

## DATA AND EXPERIMENT DESCRIPTIONS

Continuous SO<sub>2</sub> flux measurements for Vulcano Island, Italy

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**ABSTRACT**

The La Fossa cone of Vulcano Island (Aeolian Archipelago, Italy) is a closed conduit volcano. Today, Vulcano Island is characterized by sulfataric activity, with a large fumarolic field that is mainly located in the summit area. A scanning differential optical absorption spectroscopy instrument designed by the Optical Sensing Group of Chalmers University of Technology in Göteborg, Sweden, was installed in the framework of the European project "Network for Observation of Volcanic and Atmospheric Change", in March 2008. This study presents the first dataset of SO<sub>2</sub> plume fluxes recorded for a closed volcanic system. Between 2008 and 2010, the SO<sub>2</sub> fluxes recorded showed average values of 12 t.d<sup>-1</sup> during the normal sulfataric activity of Vulcano Island, with one exceptional event of strong degassing that occurred between September and December, 2009, when the SO<sub>2</sub> emissions reached up to 100 t.d<sup>-1</sup>.

**1. Introduction**

Geochemical studies of fumarolic exhalations from active volcanoes indicate that the gas emissions are directly fed by degassing magma. Over the last 20 years, many geochemical investigations have focused on the study of fumarolic gases, as tools to quantify the degassing processes during both active and quiescent phases of volcanic activity [e.g. Italiano et al. 1997, Aiuppa et al. 2005, Allard et al. 2005, Inguaggiato et al. 2011, Inguaggiato et al. 2012a]. In particular, SO<sub>2</sub> fluxes measured for the plume of volcanic systems appear to be a promising parameter to study these degassing processes [e.g. Casadevall et al. 1983, Allard et al. 2005, McGonigle 2005]. With the aim to study the SO<sub>2</sub> fluxes of Vulcano Island (Aeolian Archipelago, Italy), a scanning differential optical absorption spectroscopy (DOAS) instrument was installed at Palizzi (on the west side of Vulcano Island, near the crater) in March 2008. This instrument acquired a dataset of SO<sub>2</sub> fluxes in continuous mode directly from the summit crater.

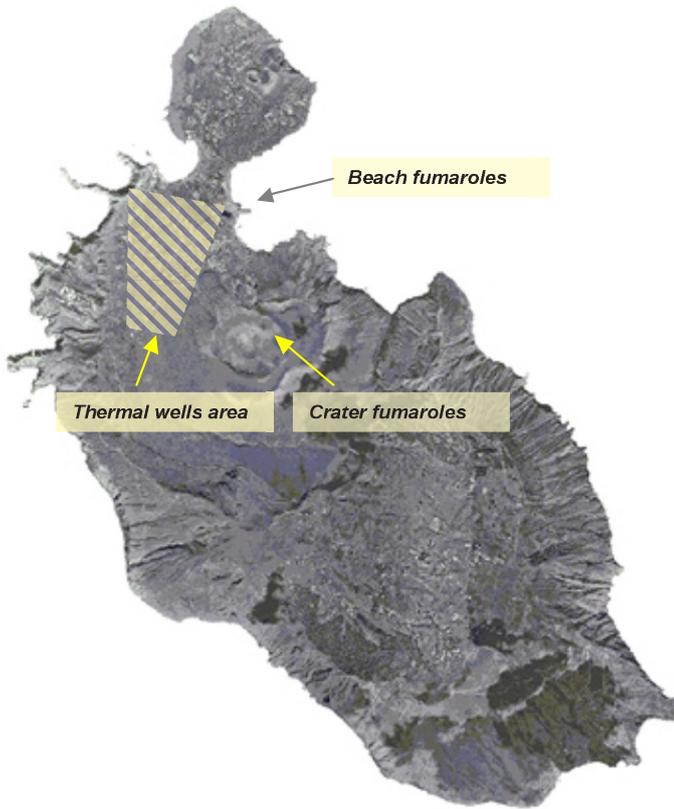
Vulcano Island is one of the currently active volcanoes of

the Aeolian Island Arc, located north of Sicily, and it is characterized mainly by explosive eruptions. The last eruption from La Fossa crater occurred between 1888 and 1890. Since that time, the crater has been affected by intense degassing of fumarolic fluids at high temperatures (up to 700 °C in 1993). There is also fumarolic activity at Forgia Vecchia, a few hundred meters north of La Fossa crater, where low-pressure fumarolic vapor escapes at 100 °C, and also at Porto di Levante Beach, where the fluids are fed by the boiling of a hydrothermal system, with their temperature buffered at 100 °C. La Fossa crater fumaroles show large variations in their flow rates, temperatures and compositions over time.

Increasing fumarolic activity preceded the 1888 eruption, but no eruption occurred after a later increase in the fumarole temperatures, from 200 °C in 1913, to 615 °C in 1923. A more recent phase of increasing heat output started in 1977, with a temperature rise in the crater fumaroles from 200 °C to 220 °C, to about 700 °C in 1993. Detailed studies of the geochemistry of the fumarolic fluids of Vulcano Island started in the late 1970's, concurrent with the rapid changes that were happening at La Fossa crater.

At present, the main exhalative activity is concentrated in the crater area (see Figure 1), with a wide fumarolic field, and with temperature ranging from 100 °C to 450 °C. Moreover, strong soil degassing that is dominated by CO<sub>2</sub> occurs all around the volcanic edifice, and particularly where the active tectonic discontinuities drive the gases (mainly water vapor and CO<sub>2</sub>) to the surface [e.g. Capasso et al. 1997, Diliberto et al. 2002]. Finally, numerous CO<sub>2</sub> thermal wells (at up to 80 °C) in the Vulcano Porto area testify to the presence of a geothermal system with an equilibrium temperature of around 200 °C [Federico et al. 2010].

Over the last few decades, Vulcano Island has shown indications of renewed activity due to increases in deeper



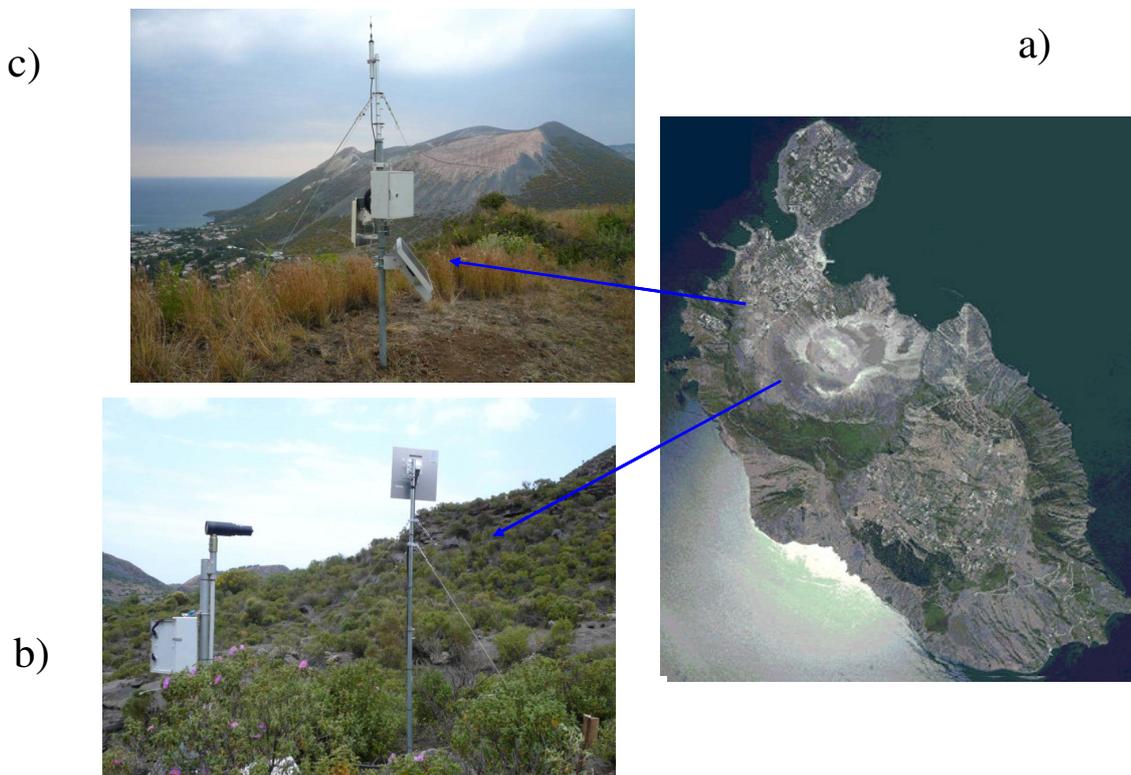
**Figure 1.** Vulcano Island, Aeolian Arcipelago, Italy. Main exhalative activity areas are indicated: crater fumaroles, beach fumaroles, and thermal wells.

fluid fluxes that have been highlighted by increases in the temperatures and volatile fluxes of the fumarolic area [Chiodini et al. 1996, Paonita et al. 2002], and strong variations in the chemical and isotopic compositions of the crater fumaroles and thermal waters [Capasso et al. 1997, 1999].

Over the last 10 years, La Fossa crater has been used as a natural laboratory to test new remote and *in-situ* sensing techniques. In particular, Mori et al. [1995] measured the  $\text{SO}_2/\text{HCl}$  ratio of the Vulcano Island plume using Fourier transform infrared (FTIR) spectroscopy, and Aiuppa et al. [2004] carried out an inter-comparison on the plume of Vulcano Island using different methodologies (FTIR, filter-packs, direct sampling). O'Dwyer et al. [2003] performed real-time measurements of volcanic  $\text{H}_2\text{S}$  and  $\text{SO}_2$  concentrations with a prototype instrument based on ultraviolet (UV) spectroscopy.

As Vulcano Island is a closed conduit volcano with only sulfataric activity and no real 'plume', telemetric measurements of  $\text{SO}_2$  fluxes of the plume have not been performed systematically in the past.

In the framework of the "Network for Observation of Volcanic and Atmospheric Change" (NOVAC), a worldwide network of permanent scanning DOAS instruments [Galle et al. 2010] was installed for 19 volcanoes around the world, to measure real-time volcanic  $\text{SO}_2$  emission fluxes. Within this context, a scanning DOAS instrument was installed at



**Figure 2.** The UV-Scanning DOAS equipment installed in the Palizzi area on the south-east flank of Vulcano island. (a) Map showing the positions of the UV-scanning DOAS and the WLAN bridge (Map © 2009 Google). (b) Photograph of the scanning DOAS station. (c) Photograph of the WLAN bridge for data transfer.

Palizzi (the west side of Vulcano Island, near the active crater) in March 2008 (see Figure 2).

## 2. Methods

A UV-scanning DOAS MARK1 instrument developed as part of the NOVAC project [Galle et al. 2010] was installed in the Palizzi area in March 2008, together with a weather station, on Lentia Hill at the same elevation (350 m a.s.l.) as La Fossa crater (Figure 3). The weather station provided wind speed and direction, rainfall, temperature, relative humidity, and UV, visible and infrared radiation. From the measurement station, these data arrived at the Vulcano Observatory through a wireless system. From Vulcano Observatory, the data were transmitted in a real time via the internet to the Istituto Nazionale di Geofisica e Vulcanologia (National Institute of Geophysics and Volcanology, INGV, Palermo).

The scanning DOAS instrument was installed very close to the source, at about 700 m from an emission area of SO<sub>2</sub> where the slant column is high enough to be detected (50 to 250 ppmm), and therefore providing the possibility to calculate the fluxes (Figure 4). The system was equipped with a single spectrometer from the Ocean Optics Company (S2000 spectrograph), an embedded personal computer, a global positioning system receiver, a timer, optical fiber, and a telescope. A technical report relating to this continuous monitoring with the scanning DOAS system, including the instrument design, station networking, and measurement geometries, is described in more detail by e.g. Zhang [2005], Galle et al. [2010] and Johansson et al. [2009]. UV light from the sun that is scattered from aerosols and molecules in the atmosphere is partly absorbed in passing the volcanic plume and then collected using the telescope with a quartz lens that defines a field-of-view of 16 mRad. The light is then transferred from the telescope to the spectrometer via the optical quartz fiber. The spectrometer used a 2,400 lines/mm grating combined with a 50 μm slit, which provided an optical resolution of ca. 0.6 nm over a wavelength range of 280 nm to 390 nm. The telescope automatically moved from horizon to horizon using a stepper motor. For further details, see Galle et al. [2010].

The settings of the station at Vulcano Island were characterized by moving the telescope from 90° to -90° (horizon in the NNW direction, to horizon in the SSE direction). For every elevation angle, 15 spectra were collated for one measurement. Every full measurement cycle contained 53 spectra: one zenith sky spectra, one dark current spectra, one offset spectra, and 50 measurement spectra. This usually took about 10 min, depending on the light conditions of the sky. The SO<sub>2</sub> fluxes were calculated with the NOVAC software [Johansson et al. 2009]. The DOAS station worked only for the time span from 07:30 hours to 16:30 hours (GMT), because of light limitations. During the first months of observations, a traditional flat scanner system was used,



**Figure 3.** The weather station in the Lentia area (same altitude of La Fossa crater), less than 2 km from the Vulcano crater.

which scanned a vertical plane that cut the transport direction of the plume; however, unfortunately, the plume was rarely hit. This was improved by changing this to a conical scanner system at the end of June 2008. The plume was then scanned along a conical surface, rather a vertical one. The new method significantly increased the wind direction range that was covered by a single scanning instrument, and at the same time, it reduced problems related to multiple scattering effects [Galle et al. 2010].

As there were low SO<sub>2</sub> concentrations in the emissions of Vulcano Island, these SO<sub>2</sub> fluxes were usually recalculated (see the DOAS analysis below), to ensure the quality of the data.

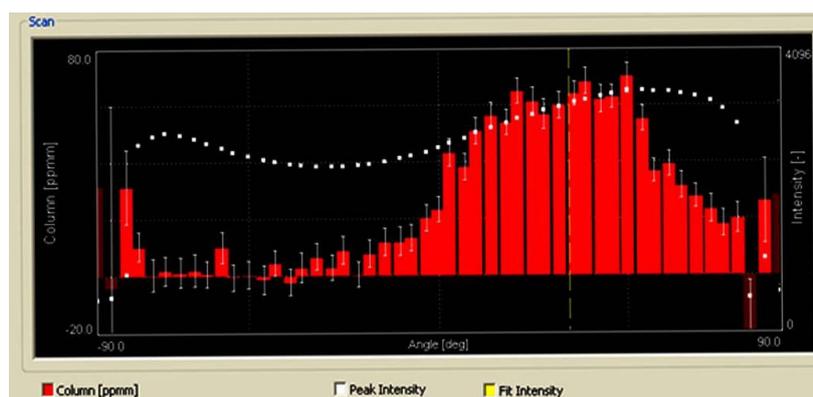
## 3. Analysis of the data evaluation of the flux

### 3.1. Evaluation of the spectra

The collected spectra were analyzed according to the DOAS procedures, which are based on the Beer Lamberts Law. Each spectrum was first corrected for the electronic noise (offset and dark current).

The optical density was obtained by dividing each spectrum measurement by a reference spectrum, as a spectrum taken outside the plume, and then a logarithm was taken of the result of this calculation. A polynomial was used to take account of any broadband extinction structures that were caused by broadband absorption of trace gases [Platt and Perner 1983, Platt 1994]. A high-pass filter was applied to the absorption cross-sections to use only the remaining high frequency structure, which is unique for any trace gas, and which can therefore be used to determine its abundance. Shift and squeeze were allowed for the absorption cross-sections [Bogumil et al. 2003], to compensate for any small shifts, caused mainly by the variations in the temperatures of the spectrometer and the detector unit.

To improve and to find the best solutions for the data evaluation, several tests were performed on data from this



a)



b)

**Figure 4.** Two examples of the distribution of the plume on two measurements made under conditions of different degassing activities. The graphs were produced by the NOVAC software, and shows the positions of the telescope during a single measurement, from  $90^\circ$  to  $-90^\circ$  (X axis; horizon to horizon), for a total 50 position for one complete measure, and the column density of  $\text{SO}_2$  (Y axis; ppmm). The white dots represent the peak intensities of the light, and the red bars the column density for each position of the telescope during the measurement. (a) Measurement during 2008 (normal activity, ca.  $12\text{t}\cdot\text{d}^{-1}$ ). (b) Measurement on November 30, 2009 (increased activity, ca.  $60\text{t}\cdot\text{d}^{-1}$ ).

'scanning DOAS' on Vulcano Island. The dataset of August 14, 2008, was chosen to carry out various sensitivity studies on the  $\text{SO}_2$  evaluation. On this day, the volcanic gas emissions were clearly inside the geometrical plane of the instrument. For all of the tests performed, the NOVAC software was used, and the data of all of the scans from this day are compared below.

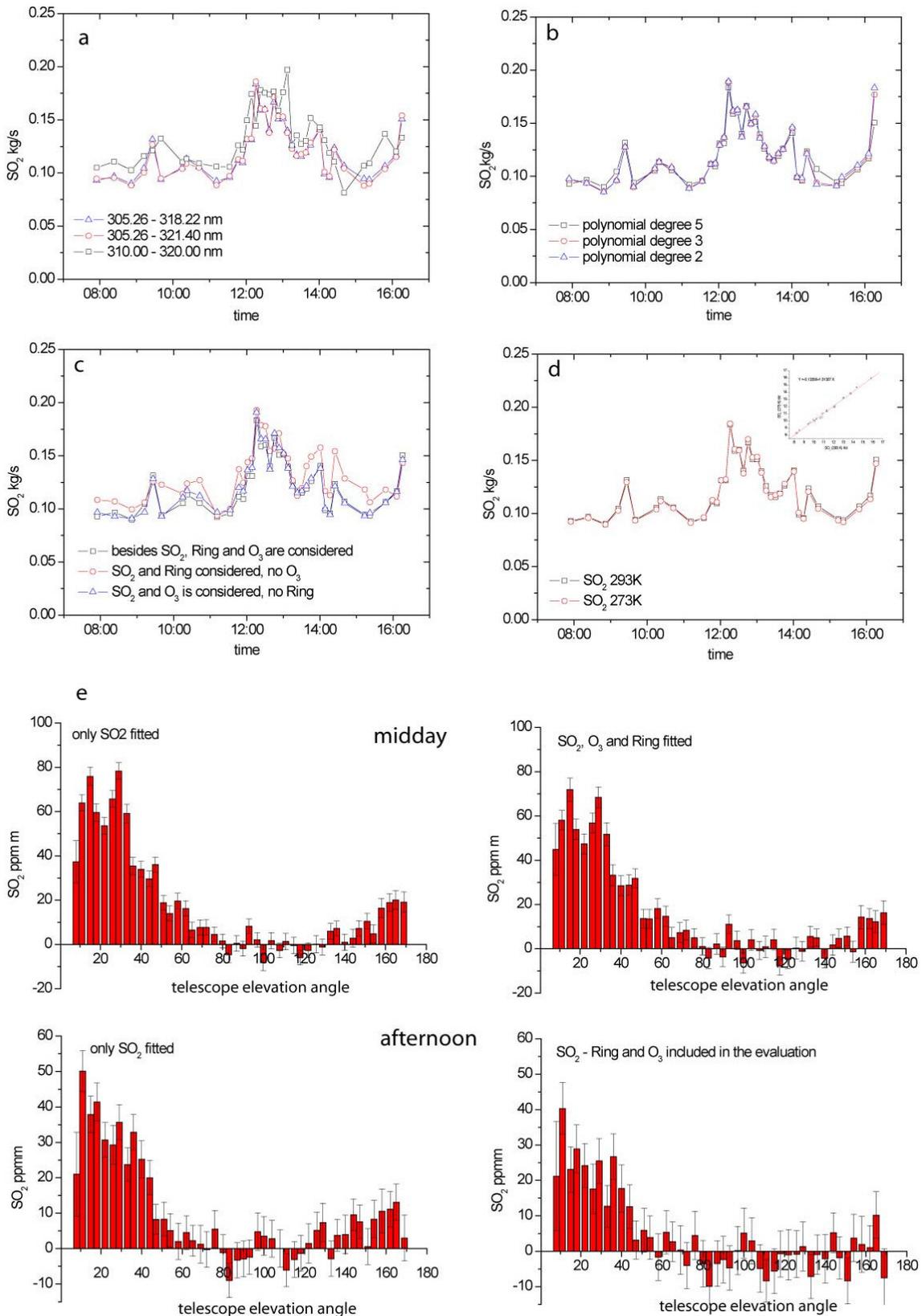
The results presented, discussed and shown in the various Figures, are always based on and compared to an evaluation carried out in the 305.26 nm to 318.22 nm range, which includes  $\text{SO}_2$ , ozone ( $\text{O}_3$ ), a Ring spectra, and a polynomial of the 5th degree. In the first tests, the wavelength evaluation was slightly expanded from the range of 305.26 nm to 318.22 nm, to 305.26 nm to 321.40 nm. The second test was performed by shifting the evaluation range to 310.00 nm to 320.00 nm. Figure 5a shows the results expressed in  $\text{kg}\cdot\text{s}^{-1}$   $\text{SO}_2$  over the day analyzed for these wavelength regions. The different data obtained in the analysis for the various wavelength ranges are shown in Figure 5a with different colors and symbols. Neither the slight shift nor the larger shift analyzed showed clear differences in the  $\text{SO}_2$  fluxes, although a systematic

increase can be noted for the shift to the larger wavelength region. Figure 5b shows the results (again displayed with different colors) from applying various polynomial degrees (2nd, 3rd and 5th degrees) to remove the broadband extinction structures from the spectra. No differences can be seen in these data. Figure 5c shows the results considering the correction or no correction for the Ring effects, as well as taking into account or not the  $\text{O}_3$  absorption. If only the Ring effects are considered (Figure 5c, red circles), the  $\text{SO}_2$  fluxes are generally larger; instead, considering only the  $\text{O}_3$  presence (Figure 5c, blue triangles) and leaving the Ring effects out of the consideration, leads to comparable results as when considering both (Ring effects and  $\text{O}_3$  absorption).

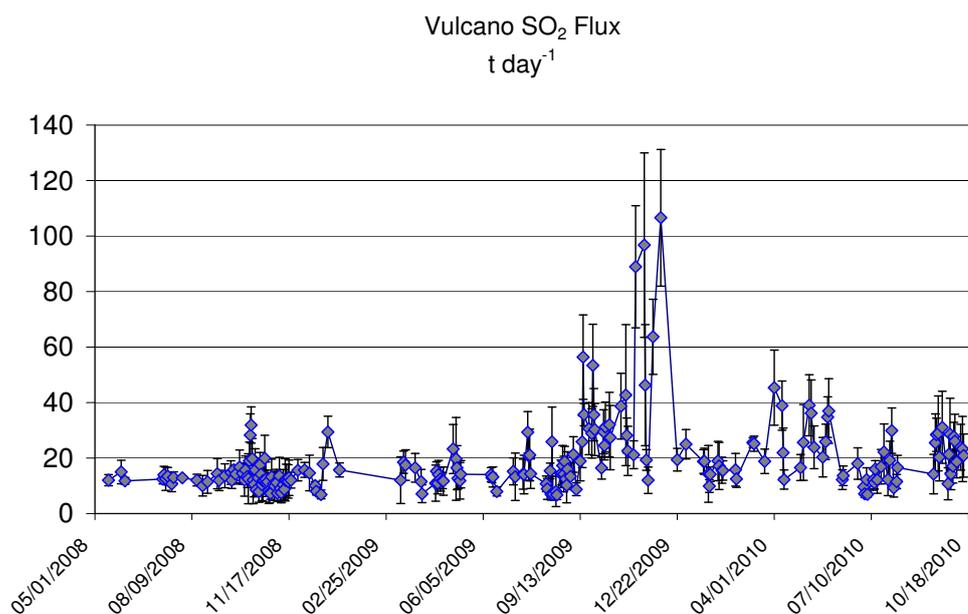
Figure 5d shows the last sensitive analysis carried out for this study; two different  $\text{SO}_2$  references were used (273 K, 293 K), and the results were compared. The data obtained are almost overlapping, in contrast to earlier studies that were performed by Bobrowski [2005], where a difference of 7% was observed.

The most notable difference is the effect of the  $\text{O}_3$  absorption, and this is shown in more detail in Figure 5e.

CONTINUOUS SO<sub>2</sub> FLUX MEASUREMENTS FOR VULCANO ISLAND, ITALY



**Figure 5.** SO<sub>2</sub> fluxes as kg/s SO<sub>2</sub> over the duration of one day (a-d), and SO<sub>2</sub> plume scans as ppmm on August 14, 2008, as a function of the telescope elevation angle, scanning from horizon 0° to horizon 180° (e). (a) A small shift in the wavelength region does not show significant changes. If the shorter wavelength range shifted to a longer wavelength, the SO<sub>2</sub> fluxes are slightly higher. (b) Applying various polynomial degrees (2nd, 3rd and 5th degrees) to remove the broadband extinction structures from the spectra. No significant difference can be seen. (c) With correction or no correction for the Ring effects, and taking into account or not the O<sub>3</sub> absorption, this leads to lower SO<sub>2</sub> fluxes, especially at the beginning of the day. (d) Using two different cross-sections of SO<sub>2</sub> (taken as 273 K and 293 K; Bogumil et al. [2003]), with very little difference noted between these evaluations. (e) The SO<sub>2</sub> plume-scan values are general higher by fitting only a SO<sub>2</sub> reference to the spectra, especially for telescope elevation angles close to the horizon. In the afternoon (lower panels) this effect gets even stronger (difference of up to 10 ppmm between the two evaluations). This is as would be expected, because of an increased ozone column due to a longer light path in the stratosphere.



**Figure 6.** SO<sub>2</sub> plume flux (t.d<sup>-1</sup>) time variations, for the 2008 to 2010 period of the continuous dataset.

This shows two plume scan examples, one at midday and the other in the afternoon, and compares the fit of the results obtained without and with the ozone (O<sub>3</sub>) included. It can be noted here that without including O<sub>3</sub> in the evaluation process, the SO<sub>2</sub> values obtained are generally higher, especially when the Fraunhofer reference spectra has a longer time distance to the measurement spectra in the end of a measurement cycle (scan). In the afternoon, this effect gets even stronger, showing a difference of up to 10 ppm. This is expected, because of an increased ozone column due to a longer light path in the stratosphere when the sunlight enters the Earth atmosphere under a higher solar zenith angle [Hönninger et al. 2004].

### 3.2. Error analysis

The errors identified in these measurements are mainly due to spectroscopy, atmospheric scattering, and wind parameters.

*Spectroscopic error* is related to nonlinear absorption, errors in the absorption cross-section, stray light, and temperature changes of the spectrometer. While the errors in absorption cross-section and nonlinear behavior in Beer's law are a general nature of this technique and have been well documented [e.g. Bobrowski 2005, Platt and Stutz 2008, Kern 2009], the major spectroscopy errors in our study came from stray light and changes in temperature. These two causes differ in size for each spectrometer [e.g. Bobrowski 2005]. To reduce the stray light error we used a filter (Hoya U330), and to account for the temperature effects, we chose the Fraunhofer reference spectra at a similar temperature and a spectra for the dark correction about every 10 min. Furthermore, a squeeze for the absorption cross-section was allowed. Overall the spectroscopic error was estimated to be in the range of 10% [Galle et al. 2010].

### 3.3. Atmospheric scattering errors

Solar light is scattered by molecules and particles in the air, like volcanic ash, condensed water and other molecules, and this can give an extended or reduced path length in the plume. The scattering error strongly depends on the meteorological conditions, volcanic activity and emissions, and the distance between the instrument and the plume [Kern et al. 2010]. In the present study, we did not correct for the atmospheric scattering errors. We assumed an error not larger than 20%, as the instrument was relatively closely situated to the plume, at less than 800 m. Also, strong condensation rarely took place and volcanic ash was not emitted during this time period.

### 3.4. Wind parameter error

To calculate the flux, the concentrations of the molecules in a cross-section perpendicular to the direction of propagation of the plume must be multiplied by the wind speed. To reduce the SO<sub>2</sub> flux error, the wind speed and the wind direction must be measured directly in the plume above the instrument, or at least at the same elevation. Therefore the meteorological station installed at Lentia Hill showed an estimated error always below 20%.

## 4. Results and conclusions

The SO<sub>2</sub> fluxes for the 2008 to 2010 period are reported in Figure 6. These were filtered from values that contained less than 80% of the plume in the scanned plane. It is worth noting that after the change from flat to conical scanning optics, the frequency that the plume was inside the scanning plane increased significantly. Sensitivity studies for the analysis of SO<sub>2</sub> showed that the chosen algorithm was relatively stable for these low SO<sub>2</sub> values. Even the evaluation over the low wavelength range with low photon statistics did not appear

to influence the data significantly. However, if O<sub>3</sub> is excluded from the fitting process, the SO<sub>2</sub> values can be slightly overestimated, especially in the later afternoon hours.

Figure 6 shows the SO<sub>2</sub> flux as a function of time, where we can observe a relatively constant value of the SO<sub>2</sub> flux of around 12 t.d<sup>-1</sup> in the period from August 2008 to August 2009. In mid-September 2009, there was a large increase in SO<sub>2</sub> emissions recorded, with fluxes up to 100 t.d<sup>-1</sup> reached in November 2009. Then, the values of this SO<sub>2</sub> flux decreased to the normal values of about 12 t.d<sup>-1</sup>, although high scattering of the SO<sub>2</sub> flux values was recorded in the spring of 2010 (up to 40 t.d<sup>-1</sup>).

It is very interesting to see that these anomalous values of the SO<sub>2</sub> fluxes that showed an increase up to about one order of magnitude occurred in the same period as the anomalous degassing processes of the soil CO<sub>2</sub> recorded by a continuous monitoring station located at the summit of La Fossa crater [Inguaggiato et al. 2012b]. Indeed, in the September to December 2009 period, the measurement of the soil CO<sub>2</sub> flux at the summit of the area of La Fossa cone recorded huge variations, passing from mean fluxes of 1,600 g.m<sup>2</sup>.d<sup>-1</sup> to values that in a few days reached 15,000 g.m<sup>2</sup>.d<sup>-1</sup>. This indicated that independently measured parameters like CO<sub>2</sub> soil flux and 'plume' SO<sub>2</sub> flux give the same geochemical indications, and reveal an increase in the volatiles 'masses' involved in the sulfataric degassing processes at Vulcano Island. This huge increase in both of these volatiles, SO<sub>2</sub> and CO<sub>2</sub>, can be interpreted as a magma depressurization of the deep volcanic system.

These contemporaneously recorded variations in CO<sub>2</sub> and SO<sub>2</sub> from the summit degassing processes at Vulcano Island did not highlight any delays linked to their different coefficients of exsolution. This behavior suggests that the degassing of the SO<sub>2</sub> and CO<sub>2</sub> were modulated by the buffering of the hydrothermal system.

This study represents the first continuous dataset of SO<sub>2</sub> fluxes as geochemical parameters acquired in continuous mode directly from the summit crater of Vulcano Island. Longer time series and correlation with other data will help us to better investigate the geochemical processes that are responsible for the plumbing degassing system of Vulcano Island, and to formulate an updated geochemical model for the volcanic apparatus.

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