Research paper

SO$_2$ loss rates in the plume emitted by Soufrière Hills volcano, Montserrat

Lizzette A. Rodríguez [a,b,*], I. Matthew Watson [b,c], Marie Edmonds [d], Graham Ryan [e], Vicky Hards [e,f], Clive M.M. Oppenheimer [d], Gregg J.S. Bluth [b]

[a] Department of Geology, University of Puerto Rico — Mayagüez Campus, PO Box 9017, Mayagüez, 00681, Puerto Rico
[b] Geological and Mining Engineering and Sciences, Michigan Technological University, 1400 Townsend Dr, Houghton, MI 49931, United States
[c] Earth Sciences, University of Bristol, Queen's Road, Bristol BS8 1RJ, UK
[d] Department of Geography, Downing Place, Cambridge University, Cambridge CB2 3EN, UK
[e] Montserrat Volcano Observatory, Flemmings, Montserrat, BWI, UK
[f] British Geological Survey, Keyworth, Nottingham NG12 5GG, UK

Abstract

To improve interpretation of volcanic SO$_2$ flux data, it is necessary to quantify and understand reactions involving SO$_2$ in volcanic plumes. SO$_2$ is lost in volcanic plumes through a number of mechanisms. Here we report SO$_2$ measurements made with miniature ultraviolet spectrometers at Soufrière Hills volcano, Montserrat; a low altitude volcano (∼1000 m above sea level) whose plume entrains humid marine air in the planetary boundary layer. Traverses very near (<400 m) beneath the ash-free plume were made at various distances from the source (from ∼2 km to ∼16 km), thereby spanning plume ages of about 6 to 35 min with minimal attenuation. We find average SO$_2$ loss rates of ∼10^{-4} s^{-1} (e-folding time of ∼2.78 h), slightly lower than estimated previously for Soufrière Hills. These are in the fast end of the range of loss rates measured at other volcanoes (10^{-3}–10^{-7} s^{-1}, e-folding times of 0.28–2778 h), indicating that Montserrat plumes have short SO$_2$ lifetimes. This work is more detailed and precise than previous work and is likely to represent the general case at Montserrat. SO$_2$ flux measurements made >2 km downwind from Soufrière Hills volcano significantly underestimate at-source SO$_2$ emission rates, on the order of 70–146%, when not accounting for the decay rate. Similar SO$_2$ loss is likely to occur in plumes from other tropical low altitude volcanoes under conditions of high relative humidity (∼20% of active volcanoes worldwide). These results suggest that the global volcanic SO$_2$ emission rate may be underestimated as the estimates are based on measurements taken downwind of volcanoes, by which time significant loss of SO$_2$ may have taken place. The loss rates calculated here could be used, in conjunction with downwind SO$_2$ fluxes, to estimate at-source SO$_2$ emission rates from volcanoes with similar environmental conditions to those at Soufrière Hills volcano.
© 2008 Elsevier B.V. All rights reserved.

Keywords: sulfur dioxide; loss rate; plume chemistry; DOAS; Soufrière Hills

1. Introduction

It is important to understand volcanic SO$_2$ reactions in different meteorological environments, because uncertainties in near-source plume chemistry can complicate interpretations of gas plume measurements, which are used for volcanic monitoring purposes and in atmospheric and environmental impact studies. Sulfur dioxide is typically the third most abundant volcanic gas (Symonds et al., 1994), and is a trace constituent of the Earth's atmosphere, mainly present in the troposphere (Vandaele et al., 1994). It is an environmentally significant species associated with direct and indirect (acidification) damage to ecosystems (Delmelle, 2003) and is potentially harmful to human health (WHO Air quality guidelines, 2006; Hansell and Oppenheimer, 2006). Sulfur emissions are also relevant to the radiative transfer of the atmosphere, particularly through the action of sulfate aerosol (Lacis et al., 1992).

The difficulties inherent in the measurement of continuous, persistent emission of relatively small concentrations of gases, occurring between and during long-lived eruptions from volcanoes, have meant that few studies have been conducted to look at their impact on the troposphere. However, the contribution of quiescent, continuous emissions of SO_2 and sulfate aerosols from volcanoes is significant and must be known when attempting to quantify anthropogenic emissions and their effects on climate (Robock, 2003). Anthropogenic emissions contribute about 78 Tg S yr^{-1} to the atmosphere (Bates et al., 1987, 1992), which is ~76% of the global S emissions (Bates et al., 1992; Spiro et al., 1992; Andres and Kasgnoc, 1998). The uncertainties in the measurements of passive emissions are caused by (1) the study of only a fraction of passive degassing volcanoes, (2) errors arising from measurement techniques (Rodríguez et al., 2004), and (3) factors unique to each volcano that can make measurements logistically difficult and less accurate (e.g., access, meteorology, volcanic activity). Even at volcanoes that are intensively monitored, there are often uncertainties in some of the parameters needed to calculate the emission rates accurately. Uncertainties in measurements from ground-based (correlation spectrometer, UV spectrometers) studies of passive emissions propagate throughout global volcanogenic SO_2 budgets. Most studies are based on a low percentage of the total number of active volcanoes (e.g., 20%, Andres and Kasgnoc, 1998). Andres and Kasgnoc (1998) calculated a time-averaged flux of ~13.4 Tg yr^{-1} SO_2, based on gas data between 1970 and 1997. Previous studies estimate a range in global volcanic emission of 1.5 and 50 Tg yr^{-1} SO_2 (from Table 5, Andres and Kasgnoc, 1998), which includes both passive and explosive degassing. The range in the estimates is an indication of the uncertainty in calculating the annual global volcanic SO_2 emissions.

Volcanic SO_2 emission rates are commonly measured using ground-based spectroscopic techniques (e.g., Galle et al., 2002; McGonigle et al., 2002; Edmonds et al., 2003). The column concentration of SO_2 is integrated across a traverse of the plume, perpendicular to the transport direction, at some distance downwind of the volcano. Traverses are generally not exactly perpendicular to the plume transport direction; therefore they have to be corrected for this effect. The corrected column concentration is then multiplied by the plume speed to yield the SO_2 flux. This is assumed to be equal to the emission rate from the source. Typically, the measurements are undertaken a few kilometers downwind from the volcano and perhaps up to several kilometers away from the plume, depending in the most part on access and safety. This allows time for SO_2 to interact with other volcanogenic gases, particles and droplets of volcanogenic and/or meteoric origin, as well as atmospheric gases and aerosols, and for the possibility of attenuation of SO_2 signal by airlight or scattering. The processes leading to SO_2 loss during transport downwind include oxidation (conversion to sulfate aerosol), dry and wet deposition, absorption, and dissolution. The effects of one process versus another are challenging to separate and, for this work, we will assume that the loss of SO_2 is dominated by in-cloud mechanisms.

Local meteorology affects the fate of tropospheric plumes initially by dispersion and transport downwind and secondly through factors such as humidity, temperature, the amount of sunlight reaching the plume (solar radiation), cloud cover, fog, and precipitation (Oppenheimer et al., 1998; Horrocks et al., 2003). An important SO_2 depletion process is the conversion of SO_2 to SO_4^{2} (sulfate). This conversion can proceed by both homogeneous and heterogeneous mechanisms to form particulate sulfate in the atmosphere (Eatough et al., 1994). In the gas phase, oxidation occurs by reaction with hydroxyl radicals to form sulfuric acid, while aqueous-phase oxidation (multiphase reaction) of SO_2 involves reaction with H_2O_2 or O_3 to form sulfate (Eatough et al., 1994).

The processes mentioned above, among others, can remove or mask the SO_2; therefore the measured SO_2 flux may not represent the at-source SO_2 emission rate. SO_2 loss rates ranging from 10^{-7} (e.g., Mount St. Helens) to 10^{-3} (e.g., Soufrière Hills) s^{-1} have been estimated for tropospheric volcanic plumes at various altitudes (e.g., Martin et al., 1986; Oppenheimer et al., 1998; McGonigle et al., 2004). The loss rates were estimated using a number of different techniques (ground and satellite-based), including the correlation spectrometer (COSPEC), UV spectrometers, photometry, the Total Ozone Mapping Spectrometer, and filters (e.g., Martin et al., 1986; Oppenheimer et al., 1998; McGonigle et al., 2004). Each technique has advantages and disadvantages, and the results may not be comparable directly, however, this also justifies the need to determine more accurately the range in SO_2 loss rates in volcanic plumes. Recent studies have shown variation on the order of at least two orders of magnitude for volcanoes with similar conditions (e.g., meteorological, SO_2 emission rates): Soufrière Hills volcano (SHV) and Masaya volcano, Nicaragua. Very fast loss rates (~10^{-3} s^{-1}, e-folding times ~17 min) were calculated by Oppenheimer et al. (1998) at SHV in 1996 (using a COSPEC), while McGonigle et al. (2004) determined very slow to negligible loss (~10^{-5} s^{-1}, e-folding times ~28 h) at Masaya in 2003 (using UV spectrometers).

This work aims to quantify SO_2 depletion rates in volcanic plumes injected into the boundary layer, from the SHV, Montserrat, using ground-based remote sensing techniques. SHV is a low altitude volcano in a humid environment (conditions typical of ~20% of active volcanoes worldwide; Smithsonian's Global Volcanism Program, Summary Lists). In order to address potential underestimates on global emission rates, SO_2 fluxes were measured near to the eruptive vent and at various distances downwind of the SHV (Fig. 1). The two main reasons for choosing SHV were: (1) it is one of the world's most heavily monitored volcanoes, with a continuous gas monitoring network (ultraviolet (UV) spectrometers) since 2002 (Edmonds et al., 2003) and (2) previous measurements of SO_2 loss indicated fast loss rates, in the order of 10^{-3} s^{-1} (Oppenheimer et al., 1998). The summit of SHV is at <1100 m asl and its average relative humidity is generally >75%. The current eruption began in July 1995 and has been characterized by periods of lava dome growth, dome collapses with pyroclastic density currents, and episodes of explosive activity. The mean SO_2 emission rate for the entire period from 1995–2005 is ~500 t d^{-1} (5.8 kg s^{-1}); with higher emission rates occurring during periods of high extrusion rate and immediately after large dome collapses (MVO data). The volcanic plume typically is at ~1000 m above sea level or lower, many times close to the ground. It typically enters the tropical, marine boundary layer and drifts towards the west under the influence of the trade winds. The location of Montserrat in the trade winds belt represents an unusually constant meteorological