A Compact Ultraviolet Spectrometer System (COMPUSS) for Monitoring Volcanic SO₂ Emission: Validation and Preliminary Observation.

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A compact and handy system for measuring SO₂ fluxes from volcanoes is newly built using a miniature UV spectrometer. This COMPact Ultraviolet Spectrometer System (Hereafter referred as "COMPUSS") is much smaller and lighter than a correlation spectrometer (COSPEC), which has been used for SO₂ flux measurements from volcanoes over 30 years. Validation of the COMPUSS was carried out comparing COMPUSS and COSPEC instruments at Sakurajima and Miyakejima volcanoes by car and heliborne traverse method, respectively. Our results show that agreement of the SO₂ flux between the COSPEC and the COMPUSS data is vouched for traverse measurements. The COMPUSS has much advantage compared to the COSPEC for volcanic SO₂ flux measurements, because of its portability and possibility for further improvement of the system.

Key words: SO₂ flux, DOAS, UV spectrometer and COSPEC

1. Introduction

For the last 30 years, correlation spectrometer (COSPEC) has been used as the only method for ground-based remote sensing to measure sulfur dioxide emission rate from volcanoes and had a prominent contribution on the progress in recent volcanology. The COSPEC was first designed for monitoring industrial emission of SO₂ and NO₂ in 1960's (Hoff and Millan, 1981) and later used for monitoring volcanic SO₂ emissions by various researchers (e.g., Moffat and Millan,1971; Stoiber and Jepsen, 1973). Integrated COSPEC measurements showed that correspondence of SO₂ flux with magma discharge rates at Unzen volcano, Japan (Hirabayashi *et al.,* 1995) or with seismicity and ground deformation at Soufriere Hills volcano, Montserrat (Watson *et al.,* 2000).

In the beginning of this century, a remarkably small miniature ultraviolet spectrometer was introduced and used successfully to volcanic SO₂ flux measurements (Galle *et al.,* 2002). This compact and low-cost UV spectrometer system offered a chance to carry out various kinds of new monitoring methods. Owing to its compactness and portability, the instrument enabled us to make walking traverse measurements (McGonigle *et al.,* 2002; Oppenheimer *et al.,* 2004). At Soufriere Hills volcano, continuous (day-time) measurements of SO₂ flux is carried out using two scanning systems with the miniature UV spectrometer (Edmonds *et al.,* 2003). Several groups developed new methods for measuring the plume speed, which has been always a fundamental problem for flux estimation, by the use of the miniature UV spectrometer (McGonigle *et al.,* 2005; William-Jones *et al.,* 2006). Under current situation, each researcher builds their own systems using the spectrometer (Galle *et al.,* 2002; Horton *et al.,* 2006; McGonigle *et al.,* 2002). In 2002, we started to develop a Japanese version of the miniature UV spectrometer system. The COMPUSS and its operating software are specially designed focusing on easy handling for measurements and on availabil-

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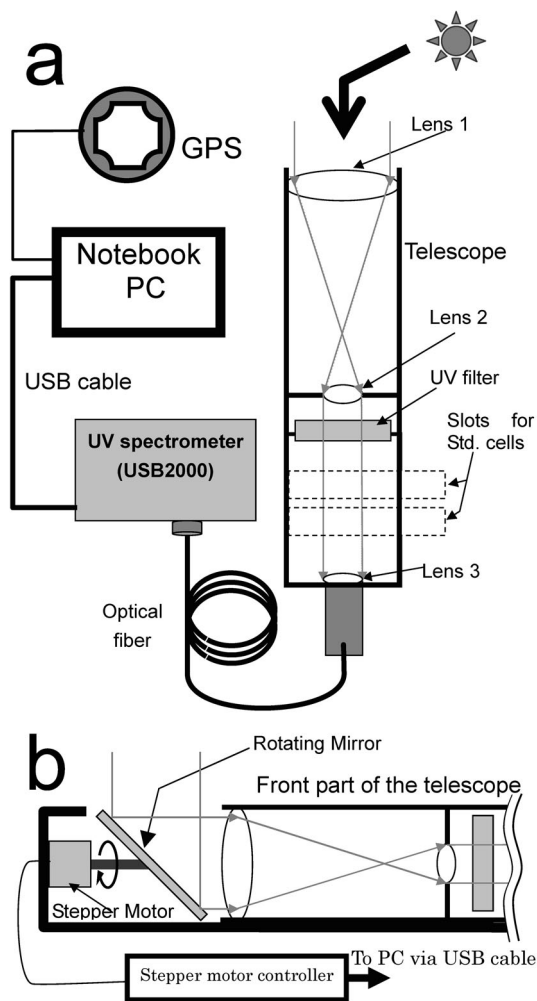Michiko Ohwada, Makoto Miyashita, Hideki Iino, Yasuhiro Nakahori

Fig. 1. (a) System configuration of the COMPUSS for traverse measurements. The system consists of mainly four parts: a UV telescope, a UV spectrometer (USB2000, Ocean Optics Inc.), a Note PC and a GPS unit. (b) The rotating mirror unit attached to the front part of the telescope in the scanning measurements. The stepper motor is connected to the controller, which is controlled by a notebook PC.
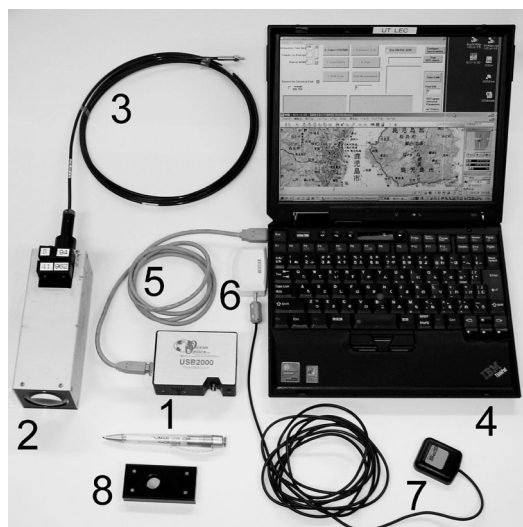

Fig. 2. A photo of the COMPUSS. (1) UV spectrometer (USB2000). (2) Telescope. (3) Optical fiber; the other end should be connected to the UV spectrometer. (4) Notebook computer. (5) USB cable. (6) GPS receiver. (7) GPS antenna. (8) Calibration Cell. Two calibration cells are inserted into the slots of the telescope. A ballpoint pen is placed below the spectrometer for size comparison.

2. Instrumentation

The compact UV spectrometer system (COMPUSS) which we developed for SO_2 column amount measurements is based on USB2000 ultraviolet spectrometer (Ocean Optics Inc.) as described by McGonigle *et al.* (2002) and Galle *et al.* (2002). The COMPUSS for traverse measurements is basically composed of four parts: the USB2000 spectrometer, a UV telescope, a computer and a GPS unit (Fig. 1a). The spectrometer features a $50\,\mu$m entrance slit and the wavelength range is adjusted to 250–400 nm, which is focused to measure the SO_2 absorption at 305–320 nm. The resulting wavelength resolution is about 0.6 nm for the whole wavelength range. The lightweight (about 0.2 kg) and compact size ($89\times64\times34$ mm) of the spectrometer enables the total system to be lighter and more compact compared to the COSPEC system. The spectrometer can be powered by USB bus power from a notebook computer and consumes 450 mW. The spectroscopic performance of USB2000 spectrometers are described in detail in Galle *et al.* (2002).

The system employs a telescope ($60\times60\times165$ mm) built with three quartz lenses (Figs. 1a and 2). The scattered UV light from the sky is collected by the first lens (5 cm diameter, focal length: 60 mm), and is then

ity for simple in-field calibration of the system. We believe that the COMPUSS and similar systems will replace the COSPEC for volcanic SO_2 flux measurements in the near future. The aim of this paper is to introduce the instrumentation and the spectral analyzing method of the COMPUSS and to show the results of comparative measurements with the COSPEC.

collimated by the second lens (2 cm diameter, focal length: 15 mm). The third lens (1 cm diameter, focal length: 30 mm) focuses the light into an optical fiber connected to the telescope. A UV filter designed to partially relieve the rapid increase in skylight intensity from 310 nm to 330 nm is inserted between the second and the third lenses (Figs. 1a and 3a). The field-of-view (FOV) of the telescope is 6.7 mrad using the 800-μm-diameter optical fiber ($\sim$1.5 m length), which connects the spectrometer and the telescope. Two slots between the second and third lenses allow insertion of one to two SO$_2$ calibration cells. This enables in-field calibration for SO$_2$ retrievals and confirmation of the system operation. Existence of the two slots for the calibration cells is one of the advantages of the COMPUSS compared to other miniature UV spectrometer systems. By having n SO$_2$ calibration cells with different SO$_2$ column amounts, it is basically possible to have $n!$ different SO$_2$ column amounts for the calibration. This not only ensures reliability of the calibration but also enables calibration for wider column amount range.

A notebook PC is used for spectral acquisition, real-time in-situ analyses and display of SO$_2$ column amount. Software used for controlling the spectrometer and analyzing the observed spectra is designed by our group considering simple operation for measurements and calibration in the field. The position of the COMPUSS can be logged and displayed simultaneously using GPS and a freeware mapping program, Kashmir3D (http://www.kashmir3d.com/index-e.html). The GPS's coordinate data are used for the SO$_2$ flux evaluations. The total weight of the traverse system is less than 1 kg without the weight of the PC. The COMPUSS can usually be operated for 1.5–4 hours with the notebook computer's battery.

Several additional equipments are needed for the scanning measurements: a rotating mirror unit, a stepper motor controller (size: 200$\times$250$\times$100 mm, weight: 700 g) and an extra 12V battery. The rotating mirror unit can be easily attached in front of the COMPUSS's telescope (Fig. 1b). A mirror with 45° angle to the light axis of the telescope is connected to the stepper motor. This configuration is similar to the scanning configuration reported by Edmonds *et al.* (2003). The rotating mirror unit enables the scanning measurements of the plume by rotating the mirror with controlled speed. The PC and the controller box are connected via USB cable in order to link the rotation and the spectra acquisitions. The scanning system works more than 6 hours using 4Ah 12V battery (about 2 kg), thus, the total weight of the system without the laptop computer adds up to $\sim$5 kg.

3. Spectrum acquisitions and evaluations

In order to obtain sufficient spectral intensity we need to select proper integration time of the spectra, or an exposure time for a CCD detector of the USB2000. Since the SO$_2$ peaks focusing on are in 305–320 nm range, integration time should be selected to maintain sufficient spectral intensity (at least several hundred counts intensity) at around 305 nm without signal saturation at 320 nm. In normal conditions, 100 ms (midday in summer)—1500 ms (late afternoon in winter) are used as the integration time depending on the weather, time and seasonal conditions. In the field measurements, preferable time interval for obtaining SO$_2$ column amounts is usually 1 to 4 seconds depending on the required spatial resolution for the measurements. Several spectra are averaged to reduce the noise of the observed spectra before spectral analyses. The SO$_2$ column amount in the plume can be obtained from absorbance of the plume, which can be expresses as

$$A_{\text{plume}}(\lambda) = -\log((P_{\text{plume}}(\lambda) - P_{\text{dark}}(\lambda))$$
$$/(P_{\text{reference}}(\lambda) - P_{\text{dark}}(\lambda))), \qquad (1)$$

where $A_{\text{plume}}(\lambda)$ is the absorbance of the plume at wavelength λ. P_{plume}, $P_{\text{reference}}$ and P_{dark} are spectral intensity for the plume, reference and dark spectra, respectively. The dark spectrum is background electrical current of the CCD inside the spectrometer and the reference spectrum is the spectrum obtained for no plume sky. The dark spectrum is obtained by covering the telescope. The reference spectrum (Fig. 3b) is obtained by aiming the no plume sky with the telescope. Since both the plume and reference spectra depend on the sky light and the atmospheric attenuation, absorbance of the plume calculated above provides attenuation (absorption and scattering) by the plume. SO$_2$ column amount can be calculated by means of differential optical absorption spectroscopy (DOAS) technique, which is widely used in atmospheric UV spectroscopy (Platt, 1994; Platt and Perner, 1983). The advantage of the DOAS technique is that interference of scattering can be reduced in the spectral analyses. McGonigle *et al.* (2002) and Galle *et al.* (2002) calculated SO$_2$ column amount from the spectra by fitting a library differential absorption spectrum of SO$_2$ to the observed differential absorption spectrum, using least squares procedure. In contrast, peak and trough height of five SO$_2$ peaks between 305 and 318 nm (between the arrows in Fig. 3 c) are used in our evaluation method for COMPUSS, for simple and in-situ real-time calculation of the SO$_2$ column amount. The peak heights for respective peaks are calculated using the vertical scale difference between peak top and a line segment bonding the troughs on the both side of the peak. This approach of using peak heights for SO$_2$ evaluation is similar with FLYSPEC's

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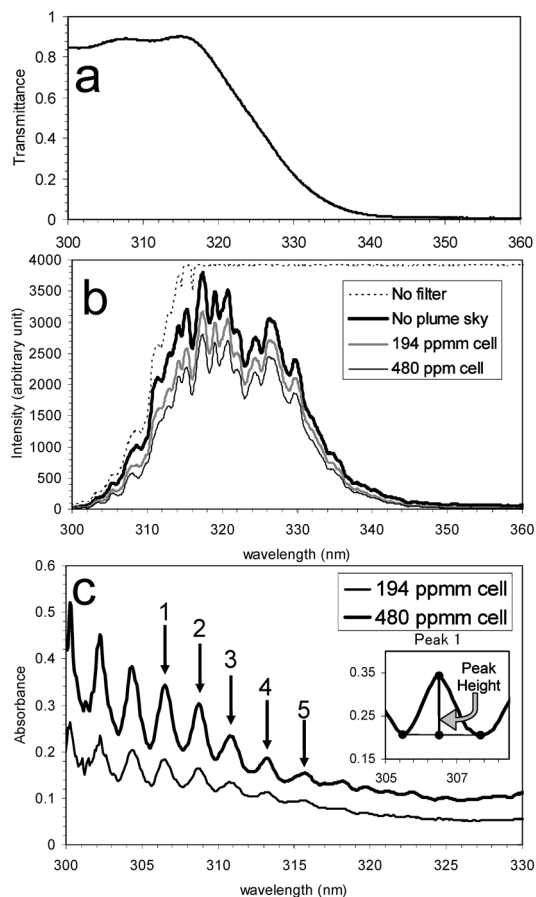

Fig. 3. (a) Transmittance of the UV filter inside the telescope between 300 nm and 360 nm. (b) Examples of the raw spectra obtained using the COMPUSS. The spectra are obtained on a clear winter day aiming zenith. The spectra (dark corrected) from the top are sky spectrum without the UV filter (dotted line), sky spectrum with the UV filter (reference spectrum: solid bold line) and two sky spectra with 194 ppmm (gray bold line) and 480 ppmm (thin solid line) calibration cells. (c) Absorbance of the two calibration cells calculated using spectra in (b). Peak tops of the five peaks used in the quantification were indicated with peak numbers and arrows. The absorbance between 300 nm and 303 nm are slightly distorted due to low spectral intensity as seen in (b). The inset in (c) shows a schematic for calculating the peak height for the peak 1.

evaluation method (Horton *et al.,* 2006). In COMPUSS's case, peak heights of 5 peaks are independently calculated and then converted to SO_2 column amounts by applying calibration parameters. The representative SO_2 column amount is obtained by averaging the results of several peaks. The operating software for the COMPUSS displays and records five SO_2 column amount values calculated using respective five peaks together with the representative value. This idea of dealing with five peaks independently is a unique feature of the COMPUSS operating software and can use as an indicator for presence of scattering interference by atmosphere between the plume and the COMPUSS.

The noise level of SO_2 column amount is usually from a several ppmm to some tens of ppmm. The noise level basically depends on the spectroscopic intensity, and number of spectra to average to obtain the sample spectra. Thus, by increasing the integration time and number of spectra to average, apparent SO_2 error can be minimized. However, this will decrease the time resolution as well as spatial resolution. Conversion factors to translate from the peak heights to SO_2 column amounts were obtained using SO_2 calibration cells prior to measurements. We also put the calibration cells between the traverse measurements to check stability of the measurements.

4. Results and discussion

In order to verify the newly built system, SO_2 standard cells with various SO_2 column amount are tested in the field. We also show the COMPUSS's results together with COSPEC's results at two Japanese volcanoes. Comparison of the results with COSPEC's results is an important issue to check and maintain the data quality and continuity for accumulated flux data over several decades using the COSPEC.

4‑1 Response validation with SO_2 calibration

The response validation for the COMPUSS was carried out on a clear day by putting the telescope on a tripod aiming zenith. Using the two slots of the telescope (Fig. 1a), variety of SO_2 column amounts are obtained with five cells, which are calibrated in the laboratory, by using one or two cells at each measurement. Figures 4a and 4b show correlations between the peak height and column amount of SO_2 synthesized with the standard cells for SO_2 peaks at 308.6 nm (Peak 2 in Fig. 3c) and 313 nm (Peak 4 in Fig. 3c), respectively. The linear or quadratic relations between the peak height and the column amount are obtained for all the peaks with correlation coefficients (r^2) over 0.99 between 0 and 2700 ppmm of SO_2. In practical use for volcanic plume monitoring, the SO_2 column amount range used here is basically enough and the results of the test show the capability of the system at least up to 2700 ppmm. This field calibration shows the advantage of
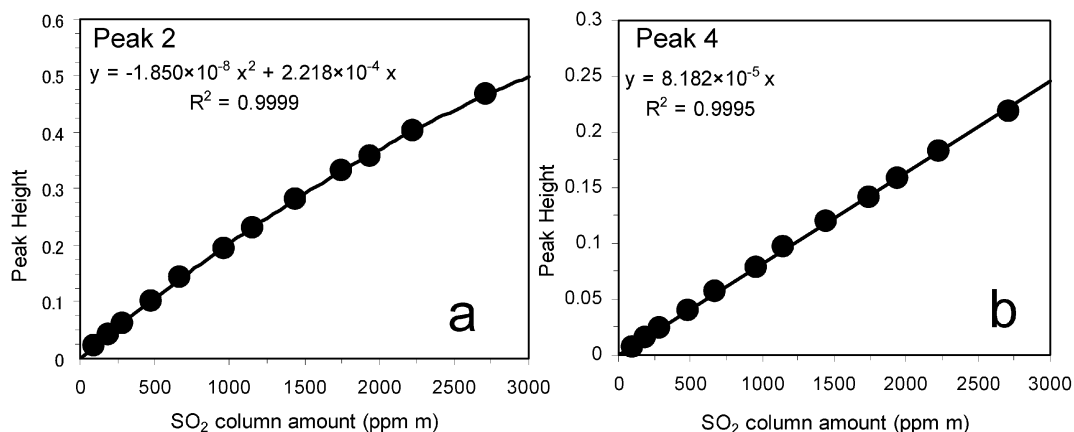
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Fig. 4. Example of plots between the peak height and the column amount of SO_2 for absorbance peaks for (a) Peak 2 and (b) Peak 4 in Fig. 3c. The regression curve and line are calculated by the least-squares-method.

the COMPUSS having two slots for the calibration cells.

4–2 Traverse measurements at Sakurajima Volcano

Sakurajima volcano locates in the southern part of Kyushu Island, Japan. The volcano emits SO_2 with an average of 2000 ton/day for last several decades (Hirabayashi *et al.,* 1998). In October 2003, a comparison measurement using three COMPUSSes was carried out at Sakurajima volcano. Three COMPUSSes (C1, C2 and C3) were mounted on three different cars aiming vertically up. Traverse measurements are made by taxiing cars under the plume in C1, C2 and C3 order. We also put the COSPEC in the last car for comparison with the C3 COMPUSS. Figure 5a shows a time versus SO_2 column amount plot of the C3 COMPUSS together with that of the COSPEC. Unlike SO_2 output by the COMPUSS, SO_2 variation obtained by the COSPEC has many spike-like noises. These spike-like noises are due to a brief covering of the FOV by trees in the case of COSPEC's output. In contrast, these obstructions in the optical path do not affect the SO_2 output with the COMPUSS, as long as the duration of the FOV covering is shorter than the integration time of the spectra acquisition which is usually a few hundred to a thousand milliseconds. When the traverse route is in the sparse woodlands with some sky light through the trees, COSPEC measurements are impossible due to the large noise.   In contrast, the COMPUSS measurements are still possible as long as there is certain amount of sky light intensity at the SO_2 absorption wavelength range. This fact expands the opportunity of measuring SO_2 flux with various conditions compared to the case of the COSPEC.

Figure 5b shows a time versus SO_2 column amount plot for three COMPUSSes. The three profiles basically have high similarity except at a few places. In the traverse measurement, we tried to keep distance be-

Table 1. Correlation and proportional coefficients of the SO_2 column amounts obtained by the three COMPUSS and the COSPEC.

Date	Pairs	Correlation Coeff. (r^2)	Proportional Coeff.
2003.10.20	C3-COSPEC	0.983	0.990
	C1-C2	0.983	1.021
	C1-C3	0.983	1.065
	C3-C2	0.996	0.959
2003.10.21	C1-C2	0.984	0.992
	C1-C3	0.985	1.019
	C3-C2	0.981	1.018

tween the three cars to be 10–50 m apart from each other which corresponds to average of 1–5 seconds time differences between the cars. However, the distance occasionally stretched over 100 m, due to interference of the local traffic. The time difference likely caused the slight disagreement in the three measurements. The SO_2 fluxes measured with the three COMPUSSes were 772, 697 and 711 ton/day for C1, C2, and C3, respectively. On 21 Oct. 2003, further evaluation measurements were carried out with three COMPUSSes (Fig. 5c). We drove more carefully than the previous day to keep the distance between cars more constant. The SO_2 fluxes measured with the three COMPUSSes were 536, 538 and 540 ton/day for C1, C2, and C3, respectively.

Table 1 shows correlation and proportional coefficients between outputs of the three COMPUSSes and the COSPEC for 20 and 21 October results. The correlation and proportional coefficients between the three COMPUSSes (Table 1) are calculated by correcting the time differences of a few seconds between the cars. For the COSPEC data, the spike-like noises are precluded from the COSPEC chart. High correlation ($r^2=0.983$) with proportional coefficient of 0.990 is
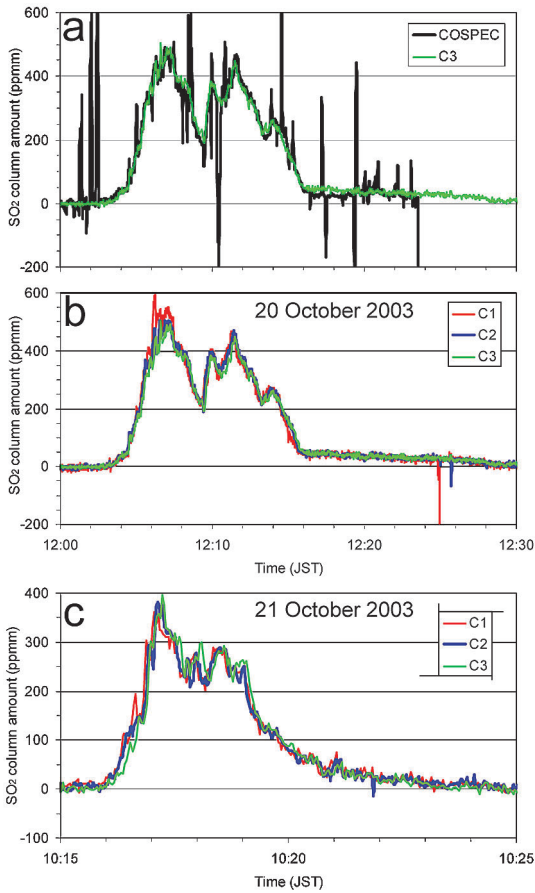
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Fig. 5. Plots of SO_2 column amounts versus time obtained by the car traverse measurements at Sakurajima. (a) COMPUSS (C3: green line) and COSPEC (black line) data obtained on 20 Oct. 2003. (b) Comparison of three COMPUSS on 20 Oct. 2003. The three COMPUSSes were on the different car taxiing with a few seconds difference in the order of C1 (red line), C2 (blue line) and C 3 (green line). (c) Comparison of three COM-PUSSes on 21 Oct. 2003.
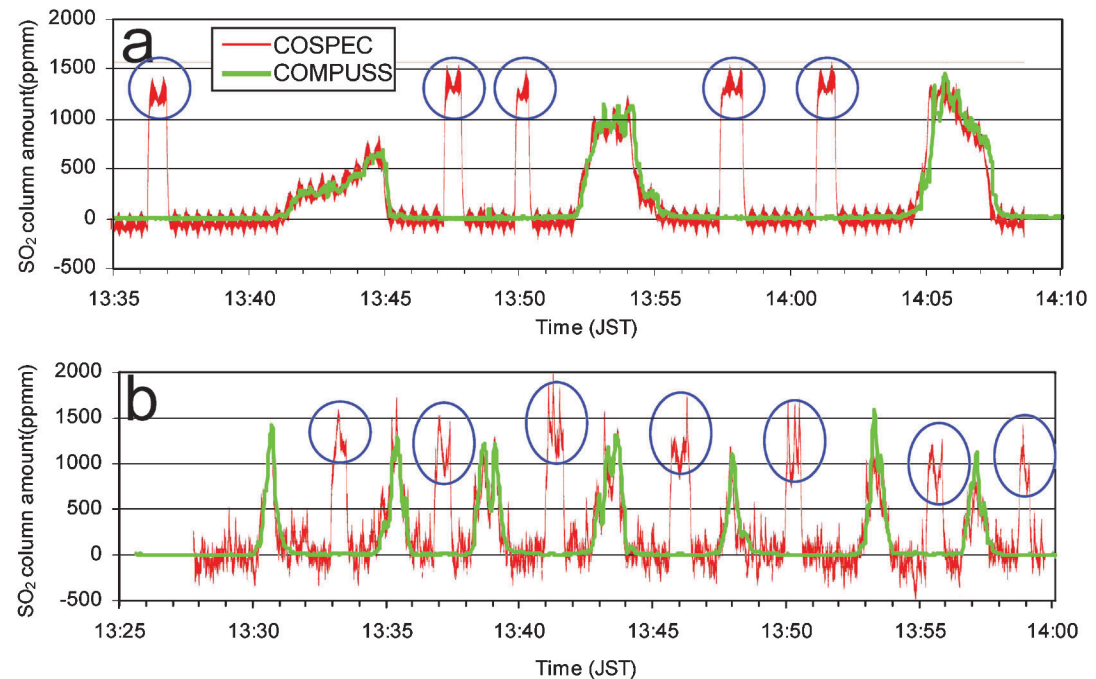
Fig. 6. Plots of SO_2 column amounts versus time obtained by the helicopter traverse measurements with COSPEC (thin red line) and the COMPUSS (bold green line) carried out at Miyakejima volcano, Japan on (a) 22 July 2003 and (b) 8 June 2004. The COSPEC's SO_2 column amount data circled in blue line correspond to output of 1438 ppmm SO_2 calibration cell. The COMPUSS profile is piled on the COSPEC profile with the same time and SO_2 column amounts level so that the calibration cell values are equal to the scale of the COMPUSS data. The COSPEC zero level and gain (the output of the standard cell) was drifted with time on 8 June 2004. The COMPUSS data are scaled with the first three data of the standard cell.

Table 2.　Comparison of SO$_2$ fluxes obtained using the COMPUSS and COSPEC at Miyakejima volcano.

Date (y, m, a)	Plume Direc. (deg.)	Speed (m/s)	No. of Trav.	SO$_2$ flux (ton/day) COMPUSS	SO$_2$ flux (ton/day) COSPEC	COSPEC / COMPUSS
03.07.22	45	6.0	3	8600	8700	1.0
03.12.16	300	7.5	3	8100	6100	0.8
04.01.22	270	3.5	2	3500	3300	0.9
04.02.17	270	22.5	4	4500	4800	1.1
04.06.08	262	11.0	7	4100	5700	1.4
04.07.24	60	8.0	2	4600	11600	2.5
04.08.17	230	16.9	3	5400	5500	1.0

obtained between the COSPEC and C3 COMPUSS data. This good agreement between the two instruments is quite significant considering the difference of the FOVs between the COSPEC (7×21 mrad) and the COMPUSS. The disagreement in proportional coefficient from unity observed on 20 October is up to 6.5% and this disagreement is mainly due to rapid time fluctuation of the plume and/or to minor misalignment of the telescope from zenith. Disagreement of the proportional factor has improved to less than 2% for all the pairs on 21 October. The results of three COMPUSSes show high reliability and reproducibility of the COMPUSS measurements. The one-to-one correspondence between the COMPUSS and the COSPEC outputs show that the COMPUSS can secure the data continuity with accumulated COSPEC data and can replace the COSPEC without any problem.

4–3　Heliborne traverse measurements at Miyakejima volcano

Miyakejima volcano is a basaltic island on the Izu-Mariana arc and is located about 180 km south of Tokyo. The volcano has been intensively discharging volcanic gas since August 2000 after the summit caldera formation. Sulfur dioxide flux from the volcano has been monitored repeatedly by heliborne COSPEC measurements until 2005. Extremely high monthly averaged SO$_2$ flux of 57000 ton/day has been recorded in Dec. 2000 and the monthly averaged flux still has 7000 ton/day at least until December 2003 (Kazahaya et al., 2004). Since July 2003, we carried out parallel heliborne SO$_2$ flux measurements using the COMPUSS and the COSPEC measurements for seven times. In the heliborne measurements, the COMPUSS's telescope was directly attached to COSPEC's telescope and the UV spectrometer was fixed on COSPEC's body. The profiles obtained by the two instruments (Fig. 6) show simultaneous and similar temporal variation patterns of SO$_2$ column amounts and accordingly similar SO$_2$ flux values. The average SO$_2$ fluxes obtained by the COMPUSS and COSPEC are commonly in good agreement within 10% disagreement (Table 2). However, there

are also a few cases where the large discrepancy of > 20% is found. Since same plume direction and speed are used in the flux analyses for both instruments, the disagreement should be purely instrumental.

COSPEC's SO$_2$ charts (Figs. 6a and 6b) are very noisy and show a several variety of noise patterns. The noise in the COSPEC chart is due to interference between rotating correlation disc in the COSPEC and rotating rotor blades which are in the view of the instruments (Daag et al., 1996; Galle et al., 2002). In contrast, the COMPUSS's output (Fig. 6a and 6b) is not affected by rotating rotor blades, since the covering duration of the view by the rotor blades are significantly shorter than the integration time of the UV spectrometer. The absence of the rotor blade noise for the UV spectrometer system is also mentioned in Galle et al. (2002). For flights on 22 Jul. 2003 (Fig. 6a), 22 Jan. 2004, 17 Feb. 2004 and 17 Aug. 2004, the noise has relatively small and regular oscillation noise in SO$_2$ column amounts. In these flights, the results of the COSPEC and the COMPUSS have good agreement. For flights on 16 Dec. 2003 and 24 Jul. 2004, beating-like noise level is sometimes larger than the SO$_2$ signals of the plume. In flights on 8 Jun. 2004, the COSPEC chart shows somehow random and peaky noise patterns (Fig. 6b). In these three flights, the noise is definitely affecting the flux estimations for the COSPEC data, and the large discrepancies between the two fluxes are more than 20%. The absence of the rotor blade noise is the clear advantages of the new system over the COSPEC. This advantage of the COMPUSS will be more important and critical, when carrying out heliborne SO$_2$ flux measurements for plumes with much lower SO$_2$ column amounts than those of Miyakejima.

5.　Concluding remarks

Newly developed COMPUSS, using a commercial compact UV spectrometer, is lighter and more portable compared with the COSPEC. From the relation between peak heights and SO$_2$ column amounts, the COMPUSS can readily measure at least up to 2700

ppmm of SO_2. The field experiment demonstrats the ability of the COMPUSS for reliable calibration of the system for wide column amount range. Simultaneous measurements of the COMPUSS and the COSPEC show good agreement. The new system can keep the continuity of the past COSPEC data obtained by traverse method. The COSPEC measurements are usually influenced and give significant noises with tree branches or rotor blades in the view of the telescope. These noises are absent in the case of COMPUSS. These evidences expand the opportunity of conducting the flux measurements in various conditions. The COMPUSS and similar systems will probably replace the COSPEC in near future in the volcanological community and contribute to further understanding of the volcanic degassing system.

Acknowledgements

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