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Discussions

Sulfur dioxide emissions from Papandayan and Bromo, two Indonesian volcanoes

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Abstract

Indonesia hosts 79 active volcanoes, representing 14 % of all active volcanoes worldwide. However, little is known about their passive degassing into the atmosphere due to isolation and access difficulties. Existing SO_2 emission budgets for the Indonesian archipelago are based on extrapolations and inferences as there is a considerable lack of field assessments of degassing. Here, we present the first SO_2 flux measurements using DOAS for Papandayan and Bromo, two of the most active volcanoes in Indonesia. Results indicate mean SO_2 emission rates of $1.4\,t\,d^{-1}$ from the fumarolic activity of Papandayan and more than 22–$32\,t\,d^{-1}$ of SO_2 released by Bromo during a declining eruptive phase.

1 Introduction

Volcanic degassing into the atmosphere constitutes one of the external expressions of subsurface magmatic and hydrothermal manifestations. Reciprocally, any changes in the chemical and physical properties of the plume are generally symptomatic of modifications in the magmatic reservoir and/or conduits. Over the last ten years, huge amounts of work in the field of volcanic degassing have significantly improved our knowledge of subsurface magmatic processes. The most abundant component in volcanic gas is H_2O (water vapor), followed by CO_2. They constitute between 60 and more than 90 mol % of the volcanic gases in the plume (Shinohara, 2008). Other components constitute only a fraction of the plume, including SO_2, H_2S, HCl, HF, H_2, CO, N_2, Ar and He. However, due to high ambient concentration, it is extremely difficult to estimate the concentrations of volcanic H_2O and CO_2. Investigations instead focus on SO_2, a relatively abundant species in the volcanic plume, typically in third place behind H_2O and CO_2 with around 5 mol % of gas content along with H_2S. SO_2 has a very low background level in the atmosphere and strong identifiable optical absorption features in the ultraviolet (UV) skylight region that offer various options for spectroscopic detection

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in the atmosphere (McGonigle et al., 2003). Furthermore, SO_2 is a readily measurable species, widely recognized as an important and highly desirable component of multidisciplinary volcano monitoring. Many observatories routinely measure SO_2 emission rates in support of their monitoring networks. SO_2 also plays important roles in the atmosphere as it is a source of sulfate aerosols which can strongly influence atmospheric chemistry, atmospheric radiation, the hydrological cycle and the weather, generate acidic precipitation and pollute air quality (Charlson et al., 1992; Jones et al., 2001; Penner et al., 2001; Stevenson et al., 2003). Despite this prominent status, huge uncertainties remain as to the global volcanic SO_2 emission budget, and the distribution of SO_2 volcanic sources worldwide has yet to be determined with precision. Recently, Bani et al. (2012) pointed out that the total SO_2 released from Vanuatu volcanoes represents about 20 % of the global volcanic SO_2 emission budget. This figure has yet to be integrated into global estimates. Furthermore, many volcanoes on earth still rarely have their SO_2 emissions measured, and many have never had their degassing rates evaluated. This is the case in Indonesia where, despite the high number of active volcanoes, SO_2 emission estimates were based on extrapolation and inference rather than field measurements (Nho et al., 1996; Halmer et al., 2002; Hilton et al., 2002).

Here, we present the first SO_2 flux measurements, obtained in June 2011 from two volcanoes in Indonesia – Papandayan and Bromo (Fig. 1). These two volcanoes are among the most active volcanoes in Indonesia and represent two end-member volcanic degassing types: fumarolic emission on Papandayan and open vent degassing from Bromo (Mazot et al., 2007, 2008; Andres and Kasgnoc, 1998; Nho et al., 1996; GVP Bromo – 03/1995 (BGVN 20:03); GVP Bromo – 05/2004 (BGVN 29:05)).

Papandayan is a complex stratovolcano culminating 2665 m a.s.l. with a base diameter of ~ 8 km (Fig. 2) located 45 km south southeast of Bandung city, West Java. The edifice is well known for its 1772 eruption that caused the collapse of the northeast flank, leading to a devastating debris avalanche over 250 km^2 that destroyed about 40 villages and killed nearly 3000 people (Abidin et al., 2006). Papandayan ranks 11th out of the 13 deadliest eruptions on Earth (Blong, 1984). Other eruptions are reported

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for this volcano in 1882, 1923–1927, 1942, 1993 and 2002 (Abidin et al., 2006). The latest eruption in 2002 is well detailed in Abidin et al. (2006) – about 6000 people were evacuated. Magmatic degassing on Papandayan occurs mainly in two fumarolic zones, aligned in a northwest–southeasterly direction and located in a deformed, horseshoe-shaped eastern crater (Fig. 2).

Bromo is located 75 km south of Surabaya, East Java. It occupies the central part of the Tengger caldera – a well-defined and a roughly square structure around 7 km wide (Fig. 2). The caldera rim culminates at more than 2600 m above sea level, and the inner caldera floor is around 2100 m a.s.l. More than 60 explosive eruptions (mainly with VEI = 2) are reported to have occurred on Bromo over the past four centuries (source: GVP). However, in contrast to Papandayan, no causalities were reported except for the two tourists killed during the 2004 eruption after they ventured too close to the volcano. The present-day active crater, through which magmatic degassing occurs, is Bromo's smallest (500 m in diameter) and northernmost crater (Fig. 2).

2 Methods

SO_2 fluxes were measured using a USB2000 ultraviolet spectrometer, and SO_2 column amounts were retrieved by following standard DOAS calibration and analysis procedures (Kraus, 2006; Platt and Stutz, 2008). The spectral range of the spectrometer is 280–400 nm with a spectral resolution of 0.5 nm FWHM. Light entered the spectrometer through a telescope (8 mrad FOV) and via a fiber optic bundle. The Vispec program (http://vispect.sourceforge.net/) was used to field track the volcanic plume. Total integration times of 3 s (exposure time 300 ms, 10 co-added spectra) and 1.6 s (exposure time 200 ms, 8 co-added spectra) were applied to Papandayan and Bromo, respectively. Reference spectra included in the non-linear fit were obtained by convolving high resolution SO_2 (Bogumil et al., 2003) and O_3 (Voigt et al., 2001) cross-sections with the instrument line shape. A Fraunhofer reference spectrum and Ring spectrum, calculated in DOASIS, were also included in the fit. The optimum fitting windows of 302–325 nm

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and 300–320 nm for Papandayan and Bromo respectively were evaluated by obtaining a near random fit residual with minimum deviation. Figures 3 and 5 show examples of the SO_2 fit. Each spectrum position was determined from a continuously recording GPS unit. Wind speeds were obtained using a handheld anemometer at high points, to the east of the Tengger caldera rim for Bromo and about 200 m above the northern fumarole zone on Papandayan. Error estimates for flux calculation (Table 1) were obtained following Mather et al. (2006) and lead to mean relative errors of 30 and 35 % for Papandayan and Bromo respectively. On Papandayan, DOAS SO_2 flux measurements were performed in walking-traverse mode (McGonigle et al., 2002). The spectrometer was carried with the telescope pointing to the zenith while walking across the northern part of the eastern crater (Fig. 2). The plume was drifting to the northwest at the time of measurement. A complementary USB4000 spectrometer was positioned on a fixed mode, operating within the 292–446 nm spectral range and with 0.3 FWHM spectral resolution. SO_2 column amounts were retrieved using the same procedures as for the USB2000. On Bromo, access into the caldera was possible using a 4WD vehicle, so DOAS traverses were done on a vehicle (Fig. 2). During measurement, the wind was from the east forcing the plume partially above the inaccessible relief zone to the west (Fig. 2). To ensure measurements across the entire plume, the telescope was positioned with an inclination of around 30° from the zenith.

3 Results and discussion

The DOAS measurements obtained in this work are summarized in Table 1 while Figs. 4 and 5 display plots of all traverses and static measurements. Non-linear fits of recorded spectra under Bromo and Papandayan highlight strong SO_2 signals in the plume (Figs. 3 and 5) with maximum concentrations largely exceeding 100 ppm m above the background level. It is therefore evident at this stage that Papandayan and Bromo release SO_2 into the atmosphere.

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3.1 Papandayan's SO_2 emission rate

Results indicate that SO_2 emission rate on Papandayan fluctuates between 0.4 and 2.8 $t\,d^{-1}$ with a mean value of 1.4 $t\,d^{-1}$. This fluctuation is consistent with DOAS static measurements (Fig. 4) where the SO_2 column amount increased progressively from ~40 ppm m to ~140 ppm m over a period of 30 min before dropping to the background level in the following 15 min. The SO_2 flux increased accordingly in traverses 1 to 5 and then decreased in traverse 6. Further measurements are required to better delimit Papandayan emissions. However, it is likely that changes in Papandayan's SO_2 emission rate come from subsurface magmatic-hydrothermal processes with regular magmatic gas discharges pumping up to 0.03 kg SO_2 s^{-1} into the atmosphere. In any case, Papandayan's SO_2 contribution to the atmosphere is relatively small compared to other volcanic sources (Andres and Kasgnoc, 1998). Assuming that the DOAS results are representative, this volcano releases only about 500 tons of sulfur dioxide into the atmosphere annually. This low SO_2 release into the atmosphere is expected for fumarolic-type activity. Mazot et al. (2008) provide a compilation of Papandayan gas chemistry, and mean SO_2 concentration (0.11 mol %) is significantly lower than H_2S concentration (0.51 mol %). Assuming that these concentrations are representative, and using the H_2S/SO_2 molar ratio of 4.6, the H_2S emission rate from Papandayan can be estimated at around 3.4 $t\,d^{-1}$, more than twice the SO_2 emission rate. According to experimental studies and thermochemical modeling of volatile partitioning between vapor and liquid in two-phase hydrothermal systems, CO_2 is the most abundant hydrothermal gas followed by H_2S – other sulfurous gases are negligible (Symonds et al., 2001; Drummond and Ohmoto, 1985; Giggenbach, 1980; Reed and Spycher, 1984, 1985; Spycher and Reed, 1989). The strong availability of H_2S suggests the existence of active hydrothermal processes beneath Papandayan's fumarolic activity, and that a portion of the SO_2 released from the magmatic source probably sinks out by hydrolysis ($4SO_2 + 4H_2O = H_2S + 3H_2SO_4$ and $3SO_2 + 2H_2O = S + 2H_2SO_4$) (Holland, 1965) or is trapped by other hydrothermal processes. Consequently, when considering

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SO_2 flux measurements for monitoring purposes, the existence of hydrothermal processes should be taken into account. Alternatively, H_2S may be a good candidate for monitoring as suggested by Symonds et al. (2001) and Aiuppa et al. (2005). In any case, the DOAS measurement results highlight the potential of SO_2 monitoring of this fumarolically active volcano, and the $1.4\,t\,d^{-1}$ of SO_2 released into the atmosphere can henceforth be used as a baseline for future SO_2 flux measurements.

3.2 Bromo's SO_2 emission rate

The SO_2 flux measurements from Bromo vary roughly between $0.7\,t\,d^{-1}$ and $32\,t\,d^{-1}$, but unlike the Papandayan survey, there were no static measurements to support this investigation. Furthermore, despite the strong signal obtained in the SO_2 fit procedure (Fig. 5), all the traverses were not completed (Fig. 5), and SO_2 fluxes for traverses 1, 3 and 5 were dramatically reduced in comparison to traverses 2 and 4. The reason for this disparity was the inclination of the telescope since part of the plume dispersed above the western relief (Fig. 2). Traverses 2 and 4 commenced below the plume but the relief configuration precluded a complete profile across the plume, while traverses 1, 3 and 5 began away from the plume and the telescope inclination was not sufficient to catch the bulk plume concentration when the vehicle reach the relief. The outcome of these DOAS SO_2 flux measurements may not be representative of the volcano's activity at the time of the survey. However, in the configuration described above, the measurements from traverses 2 and 4 are much closer to reality, suggesting an SO_2 flux of more than $22–32\,t\,d^{-1}$. In the past, the SO_2 emission rate of this volcano was estimated during two eruptive periods, on 8–27 March 1995 (6, 22 and $22\,t\,d^{-1}$) (GVP, 03/1995 – BGVN 20:3), and on 14 June 2004 ($200\,t\,d^{-1}$) (GVP, 05/2004 – BGVN 29:05). The SO_2 flux published in Andres and Kagnoc's (1998) well-known paper were derived from the March 1995 COSPEC measurements as no DOAS results exist prior to this work. Our DOAS measurements were carried out in June 2011 after a strong eruptive phase which commenced in November 2010 and persisted until April 2012. Thus, our

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results likely reflect the continuous decline of the eruptive phase. In May 2012, a 2nd DOAS survey was organized on Bromo but, surprisingly, the results showed no SO_2 emission from the active crater (Fig. 6). Bromo is therefore not a persistent source of SO_2 in the atmosphere, as widely thought. However, this volcano has a high frequency of eruptive activity – about one eruption every 6–7 yr since 1804 (http://www.volcano.si.edu/index.cfm), which indicates that it is nevertheless a major contributor of SO_2 to the atmosphere. Regular measurements over a period of 6–7 yr are necessary to better determine the SO_2 emission rate of this volcano. In any case, this work highlights the potential for DOAS traverses on Bromo and encourages systematic DOAS deployment for monitoring and degassing studies given the short periodicity of eruptive events.

4 Conclusion

We present the first DOAS SO_2 flux estimates for Papandayan and Bromo, two of the most active volcanoes in Indonesia. Results indicate mean SO_2 emission rates of 1.4 $t\,d^{-1}$ from Papandayan's fumarolic activity and more than 22–32 $t\,d^{-1}$ of SO_2 released by Bromo during a declining eruptive phase. Results further indicate that Papandayan's SO_2 release is sustained by the regular discharge of gas, although much of the SO_2 is likely trapped by subsurface hydrothermal processes, leading to significant release of H_2S into the atmosphere. Bromo's SO_2 releases appear not to be persistent over time. This volcano is nevertheless a major source of volcanic degassing into the atmosphere given its 6–7 yr cycle of periodic eruptive activity. In contrast, the permanent degassing on Papandayan represents a negligible contribution of SO_2 to the atmosphere outside eruptive periods. Finally, the DOAS measurements obtained on Papandayan and Bromo are very encouraging given the numerous volcanoes in Indonesia whose degassing has never been evaluated. In addition, this work establishes benchmarks for SO_2 flux monitoring on both Bromo and Papandayan.

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Table 1. Estimated SO_2 emission rates for Papandayan and Bromo.

Volcano	State of activity during measurements	Date of measurements	Traverse	Start time (UT) local time = UT +7	Mean measurement distance from sources (km)	Plume width (km)	Average column amount ($mg\,m^{-2}$)	SO_2 flux ($kg\,s^{-1}$)
Papandayan	Degassing through fumaroles	18 Jun 2011	Trav_1	06:47:04	0.6	0.14	37	0.005
			Trav_2	06:51:03	0.5	0.13	85	0.004
			Trav_3	06:56:45	0.6	0.11	110	0.014
			Trav_4	06:59:51	0.5	0.10	114	0.014
			Trav_5	07:04:35	0.5	0.12	200	0.033
			Trav_6	07:08:11	0.5	0.11	209	0.026
							Mean SO_2 emission rate = $1.4 \pm 0.5\,t\,d^{-1}$	
Bromo	Open vent degassing	23 Jun 2011	Trav_1	03:11:58	2.3	0.30	49	0.028
			Trav_2	*03:20:48*	*2.3*	*0.75*	*120*	*0.372*
			Trav_3	03:32:50	2.3	0.14	54	0.008
			Trav_4	*03:37:20*	*2.3*	*0.68*	*79*	*0.255*
			Trav_5	03:49:23	2.3	0.16	30	0.008
							Mean SO_2 emission rate = $27.1 \pm 9.5\,t\,d^{-1}$	

The mean emission rate is deduced from traverses 2 and 4 (in italics) whose profiles are closer to the real degassing of Bromo. Traverses 1, 3 and 5 account for a small portion of the plume (see text for further detail).

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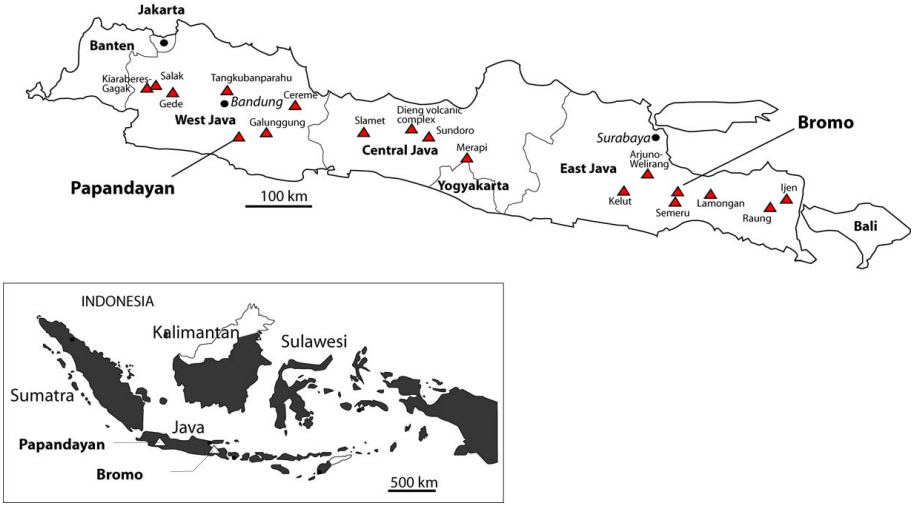

Fig. 1. Java island with its 18 active volcanoes. The target volcanoes, Papandayan and Bromo, are indicated. The location of Java and the studied volcanoes within the Indonesian archipelago is provided below.

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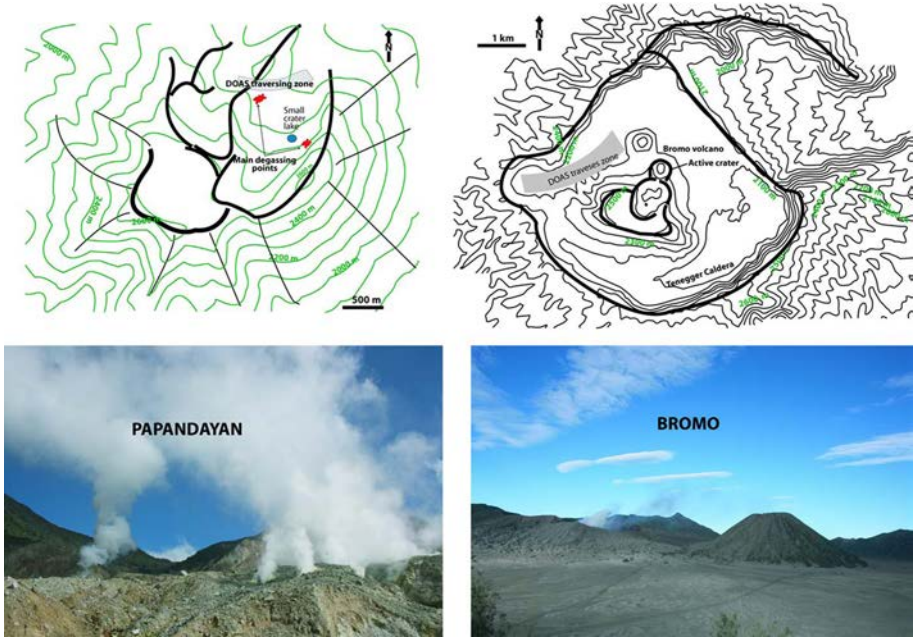

Fig. 2. Maps of the Papandayan summit and the Bromo caldera. The DOAS traverse zones are shaded in gray. The main degassing points are shown: fumarole zones on Papandayan and the active crater on Bromo. Pictures provide a synoptic view of degassing during the field measurements.

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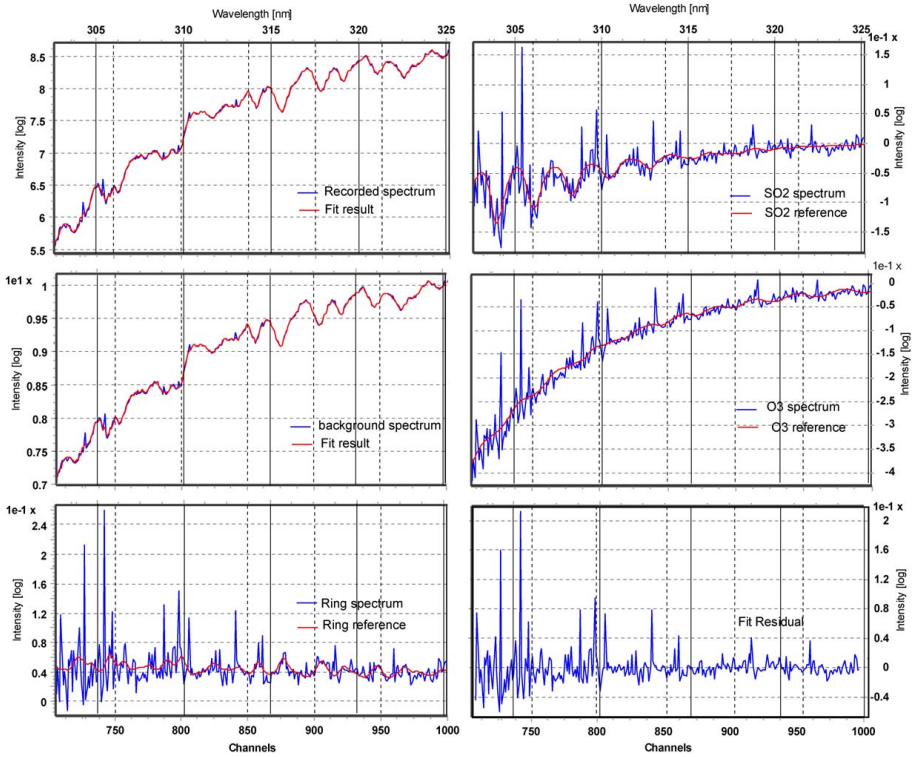

Fig. 3. Example of DOAS SO_2 fit on Papandayan. Blue lines are recorded spectra. The background spectra were acquired by pointing outside the plume.

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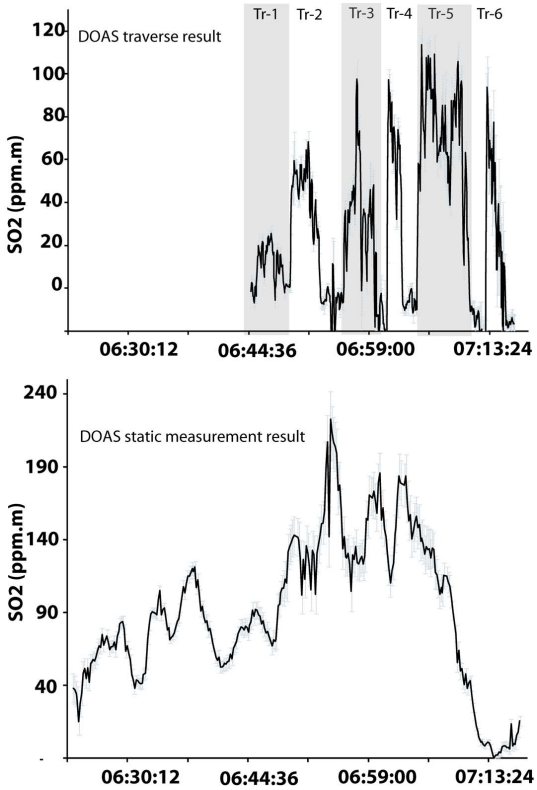

Fig. 4. Plots of traverse (above) and static (below) measurement results obtained on Papandayan. Time axis were aligned highlighting the increase of SO_2 column amounts in both static and traverse measurements.

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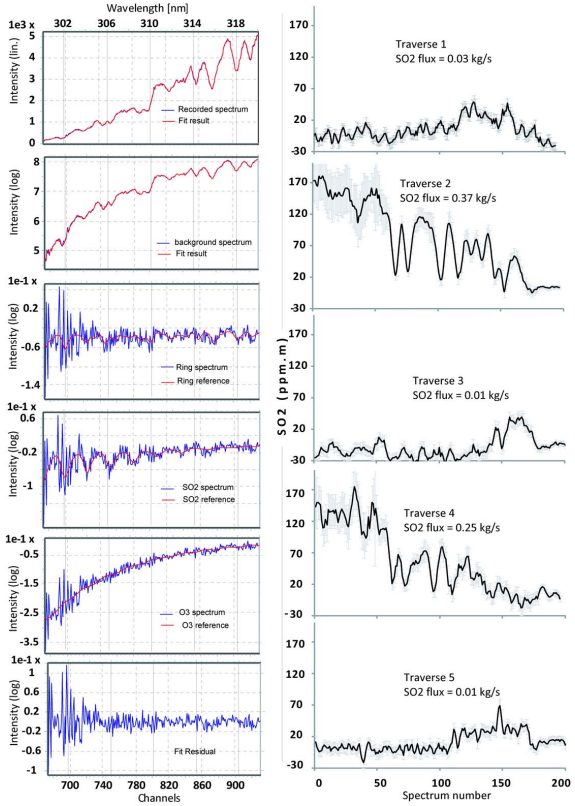

Fig. 5. Example of DOAS SO_2 fit on Bromo (left). Measurement spectra are blue. Traverse measurement profiles (right). Traverses 1, 3 and 5 did not catch the bulk concentration while traverses 2 and 4 commenced in the plume (see text).

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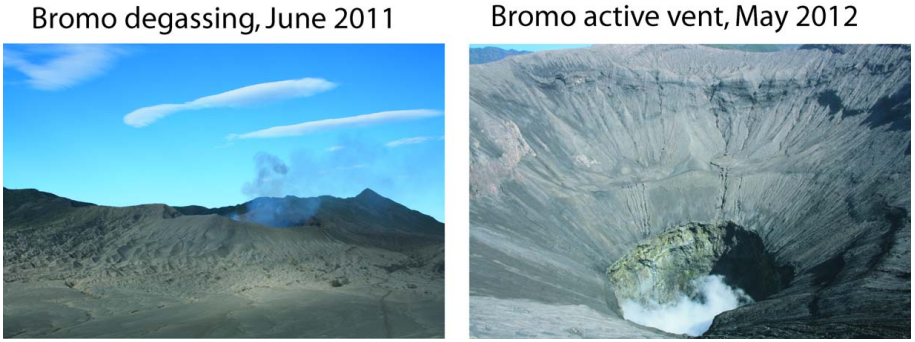

Fig. 6. Bromo degassing observed during two different periods. In June 2011 (left), during measurement, the degassing was clearly visible 2 km from the volcano. In May 2012, there was no plume on Bromo and no SO_2 detected by DOAS. Only a stagnate white vapor was observed in the active crater (right).

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