



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 SO₂ contribution into the atmosphere, due to isolation and access difficulties. Existing SO₂ 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 SO₂ flux measurements using differential optical absorption spectroscopy (DOAS) for Papandayan and Bromo, two of the most active volcanoes in Indonesia. Results indicate mean SO₂ emission rates of 1.4 t d⁻¹ from the fumarolic activity of Papandayan and more than 22–32 t d⁻¹ of SO₂ released by Bromo during a declining eruptive phase. These DOAS results are very encouraging and pave the way for a better evaluation of Indonesian volcanic emissions.

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. Among the volcanic volatile components released into the atmosphere, sulfur dioxide (SO₂) enhances considerable research interest due to its non-negligible roles in the atmospheric chemistry, atmospheric radiation, hydrological cycle and climate, as well as acidic precipitation and air

quality (Charlson et al., 1992; Jones et al., 2001; Penner et al., 2001; Stevenson et al., 2003). It is a relatively abundant species in the volcanic plume, typically in third place behind H₂O and CO₂ with around 5 mol% of gas content along with H₂S (Shinohara, 2008). SO₂ 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 in the atmosphere (McGonigle et al., 2003). SO₂ is thus a readily measurable species, widely recognized as an important and highly desirable component of multidisciplinary volcano monitoring. Many observatories routinely measure SO₂ emission rates in support of their monitoring networks. The global SO₂ emission budget estimates over the last four decades range from 1.5 to 40 Tg yr⁻¹ (Table 1) – a very large range due to the diversity of methodology used, the extending number of studied volcanoes and the increasing development of measurement techniques.

According to the IPCC report (2001), the global volcanic SO₂ emission budget is highly uncertain, because only very few of the potential sources have been measured and the variability between sources and between different stages of activity is considerable. Over the last ten years, the increasing development in the field of remote sensing has improved our knowledge on the distribution of volcanic volatile sources across the earth and even on the remote and less accessible edifices (McGonigle et al., 2004; Mather et al., 2006; Bani et al., 2012; McCormick et al., 2012). But still many volcanoes on earth have never had their degassing rates evaluated. This

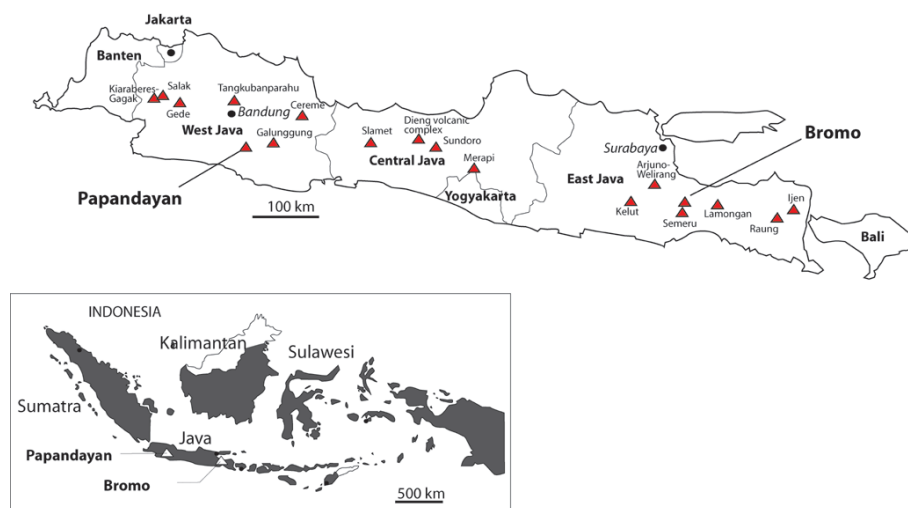


Fig. 1. Java island with its 18 active volcanoes. The target volcanoes, Papandayan and Bromo, are indicated. The location of Java and the studied volcanoes within the Indonesian archipelago is provided below.

is the case in Indonesia where, despite the high number of active volcanoes, the past SO_2 emission estimates were based on extrapolation and inference (Nho et al., 1996; Halmer et al., 2002; Hilton et al., 2002), whilst SO_2 flux measurements were carried out only on 4 volcanoes (Table 1). This present work aims to point out this misrepresentation and further highlight a possibility to constrain the SO_2 emission better from Indonesian volcanoes using differential optical absorption spectroscopy (DOAS).

We present SO_2 flux measurements, obtained in June 2011 from two volcanoes in Indonesia – including Papandayan and Bromo (Fig. 1), two volcanoes among the most active in Indonesia, and they represent two end-members of volcanic degassing types: a fumarolic emission on Papandayan and an open vent degassing from Bromo

2 Papandayan and Bromo volcanoes

Papandayan is a complex stratovolcano culminating 2665 m above sea level with a base diameter of ~ 8 km (Fig. 2) located 45 km south–southeast of Bandung city, West Java. The edifice became well known after its 1772 eruption that caused the collapse of the northeast flank, leading to a devastating debris avalanche over 250 square kilometers 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 have been reported 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) (Mazot et al., 2007, 2008).

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 $\text{VEI}=2$) have been reported to have occurred on Bromo over the past four centuries (source: GVP). However, in contrast to Papandayan, no casualties were reported except for the two tourists killed during the 2004 eruption after venturing 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) (Andres and Kasgnoc, 1998; Nho et al., 1996; GVP Bromo – 03/1995 (BGVN 20:03); GVP Bromo – 05/2004 (BGVN 29:05)).

3 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 (full width at half maximum). 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

Table 1. SO₂ flux contributions from Indonesian volcanoes in the global volcanic emission budgets.

Authors	Global SO ₂ emission budget (Tg yr ⁻¹)	Estimation technique	Contribution from Indonesian volcanoes.
Kellogg et al. (1972)	1.5	SO ₂ emission budget deduced from the volume of erupted lava and assuming that SO ₂ constitutes 0.5 % of the erupted gas.	SO ₂ contribution from Indonesian volcanoes not specified.
Cadle (1975)	7.5	SO ₂ emission budget deduced from the volume of erupted lava and assuming that SO ₂ constitutes 2.5 % of the erupted gas.	SO ₂ contribution from Indonesian volcanoes not specified.
Le Guern (1982)	5–10	SO ₂ budget estimation was based on lava density, gas content and viscosity.	0.073 Tg yr ⁻¹ from Indonesia (only Merapi volcano was considered in this estimation).
Berresheim and Jaeschke (1983)	15.3	SO ₂ emission budget obtained from a classification of volcanic activities in the literature.	SO ₂ contribution from Indonesian volcanoes not specified.
Stoiber et al. (1987)	18.7	Based on 35 volcanoes monitored by COSPEC and questionnaire sent to volcanologists to estimate plume sizes, from which SO ₂ burdens were calculated.	SO ₂ contribution from Indonesian volcanoes not specified.
Spiro et al. (1992)	19.2	SO ₂ budget estimation derived from the Volcanic Explosivity Index (VEI) with data from Simkin et al. (1981) and as function of location (at convergent plate margin or not). For some volcanoes, estimations were derived from plume size.	0.41 Tg yr ⁻¹ attributed to Indonesian volcanoes.
Andres and Kasgnoc (1998)	13	Data from TOMS and COSPEC, as well as data from journals, conferences and personal communication.	Indonesian volcanoes contributed only 0.10 Tg yr ⁻¹ (3 volcanoes were considered, including Merapi, Tangkuban Parahu, Bromo and Slamet).
Graf et al. (1997)	34.9	SO ₂ emission budget derived from atmospheric circulation–chemistry model.	SO ₂ emissions not specified for Indonesia. However the authors indicate strong sulfate burden over the archipelago.
Halmer et al. (2002)	15–21	SO ₂ emission budget based on 360 explosively degassing subaerial volcanoes, of which 50 were monitored directly by TOMS and/or COSPEC; the sulfur emission of the remaining 310 volcanoes was extrapolated.	2.1–2.6 Tg yr ⁻¹ attributed to Indonesia subduction zone.
Diehl et al. (2012)	26	SO ₂ emission budget is derived using the Volcanic Sulfur Index (VSI) and based on 1167 volcanoes considered to be active in the Global Volcanism Program from 1979 to 2009. Data are replaced by specific observation from TOMS and OMI when available or in some cases with COSPEC measurements and more detailed analyses from open literature.	SO ₂ emissions not specified for Indonesian archipelago.

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 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 SO₂

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. On this latter volcano, DOAS SO₂ flux measurements were performed in

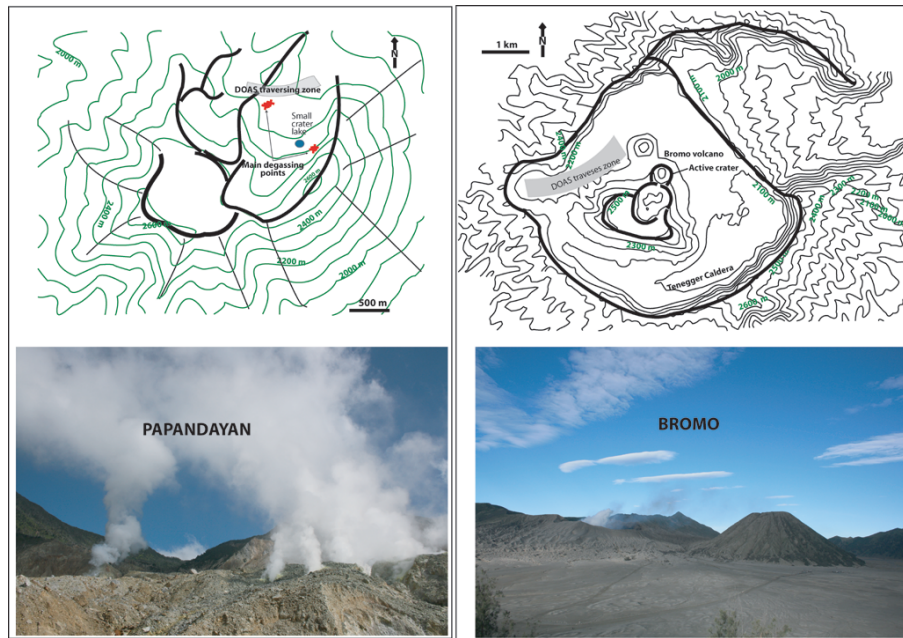


Fig. 2. Maps of the Papandayan summit and the Bromo caldera. The DOAS traverse zones are shaded in gray. The main degassing points are shown: fumarole zones on Papandayan and the active crater on Bromo. Pictures provide a synoptic view of degassing during the field measurements.

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.

3.1 Errors in the SO_2 flux measurements

Error in the SO_2 flux measurements is derived from four different factors, including the retrieved column amount, the distance perpendicular to the plume transport direction, the angle between the assumed wind direction and the traverse path and the plume transport speed (Mather et al., 2006). Error in the retrieved SO_2 column amount depends on many factors (Stutz and Platt, 1996; Hausmann et al., 1999; Kern et al., 2010), but we assume that the dominant error is induced by variable cloudiness that we compensate using artificial constant dark, calculated from each recorded spectrum, in the range of blind pixel (pixel below 290 nm) (Tsanev, 2008). Such corrections account for dark spectrum, offset and stray

light. We estimate that the error in the column amount contributes 0.006–0.014 to the squared variation coefficient of the total flux, whilst error contributions from the distance traversed perpendicular to the plume and from the assumed wind direction following the approached detailed in Mather et al. (2006) are 0.001–0.006 and ~ 0.003 respectively. Note however that all these errors are negligible in comparison to uncertainties in the plume speed (e.g., Stoiber et al., 1983). We assumed that the plume transport speed is conservative throughout our measurements period with a relative error of ~ 30 – 35% , consistent with Stoiber et al. (1983).

4 Results and discussion

The DOAS measurements obtained in this work are summarized in Table 2, 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.

4.1 Papandayan's SO_2 emission rate

Results indicate that SO_2 emission rate on Papandayan fluctuates between 0.4 ± 0.1 and $2.8 \pm 0.8 \text{ t d}^{-1}$ with a mean value of $1.4 \pm 0.5 \text{ t d}^{-1}$. This fluctuation is consistent with DOAS static measurements (Fig. 4) where the SO_2 column amount

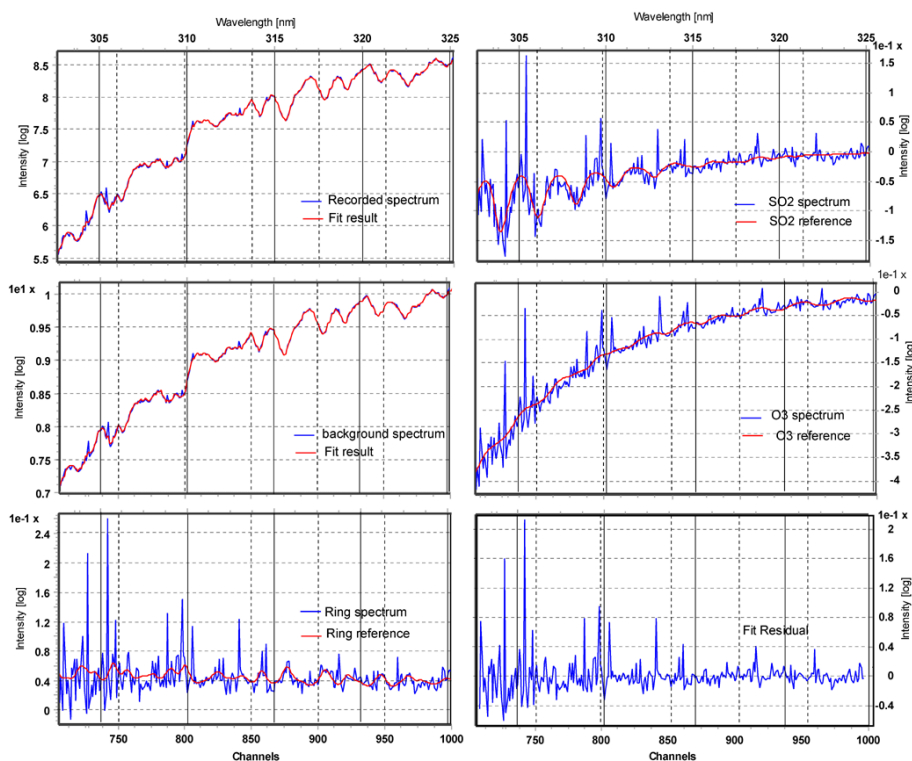


Fig. 3. Example of DOAS SO₂ fit on Papandayan. Blue lines are recorded spectra. The background spectra were acquired by pointing outside the plume.

Table 2. Estimated SO₂ 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 (t d ⁻¹)
Papandayan	Degassing through fumaroles	18/06/11	Trav_1	06:47:04	0.6	0.14	37	0.4 ± 0.1
			Trav_2	06:51:03	0.5	0.11	110	1.2 ± 0.4
			Trav_3	06:56:45	0.6	0.11	110	1.2 ± 0.4
			Trav_4	06:59:51	0.5	0.10	114	1.2 ± 0.4
			Trav_5	07:04:35	0.5	0.12	200	2.8 ± 0.8
			Trav_6	07:08:11	0.5	0.11	209	2.2 ± 0.7
							Mean SO ₂ emission rate = 1.4 ± 0.5 t d ⁻¹	
Bromo	Open vent degassing	23/06/11	Trav_1	03:11:58	2.3	0.30	49	2.4 ± 0.8
			Trav_2	03:20:48	2.3	0.75	120	32.1 ± 11.2
			Trav_3	03:32:50	2.3	0.14	54	0.7 ± 0.2
			Trav_4	03:37:20	2.3	0.68	79	22.0 ± 7.7
			Trav_5	03:49:23	2.3	0.16	30	0.7 ± 0.2
							Mean SO ₂ emission rate = 27.1 ± 9.5 t d ⁻¹	

The mean emission rate is deduced from traverses 2 and 4 (in bold), 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).

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 SO₂ flux increased accordingly in traverses 1 to 5 and then decreased in traverse 6. Further measurements are required to delimit Papandayan emissions better. However, it is likely that changes in Papandayan’s SO₂ emission rate come from subsurface magmatic–hydrothermal processes with regular magmatic

gas discharges pumping up to 0.03 kg SO₂ s⁻¹ into the atmosphere. In any case, Papandayan’s SO₂ 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 SO₂ release into the atmosphere is expected for fumarolic-type activity. Mazot et al. (2008) provide a

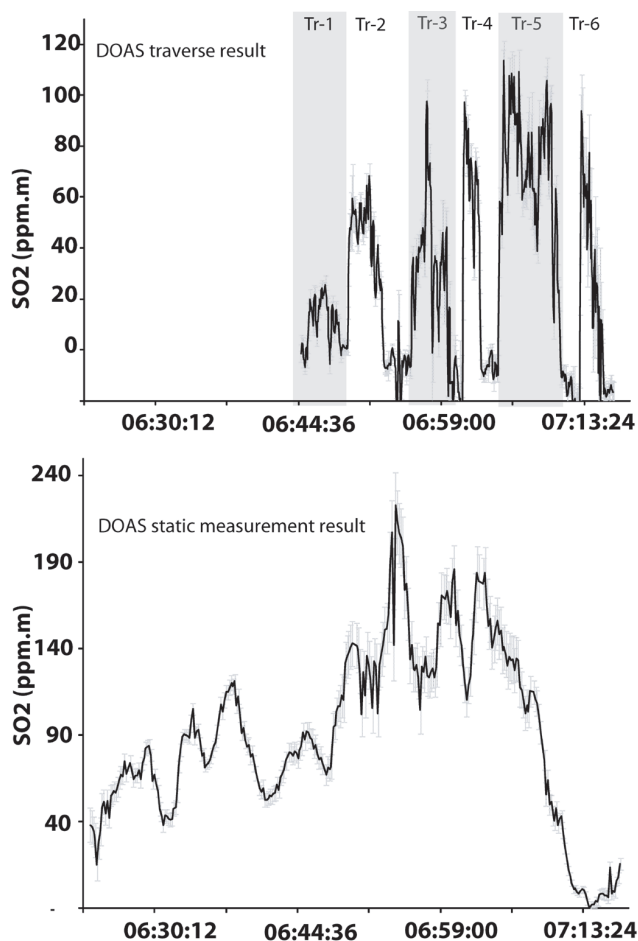


Fig. 4. Plots of traverse (above) and static (below) measurement results obtained on Papandayan. Time axes were aligned highlighting the increase of SO₂ column amounts in both static and traverse measurements.

compilation of Papandayan gas chemistry, and mean SO₂ concentration (0.11 mol%) is significantly lower than H₂S concentration (0.51 mol%). Assuming that these concentrations are representative, and using the H₂S/SO₂ molar ratio of 4.6, the H₂S emission rate from Papandayan can be estimated at around 3.4 t d⁻¹, more than twice the SO₂ emission rate. According to experimental studies and thermochemical modeling of volatile partitioning between vapor and liquid in two-phase hydrothermal systems, CO₂ is the most abundant hydrothermal gas followed by H₂S – 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 H₂S suggests the existence of active hydrothermal processes beneath Papandayan's fumarolic activity, and that a portion of the SO₂ released from the magmatic source probably sinks out by hydrolysis ($4\text{SO}_2 + 4\text{H}_2\text{O} = \text{H}_2\text{S} + 3\text{H}_2\text{SO}_4$ and $3\text{SO}_2 + 2\text{H}_2\text{O} = \text{S} + 2\text{H}_2\text{SO}_4$) (Holland, 1965) or is trapped by other hydrothermal processes.

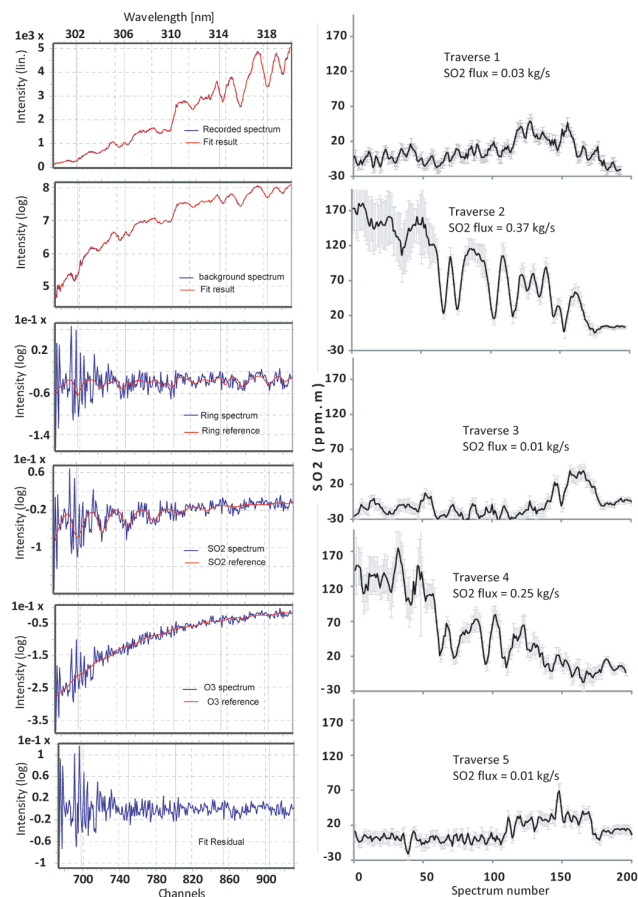


Fig. 5. Example of DOAS SO₂ fit on Bromo (left). Measurement spectra are blue. Traverse measurement profiles are shown (right). Traverses 1, 3 and 5 did not catch the bulk concentration while traverses 2 and 4 commenced in the plume (see text).

Consequently, when considering SO₂ flux measurements for monitoring purposes, the existence of hydrothermal processes should be taken into account. Alternatively, H₂S 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 SO₂ monitoring of this fumarolically active volcano, and the 1.4 t d⁻¹ of SO₂ released into the atmosphere can henceforth be used as a baseline for future SO₂ flux measurements.

4.2 Bromo's SO₂ emission rate

The SO₂ flux measurements from Bromo vary roughly between $0.7 \pm 0.2 \text{ t d}^{-1}$ and $32 \pm 11.2 \text{ t d}^{-1}$, but unlike the Papandayan survey, there were no static measurements to support this investigation. Furthermore, despite the strong signal obtained in the SO₂ fit procedure (Fig. 5), all the traverses were not completed (Fig. 5), and SO₂ 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



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 from Bromo and no SO₂ detected by DOAS. Only a stagnate white vapor was observed in the active crater (right).

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 reached the relief. The outcome of these DOAS SO₂ 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 SO₂ flux of more than 22–32 t d⁻¹. In the past, the SO₂ 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 20:03) and on 14 June 2004 (200 t d⁻¹) (GVP, 05/2004 – BGVN 29:05). The SO₂ fluxes 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 that commenced in November 2010 and persisted until April 2012. Thus, our results likely reflect the continuous decline of the eruptive phase. In May 2012, a second DOAS survey was organized on Bromo, but, surprisingly, the results showed no SO₂ emission from the active crater (Fig. 6). Bromo is therefore not a persistent source of SO₂ 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 SO₂ to the atmosphere. Regular measurements over a period of 6–7 yr are necessary to determine the SO₂ emission rate of this volcano better. 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.

5 Conclusions

We present the first DOAS SO₂ flux estimates for Papandayan and Bromo, two of the most active volcanoes in Indonesia. Results indicate mean SO₂ emission rates of 1.4 t d⁻¹ from Papandayan's fumarolic activity and more than 22–32 t d⁻¹ of SO₂ released by Bromo during a declining eruptive phase. Results further indicate that Papandayan's SO₂ release is sustained by the regular discharge of gas, although much of the SO₂ is likely trapped by subsurface hydrothermal processes, leading to significant release of H₂S into the atmosphere. Bromo's SO₂ 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 SO₂ 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 SO₂ flux monitoring on both Bromo and Papandayan.

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