observatoire Volcanologique de Goma, Centre de Recherche en Sciences Naturelles, Lwiro, République Démocratique du Congo

[3] Institute of Environmental Physics, Heidelberg University, Heidelberg, Germany

[4] Department of Earth Sciences, Uppsala University, Uppsala, Sweden

*Corresponding author: santiago.arellano@chalmers.se

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Abstract

The activity of open-vent volcanoes with an active lava-lake, such as Nyiragongo, is characterized by persistent degassing, thus continuous monitoring of the rate, volume and fate of their gas emissions is of great importance to understand their geophysical state and their potential impact. We report results of SO_2 emission measurements from Nyiragongo conducted between 2004 and 2012 with a network of ground-based scanning-DOAS (Differential Optical Absorption Spectroscopy) remote sensors. The mean SO_2 emission rate is found to be 13 ± 9 kg s^{-1}, similar to that observed in 1959. Daily emission rate has a distribution close to log-normal and presents large inter-day variability, reflecting the dynamics of percolation of magma batches of heterogeneous size distribution and changes in the effective permeability of the lava lake. The degassed S content is found to be between 1000 and 2000 ppm from these measurements and the reported magma flow rates sustaining the lava lake. The inter-annual trend and plume height statistics indicate stability of a quiescently degassing lava lake during the period of study.

1. Introduction

The segregation and release of magmatic volatiles as gases into the atmosphere is a fundamental process of active volcanism. The total emission or magnitude as well as the rate of emission or intensity are often related to the volume of magma at shallow levels and its rate of ascent, which in turn define the style, explosiveness and duration of volcanic eruptions (Galle *et al.*, 2010, Parfitt & Wilson, 2008, Sparks, 2003). In particular, volcanic degassing is the defining process of volcanoes with permanent quiescent activity, where other geophysical signals, indirectly linked to the presence or ascent of magma, such as seismicity or ground deformation, are usually less pronounced. This underlines the importance of long-term monitoring of volcanic degassing, but its actual implementation has proven to be technologically or logistically challenging, leaving operational gas monitoring behind seismic or geodetic methods in most volcano observatories until recently.

A qualitative change in the implementation of gas monitoring on volcanoes resulted from the implementation of modern developments giving sensitive and fast multichannel array detectors, advances in computers, and algorithms for the analysis of differential optical absorption spectroscopy (DOAS). The first flux measurements of volcanic gas emissions using a miniaturized spectrometer were made in Nicaragua on April 2001 (Galle *et al.*, 2002). Subsequently, the first time-resolved measurements with an automatic scanning miniaturized-DOAS instrument were initiated on Montserrat in January 2002 (Edmonds, 2003), followed by similar implementations in Congo, Italy, Ecuador, Hawaii, Japan and New Zealand. Subsequently, a growing network of scanning instruments has been built up since 2005 (Galle *et al.*, 2010).

1.1 Nyiragongo volcano

An archetypical case of a permanent degassing volcano which due to its associated risk demands continuous monitoring is Nyiragongo (Lat. 1.52°S, Lon. 29.25°E, Alt. 3470 m). An active alkaline-basaltic stratovolcano located in the western branch of the East African Rift, Nyiragongo is situated just about 15 km N of the city of Goma (~1 million inhabitants) in the Lake Kivu province of the Democratic Republic of Congo, close to the border with Rwanda. Nyiragongo and its neighbor, the basaltic shield volcano Nyamuragira (1.41° S, 29.20°E, Alt. 3058 m), are the only presently active volcanoes of the Virunga Volcanic Province (see Figure 1). Morphologically, Nyiragongo presents a relief of about 2000 m and a 1-2 km wide open crater hosting an active lava lake since at least 1928 (at a level of about 1500 m above the altitude of Goma and with a typical area of 200×300 m^2), which has drained out catastrophically during historical times (Burgi *et al.*, 2014). The most recent and better studied cases occurred in 1977 (Tazieff, 1977) and 2002 (Komorowski *et al.*, 2003, Tedesco *et al.*, 2007). In the latter case, a fissure eruption produced about $14\text{-}34\times10^6$ m^3 of lava that drain down the flanks and destroyed about 15% of the city of Goma, causing at least 200 fatalities and leaving ~250 000 people homeless (Burgi *et al.*, 2014, Tedesco *et al.*, 2007). Inside the crater,

three different terraces indicate levels of the lava lake today as well as before the eruptions of 1977 and 2002. The present level lies at about 400 m below the summit, and fluctuations of a few m occur within a few minutes. The geometry of the upper part of the lava lake is assumed to be that of an inverted truncated cone with an upper diameter of ~200-300 m, a lower diameter of ~50-120 m and a repose angle of ~55-80°. The conduit connecting the lower part of this crater with the reservoir is thought to be ~15 m diameter and a depth of 1-4 km (Burgi *et al.*, 2014).
Nyiragongo's magma composition is mafic, with leucite-bearing nephelinites and melilitites, enriched in Na_2O and K_2O and under-saturated in SiO_2 (38-40%). This composition determines a relatively low magma viscosity, which facilitates gas segregation and fast transport of lavas, in the case of an outflow.

Recent activity has been mostly localized in the central vent, but has also migrated to the complex system of lateral fissures. The onset of major outbursts seems to be controlled by regional tectonic stress of the rift system, although phreato-magmatism has also been linked to the initiation of major eruptions (Komorowski *et al.*, 2003).

Besides the hazard of fast-moving (up to 60 km h^{-1}) lava flows caused during major eruptions, the most important risk of this volcano is associated with the permanent emission of gases such as CO_2 from the flanks and surroundings of the volcano. Being the density of CO_2 higher than that of the ambient air, this gas tends to accumulate in depressions reaching concentrations higher that 10%vol, which are lethal. These zones are known as "mazukus", the Swahili term for "evil winds" (Smets *et al.*, 2010).

1.2 Monitoring network

To assess the risk of this volcano a number of local and international projects have been carried out to support the surveillance in charge of the Goma Volcanological Observatory. The present monitoring capabilities of this volcano include a network of seismic sensors, tiltmeters, GPS sensors and InSAR imagery, temperature probes, in-situ gas analyzers, geochemical analysis of hydrothermal samples, and occasional petrological analyses of collected rocks. The surrounding area has been properly mapped for hazards and dissemination of the activity and prevention has taken place among the population (Mavonga *et al.*, 2010). A major difficulty for long term monitoring; however, is the agitated political situation in the region, which has sometimes resulted in the impossibility for installing, maintaining or operating the geophysical sensors. Under such circumstances, remote sensing acquires additional significance.

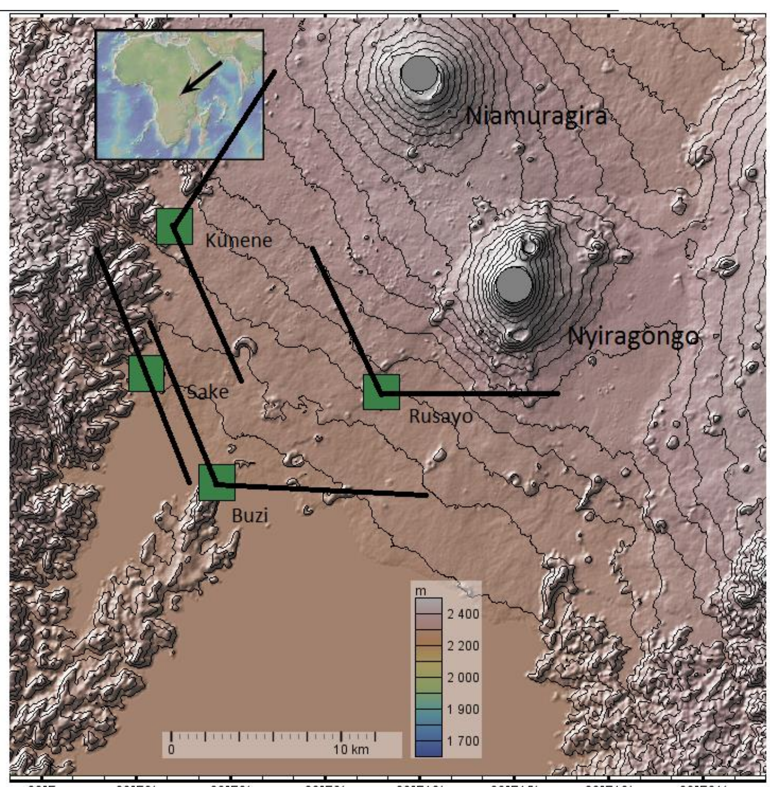


Figure 1. Map of the gas monitoring network (green squares) around Nyiragongo (and Nyamuragira) volcano. The black lines represent the angular coverage and orientation of the scanners (base map from GeoMap.org)

1.3 Degassing measurements and mechanism

The current activity of Nyiragongo is characterized by variations in the level and convection intensity of the lava lake, which is associated with a persistent, and occasionally prodigious, open vent degassing (Carn, 2004). During the 2002 eruption, it reached emission rates of about 10 Gg d^{-1}, whereas in the 1972 eruption it was estimated to have reached up to 23 Gg d^{-1} (Le Guern *et al.*, 1988). The low viscosity of the low-Si content Nyiragongo's lava facilitates a highly dynamical convection thought to be driven by a degassing-crystallization-densification mechanism, as suggested by various authors (Burgi *et al.*, 2014, Le Guern, 1987, Sawyer *et al.*, 2008, Tazieff, 1994).

There is compelling evidence in the literature regarding convection in the conduit as the main mechanism for steady-state degassing at this volcano. Succinctly, this mechanism consists in the equilibrium (i.e., closed respect to degassing) ascent of volatile-rich, vesiculated batches of magma through the conduit. This is followed by outgassing at a certain depth, which is determined by the amount of volatiles, their solubility and bulk permeability of the magma column. As a consequence, there is a reduction of vesiculation leading to an increase in density and sinking (down the conduit) of the degassed magma, which is then replaced by another batch of ascending lower density magma. The degassed magma might accumulate as a plutonic rock (Arellano *et al.*, 2008, Kazahaya *et al.*, 1994, Palma *et al.*, 2011, Stevenson & Blake, 1998). This process is sustained as long as there is enough supply of volatile-rich magma from depth, which in the case of Nyiragongo, seems to be related to a deeper mantle source than the neighboring Nyamuragira (Chakrabarti *et al.*, 2009a, Chakrabarti *et al.*, 2009b). Steady-state convection is a function of the amount of volatiles, the viscosity and geometry of the magmatic system; hence the degassing rate is a key parameter to monitor, especially because other disturbances (seismicity, deformation) are indirect and less pronounced in this volcano, and because understanding of the activity, although seemingly well characterized by the convection model, remains highly qualitative without actual measurements to define its magnitude.

Past measurements (Galle *et al.*, 2005, Le Guern, 1982, Sawyer *et al.*, 2008) of Nyiragongo's plume composition indicate interesting changes over time. The relative volumetric concentrations measured in 1959 and 1972 indicated contents of about 45-55% H_2O, 35-50% CO_2, 1-2% SO_2, 2-3% CO, 1.5-2.5 H_2S, and <2% for H_2, S_2, HCl, HF, and COS (Gerlach, 1980). After the 2002 eruption, the estimated concentrations were of about 68-72% H_2O, 22-26% CO_2, 4-5% SO_2 and <1% for other species (Sawyer *et al.*, 2008). Conceding that methodological differences between direct sampling (prior to 1977) and remote sensing measurements are properly accounted for, it is not clear which is the cause of these differences, either an actual change in the composition of the magma after the 1977 eruption or the progressive depletion of CO_2 caused by long-term degassing of the same source of magma feeding the lava lake.

Regarding gas emission rates, SO_2 has been the species focus of monitoring by ground-based and satellite-based instruments. The record of satellite measurements for this volcano dates back to the era of Total Ozone Mapping Spectrometer, and continued principally with the Ozone Mapping Spectrometer (OMI) and the Ozone Mapping and Profiler Suite (OMPS) (Carn *et al.*, 2003), as well as other UV sensors, such as the Global Ozone Monitoring Experiment-2 (GOME-2), or IR sensors, such as the Infrared Atmospheric Sounding Interferometer (IASI) or the Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER). This dataset is particularly valuable for the study of large plumes from Nyiragongo, as during the 2002 eruption and its aftermath (Carn, 2004). However, the spatial and/or temporal resolution and sensitivity profile of satellite sensors are insufficient for detecting low altitude quiescent plumes with low burdens (~<1 kt) of SO_2, which dominate the activity of this volcano.

In this article, we present the record of ground-based SO_2 emission rate measurements conducted at Nyiragongo volcano during the period 2004-2012 with the scanning-DOAS technique. After presenting details of the method and the resulting measurements, we discuss the implications for the long-term degassing of the lava lake, its trends and magnitude, as well as the fate of the volcanic plume to assess the impact of the emissions.

2. Methods

2.1 Scanning DOAS network

Our measurements of the SO_2 emission rate from Nyiragongo started in 2003. The first permanent installation of a scanning-DOAS system was done in March 2004 at Rusayo, where a seismic station operated by the GVO was in place. Within the EU-project NOVAC (Network for Observation of Volcanic and Atmospheric Change) (Galle *et al.*, 2010), three further instruments were installed at the sites Sake, Kunene and Buzi (see Figure 1) However, Buzi was soon after put out of operation and did not produce valid measurements, unfortunately. The configuration of this network was determined based on the criteria of maximizing the chances of capturing the plume of Nyiragongo (i.e., considering the prevalent wind patterns in the area), while respecting accessibility, security and the existence of a telemetric grid to transmit data in real time to the observatory (Buzi hosts a repeater station). Assuring the permanent operation of the network has not always been possible, due to frequent social unrest that has resulted in vandalism or in the unfeasibility of conducting proper maintenance of the stations. Therefore several gaps exist in the dataset and at least one visit to the sites every year has been necessary to recover and maintain the stations.

The scanning-DOAS method is well known and the interested reader is referred to the extensive literature for details (Bobrowski *et al.*, 2003, Galle *et al.*, 2010, Galle *et al.*, 2002, Williams-Jones *et al.*, 2008). In short, an automatic scanning system acquires spectra of scattered solar UV radiation over either a flat or conical scanning surface. These spectra are taken at Nyiragongo at angular steps of 3.6 deg, requiring therefore 51 steps to complete one scan. Each spectrum is evaluated by DOAS to get the SO_2 column density (number of absorbing molecules per unit area) relative to the background column of the gas (Platt & Stutz, 2008). If the volcanic plume is intercepted by the scanning path of the instrument, its total gas content can be obtained by integration of the column densities on the scan (which involves a conversion from slant to vertical column densities by a geometrical air-mass-factor correction), and the angular position of the centre of mass of the plume for each instrument can be determined. Finally, the gas flux is calculated by multiplying the line-integrated column densities with plume height (typically calculated from triangulation of quasi-simultaneous observation of the plume by two scanners) and plume speed (typically assumed to be equal to the wind speed at plume altitude). This method lies on the principle of conservation of mass in the volume defined by the scanning surfaces enclosing the volcano, for within this volume, the flux across the surface equals the emission rate or source strength of the volcano (in kg s^{-1}), if other possible sources (e.g., other volcanoes), sinks (e.g., chemical reactions, scavenging by adsorption or dilution), or accumulation (e.g., deposition) in the volume can be neglected. The plume transport speed normal to the scanned surface has to be determined by independent methods, although, under certain conditions, it can also be measured by autocorrelation of two time series of gas columns obtained by simultaneous measurements at two pointing directions along the plume axis (Johansson *et al.*, 2009). In this study, the source of plume speed that was systematically used corresponds to analyzed observations of the wind speed at crater altitude provided by the European Center of Medium-range Weather Forecasts (ECMWF), which has a temporal resolution of 6 h.

During the period April 2010-March 2011, a more detailed study of the local meteorology was conducted (Dingwell *et al.*, 2016). Meteorological data with 0.75 deg resolution from the ERA reanalysis product was used as an input for the Weather Research and Forecasting (WRF) model. This model was used, to downscale from 54 to 2 km spatial resolution in 4 steps, thus taking into account topographical effects on the wind-fields. As a result, wind profiles above the crater were produced at altitudes of 3200, 3300, 3400, 3500, 3750, 4000, 4250, 4500, 4750, and 5000 m a.s.l. for every hour. These profiles were then compared with the angular position of the measured columns to iteratively find the altitude at which the meteorological model and the measurement give a similar wind direction. If such convergence is found, the altitude, wind direction and corresponding wind speed are simultaneously determined. This was necessary because most of the time there were no two stations in operation to produce plume localization by triangulation. Thus the mean altitude from the statistics of this 1 year period was used as default value for the flux calculations of the other periods.

All stations have been subject to change, either due to technical problems or to improvements in the scanning geometry (flat scanning at 90 deg or conical scanning at 60 deg). Conical scanning refers to a surface of scanning defining a semi-cone open towards the volcano. This geometry allows a larger coverage of the plume and less sensitivity to geometrical errors in the emission rate calculation (Johansson, 2009). The different configurations set at this volcano are described in Table 1.

Table 1. Configurations of the scanning-DOAS instrumental network at Nyiragongo volcano

	Rusayo		**Sake**	**Kunene**	**Buzi**
Serial Number	D2J1840	D2J2133	D2J1840	D2J2081	I2J8554
Time from (yyyy/mm/dd)	2004-03-06	2007-07-06	2007-06-27	2007-06-27	2007-07-06
Time to (yyyy/mm/dd)	2007/06/01	Time of writting	Time of writting	Time of writting	Time of writting
Latitude (UTM)	-1.576987	-1.576987	-1.567100	-1.489283	-1.625033

Longitude (UTM)	29.179893	29.179893	29.055583	29.070400	29.09305
Altitude (m a.s.l)	1688	1688	1509	1807	1698
Azimuth (deg)	106	52	78	100	56
Scanning angle (deg)	90	60	90	60	60
Number of valid measurements (this study)	7878	66090	6440	656	0

The NOVAC scanning-DOAS instrument used at Nyiragongo is described in (Galle *et al.*, 2010). This instrument is automatic and capable of acquiring and transmitting data to be analyzed by dedicated software at the local observatory in real-time and then duplicate the raw data into a centralized archive. The logistical limitations at this particular volcano did not allow permanent connection with the observatory, thus the data collected in the control computer of the system had to be downloaded in-situ during occasional visits to the site and then post-processed. The entire dataset presented in this work was reprocessed with the NOVAC Post-Processing-Software (Johansson, 2011), which uses the same routines that the observatory software but it is tailored for batch-processing of large datasets in a standardized format. Details of the evaluation parameters are given in Table 2. These are standard DOAS variables which can be referred to elsewhere (Platt & Stutz, 2008).

Table 2. Instrumental specifications of the scanning-DOAS systems at Nyiragongo

Instrument type	Scanning DOAS
Wavelength range (nm)	277–464
Spectral resolution[a] (nm)	0.6
Angular (scan) resolution (deg)	1.8 (measuring every second step)
Exposure time[b] (ms)	~100-400
Sampling time (s)	~360 (per scan)
Field of view (mrad)	11
Additional spectra for flux measurement	Zenith (sky), nadir (dark)
Power / W	~5
Dimensions / mm (L×W×H)	~ 600×300×200
DOAS implementation	Dark subtraction, division by sky, high-pass filtering, NL-fitting
Species included in fitting	SO_2, O_3, Ring-effect pseudo-absorber
Wavelength shift correction	Fit to Fraunhofer spectrum
Geometrical calculations	Triangulation if possible, if only one station available calculate plume direction assuming plume at crater altitude
Rejection criteria	Spectra saturated or over-attenuated, completeness factor[c] <0.8, distances to plume >~10 km, altitude error >1000 m, direction error >30 deg

[a] FWHM of 302.15 nm line of Hg measured at the laboratory

[b] Calculated to reach 80% of saturation level at the peak intensity of the full spectral range

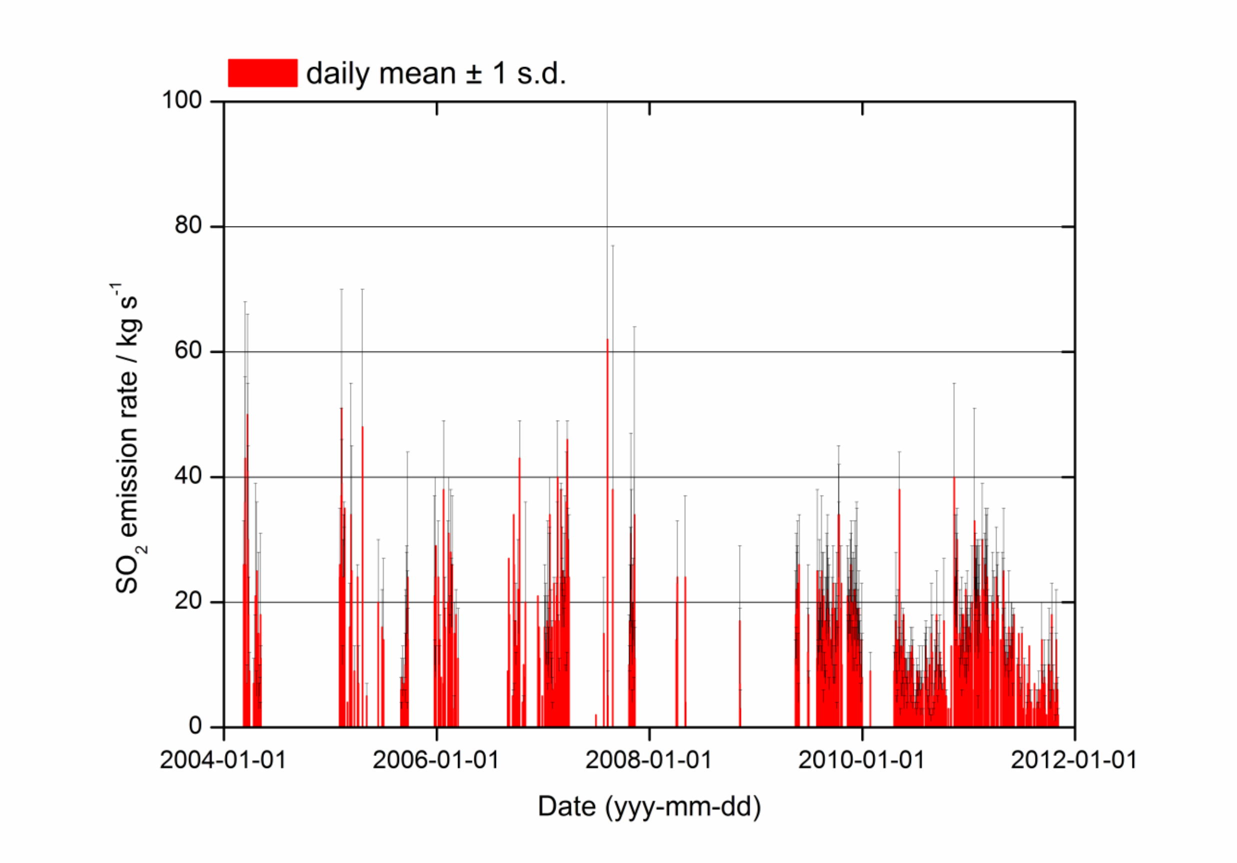
[c] Estimation of the coverage of the plume in one scan (0.5 for plume in the horizon, 1 for complete plume)

2.2 Measurement uncertainty

The sources of uncertainty in this type of measurements are diverse and sometimes difficult to quantify. They correspond both to the assumption of mass conservation, to the spectroscopic measurement of slant column densities, to radiation transport effects, and to the across column integration and estimation of plume height and transport velocity. The first type of uncertainty includes effects such as multiple sources of emission, scavenging by liquid or solid aerosols, chemistry involved in the time of flight of the gas up to the measurement, or dry and wet deposition. Under typical conditions the lifetime of SO_2 before is converted to sulfate aerosol in the equatorial troposphere is in the order of 20 h throughout the year (Lee *et al.*, 2011), although the complex composition of volcanic plumes may reduce this value via heterogeneous processes (Carn *et al.*, 2016). Spectroscopic errors include effects such as improper correction of dark and offset intensities, shot-noise, digitalization and read-out noise, inter-pixel variability, stray light, sensitivity of cross sections to temperature/pressure conditions, spectral shifts and line-shape variations, the "I_0-effect" and inadequate representation of the Ring effect (Stutz & Platt, 1996). These problems are to a high degree controlled by characterizing the instrumental characteristics of the stations (e.g., by measuring the dependence of the instrumental line-shape with temperature, which is measured at the station). The evaluation also takes into account correction of some of these effects, for instance, by co-adding spectra, dividing each spectrum by a measured reference with the same instrumental characteristics, applying wavelength shift corrections by fitting the known features of a solar spectrum, etc. These measures have been adopted for the data analysis presented here. Radiative transfer may be a large source of uncertainty due to complicated effects such as multiple-scattering within the plume, strong-absorption features, or dilution of the signal by radiation entering the spectrometer from a region of the sky not passing through the plume. By performing spectral evaluation at different wavelength ranges with differing sensitivity to these effects, it is possible to detect and, in some cases, correct for them. Another possibility is to reconstruct the measured spectrum out of radiative transfer modeling (Kern *et al.*, 2012, Kern *et al.*, 2010), but this approach is computationally expensive and impractical for the evaluation of a large dataset, like the one presented in this study. Finally, pointing and integration errors, errors in the air-mass-factor corrections and errors in the height, direction and speed of the plume are critical. These are largely reduced when the plume geometry can be constrained by triangulation of the measurements from two or more stations, which was rather the exception in this case. However, the distribution of measured column densities by a single scan gives an indication of the coverage of the plume and its distance to the scanner. It is possible to reduce these types of errors by imposing quality assurance criteria on the results. To the extent of what is assessable by the measurements alone, we estimate that the different uncertainty sources amount to a value of the order of 30-60% (usually skewed towards underestimation of the source strength). This estimation is based on uncertainty analysis by Monte-Carlo sampling from the distributions of the variables involved in the flux calculation (Arellano, 2014).

3. Results

The time series of SO_2 gas emission rates from Nyiragongo for 2004 -2012 is shown in Figure 2a. The intermittency denotes mostly periods when there were no stations in operation; nevertheless, the coverage represents reasonably well the long-term degassing of this volcano, since 669 out of 2797 (~24%) possible days had validated gas emission rate measurements during the period 2004-03-09 to 2011-11-05. The periods of eruption of Nyamuragira volcano where also discarded from this analysis, because it is difficult or impossible to differentiate between emissions from both volcanoes under certain conditions -for a methodology see (Smets *et al.*, 2013)-, and because the magnitude of the eruptive emission from Nyamuragira is usually much larger than the emission from Nyiragongo and will most likely result in a plume that covers all angles of observation. These periods are: 2004-05-10 to 2004-06-13; 2006-11-27 to 200612-03; 2010-01-02 to 2010-01-25; 2011-11-07 to 2012-01-17 (Carn, 2015).


daily mean ± 1 s.d.
SO$_2$ emission rate / kg s^{-1}
100
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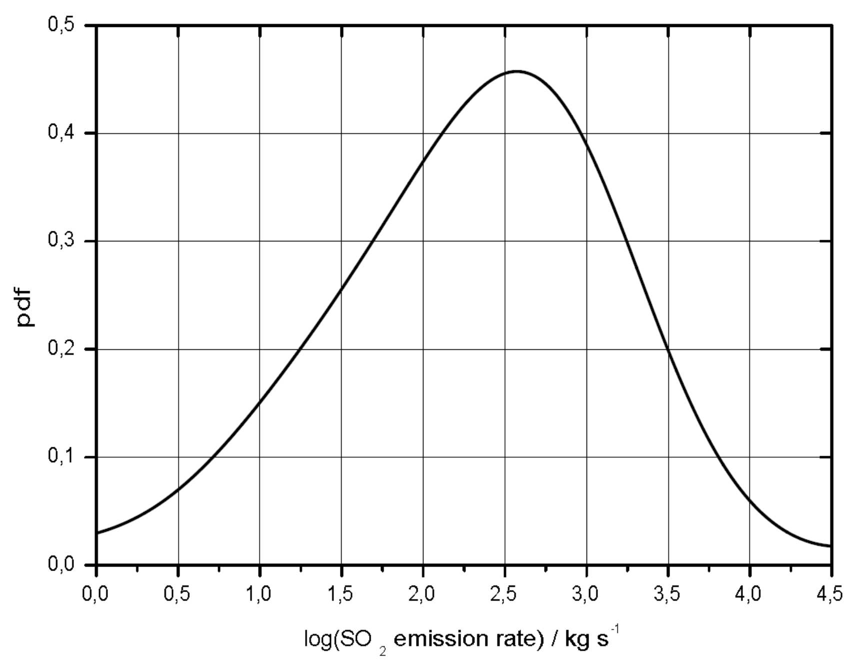
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log(SO$_2$ emission rate) / kg s^{-1}

Figure 2a. SO_2 gas emission rates from Nyiragongo volcano during 2004-2012. The plot shows daily averages and their standard deviations. The range of uncertainty of each measurement is estimated at 30-60%. Gaps in the time series corresponds mostly to periods when the stations were not operational except for the periods of eruption of Nyamuragira volcano (see text). **2b.** Empirical distribution function of the flux measurements from Nyiragongo for the period 2004-2012. The distribution is calculated with a kernel density estimator, and it can be approximated as a log-normal distribution.

The empirical probability density function of the daily emission rate values is shown in Figure 2b. It has been calculated by a kernel density estimator which optimizes the size of the bins in the histogram (Botev *et al.*, 2010). The distribution is approximately log-normal.

Another important parameter for assessing the emission energy and potential impact of volcanic activity is the plume altitude. This is a function of the source conditions, mainly the thermal power and amount of volatiles, as well as of the meteorological conditions in the surrounding atmosphere, principally the wind patterns, stability (vertical temperature) and relative humidity (Sparks *et al.*, 1997). Since most of time there was only one station running during the period of analysis, the plume altitude could not be calculated, except for the ‘focus’ period of April 2010-March 2011, where it was derived by a combination of measurements and meteorological modeling. For the rest of the period of study, the plume altitude was assumed to correspond to the mean altitude of the focus period. This value lies consistently above the summit altitude, which is reasonable according to visual observations of a lifted plume. When combined measurements were available for calculation of the plume height by triangulation, the retrieved values were adopted for measurements performed within the same day. Statistics of the retrieved values are presented in Table 3 and Figure 3, along with other important results of the measurements.

Table 3. Statistics of plume measurements at Nyiragongo volcano during 2004-2012 from ~12000 valid flux measurements taken on 669 days.

	Mean	Std. dev.	Range
SO2 emission rate / kg s^{-1}	13	9	1-62
Plume height / m a.s.l.	3500	277	3470-8530
Plume speed / m s^{-1}	5.2	2.1	0.3-11.9
Plume direction / deg	57.4	15.8	22.3-148

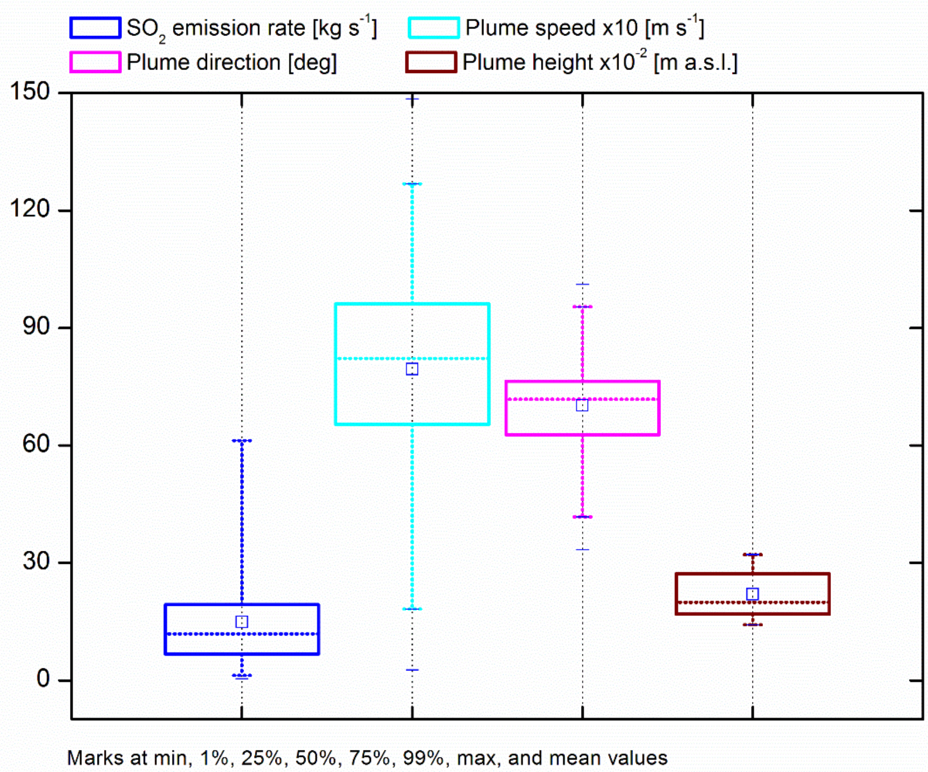

Figure 3. Box-chart plots of the main characteristics of the plumes from Nyiragongo (2004-2012). Note the skewed distribution for gas emission rates, and the more symmetrical distributions for the observed plume height, speed and direction

4. Discussion

The results presented here are representative of the multi-year degassing behavior of Nyiragongo during a period of lava lake stability and quiescent degassing. The inter-day variability is high, being not unusual to have an order of magnitude change between two consecutive days. This characteristic has also been observed in other quiescently degassing volcanoes, of quite different magmatic composition or tectonic environment, monitored with the scanning-DOAS technique (Arellano, 2014). We recall that individual flux measurements were selected based on quality check of the distance to the plume, coverage of the plume and other factors, in order to keep the measurement uncertainty below ~60%. By further averaging measurements taken on the same day, we think the results presented here give a reliable picture of the degassing intensity of Nyiragongo. The inter-day variability reflects the complex dynamics of the lava lake. In order to maintain the lava lake for a sustained period of time, cooling and crystallization produced by outgassing has to be counterbalanced by the influx of gas-rich magma from below. This influx can be periodic, as in the case of bi-directional magma flow, with periods of a few minutes. For example, (Ilanko *et al.*, 2015, Oppenheimer *et al.*, 2009) find cycles of about 10 min and discussed that shorter periods are expected for magmas of lower viscosity, as in the case of Nyiragongo. Such periodicity cannot be resolved for daily flux measurements presented here, and a wavelet analysis of the time series (Grinsted *et al.*, 2004) during 4 periods of 70 days of consecutive measurements each, reveals no signs of characteristic frequencies in days or longer scales. The inter-day variability can be the result of irregular changes in the permeability of the lava lake, due to localized thickening of the crust and the percolation of magma batches of heterogeneous size distribution.

In terms of the total distribution of daily emission rate, 80% of the observed SO_2 fluxes are below 20 kg s^{-1}. Based on observations of the lava lake level and fluid dynamical modelling, (Burgi *et al.*, 2014) inferred that the mass flux of magma necessary to keep the lava lake in equilibrium, against heating losses by degassing, radiative cooling and crystallization, should be between ~9200 and 1700 kg s^{-1}. These two end-members are considered minimum estimates that correspond to two enthalpy models, for dyke intrusion and for cumulate emplacement, respectively (Francis *et al.*, 1993). The distribution of gas fluxes should also be relate to this range of magma gas flow rates at depth. For a SO_2 flux between 7 and 18 kg s^{-1} (25-75 percentiles), assuming that the lower gas fluxes correspond to the lower magma flow rates, the corresponding amounts of S degassed from the magma is between ~2000 and 1000 ppm. Analysis of the S content of degassed lavas after the January 2002 eruption indicated a total S content of ~2500 ppm (Carn, 2004). For this eruption, the amount of S contained in the lava was an order of magnitude larger than the amount of S degassed, a low outgassing efficiency attributed to the high lava effusion rate. The S content derived from our measurements in the period of lava lake stability will on the contrary reflect the predominance of outgassing, with a lower component of gas remaining in the solution, because quiescent degassing would on one hand facilitate gas segregation and occur in equilibrium with the confining pressure, and, on the other hand, it will be reinforced by a more efficient convection rate caused by a larger viscosity/density contrast between degassed and gas rich magma. Conceding the uncertainties involved in the model calculations of the magma influx and the uncertainties in gas flux, it is still remarkable the agreement in the range of S content for this volcano derived by this and independent studies.

On a yearly basis, there is a slightly decreasing trend in the emissions. As the degassed magma sinking down the conduit should be accommodated inside the plumbing system of the volcano (Allard, 1997), it would be interesting to look for signals of ground deformation, considering the magnitude of the involved volumes of magma (Burgi *et al.*, 2014). The mean emission rate during the observed period is similar to that reported by (Le Guern, 1987) during the expedition of 1959, when the level of the lava lake was also similar.

Observations of the plume height, direction and speed are relevant for assessing the impact of the emissions in the surrounding area. Plume height is observed to some hundreds meters above the summit but still within the free troposphere. Nyiragongo is located in the tropics, and the local troposphere is characterized by a high relative humidity. This produces a typical foggy environment, which has implications for the life-time of the plume, since wet deposition, scavenging and oxidation of SO_2 to sulfate aerosol may be important sinks for the crater emission that escape our measurements, resulting in a net underestimation of the emission rate (also by an increased radiative transfer “dilution” effect (Kern *et al.*, 2010)). Another effect of deposition is the occurrence of acid rain, which consequences are evidenced in the surroundings of the volcano, like the acidification of water reservoirs, soil and vegetation, as well as large incidence of health affectations like fluorosis (Baxter, 1990). From the reported values of emission rate, and knowledge of the meteorological conditions, it is possible to estimate ground-level concentrations of the emitted gases at different distances downwind the crater. This study has been recently done by (Dingwell *et al.*, 2016) for the SO_2 ground-level concentration at the most important villages around Nyiragongo.

It is also interesting to compare the record of long-term with past measurements of the bulk gas emission. Such comparison is shown in Figure 4, and highlights the fact that previous reported values correspond to sporadic measurements performed during eruptive periods. In fact our measurements also show occasional bursts of emission, even comparable to those observed during eruptions. However, the bulk emission remains relatively stable, a feature that is only deducible from long-term monitoring.

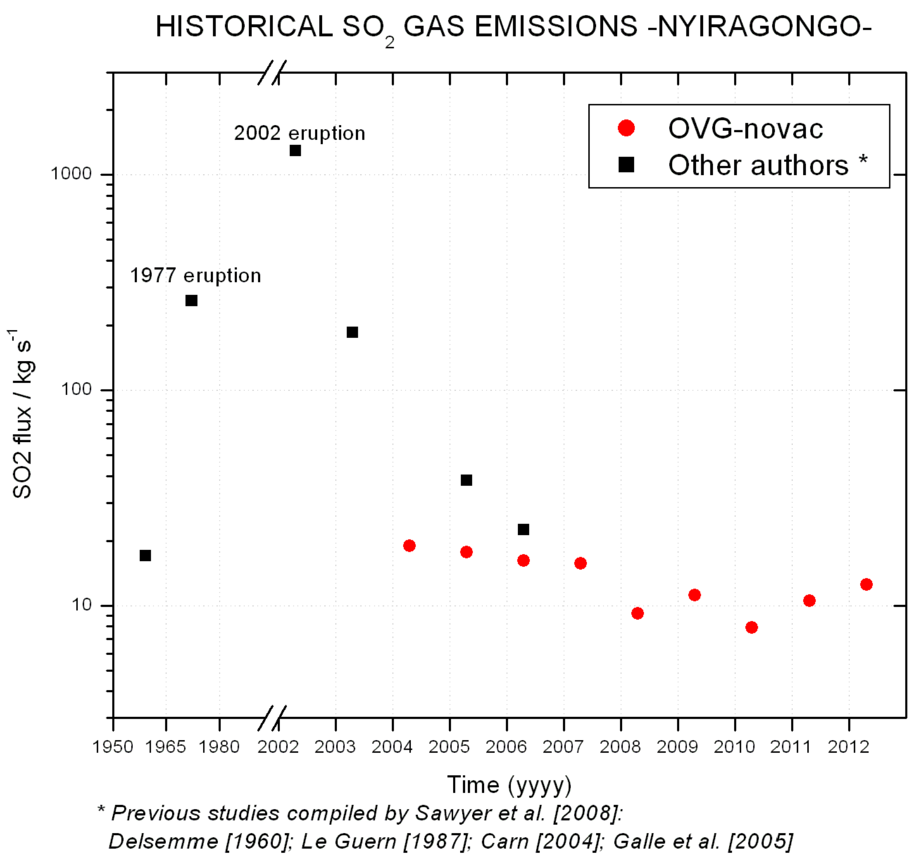

Figure 4. Record of the annual averages of measurements of SO_2 emission rate from Nyiragongo (1959-2012). Measurements before the long-term monitoring were done during short-term field surveys, especially during or after major eruptive events. The record of permanent surveillance indicates relatively stable conditions and no signals of a net increase over the years.

5. Conclusions

By conducting long-term automatic measurements of the emission rate of SO_2 from Nyiragongo volcano with a network of scanning-DOAS remote sensors during 2004-2012, we characterized the time evolution of degassing intensity. This characterization gives a much more complete view which could not be noticed by sporadic field measurements or lower sensitivity satellite-based observations. The measurements have been obtained under particularly demanding circumstances in a politically conflictive area and reveal a remarkable stability in the degassing behavior during the studied years, altered by short-term variations in the gas flux that could be related to variations in the effective permeability of the lava lake and to fluctuations in the rate of magma convection that sustain the lava lake during these years. From the scaling of magma flow rate required for this stability and the measurements of gas flux, it is possible to estimate the S content in the magma to be 1000-2000 ppm. The flux of SO_2 during 2004-2012 is similar to that observed in 1959. This paper emphasizes the importance of short-range, long term gas monitoring to understand the activity of volcanoes like Nyiragongo, characterized by persistent, quiescent plumes and high associated levels of risk.

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Supplementary material

Dataset of daily mean SO2 flux from Nyiragongo volcano during 2004-2011 (Arellano_etal_nyiragongo_so2Flux_2004_2011.csv).

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