京都大学防災研究所年報 第47号 C 平成16年3月
Annuals of Disas. Prev. Res. Inst., Kyoto Univ., No.47 C, 2004

SO_2 gas monitoring by DOAS at Sakurajima and Suwanosejima volcanoes

Takehiko MORI*, Kazuhiro ISHIHARA, Junichi HIRABAYASHI**,
Kouhei KAZAHAYA***and Toshiya MORI****

*Aso Volcanological Laboratory, Graduate School of Science, Kyoto University
** Volcanic Fluid Research Center, Tokyo Institute of Technology
*** National Institute of Advanced Industrial Science and Technology
**** Laboratory for Earthquake Chemistry, Graduate School of Science, University of Tokyo

Synopsis

We have produced trial instrument for measurement of SO2 emission using differential optical absorption spectrometer (DOAS). Its performance has been tested at several volcanoes in Japan since 2003. The performance of the DOAS and its practice measurement were executed in Sakurajima and Suwanosejima. The SO2 emissions from Sakurajima and Suwanosejima were measured by the DOAS. Sakurajima emitted 500~1000ton/day due to dormancy of the volcanic activity. The excellent portability of the DOAS enables to estimate the SO2 emission from Suwanosejima at 1000ton/day. In addition, the DOAS enables to monitor temporal variation in the SO2 column concentration in the vicinity of the crater. We carried out measurement of the SO2 column concentration (ppmm) in the plume at Suwanosejima.

Keywords: Volcanic gas; COSPEC and DOAS; SO2 emission; SO2 column concentration

1. Introduction

Volatile components are the most mobile materials in magma and change in chemical composition of fumarolic gases is often observed as one of the remarkable phenomena associated with activation of volcanism, which has been generally acknowledged (e.g. Saint-Claire Deville and Leblance, 1858). Increase in SO2/H2S molar ratio in fumarolic gases at Mt. Kusatsu-Shirane located in the central Honshu, Japan had been detected one year before the 1976 eruption, which resulted in success of prediction of the eruption (Ossaka et al, 1980). Monitoring of chemical composition of volcanic gases can contribute toward the prediction of volcanic eruptions.

Flux of volcanic gases will also provide information on volcanism. Increase in carbon dioxide gas flux was detected on the summit caldera six months before the 2000 eruption of Usu volcano (Hernandez et al, 2001). COSPEC (COrrelation SPECtrometer) is a spectrometric instrument, which was contrived for SO2 measurement, has been widely utilized in the observation of gas flux at several volcanoes since 1970's (e.g. Stoiber et al., 1983). Increase in SO2 gas emission prior to the eruption of Mt. Asama from 1982 to 1983 is noticeable and emission of SO2 progressively declined when the volcanic activity became dormant (Ohta et al, 1984).

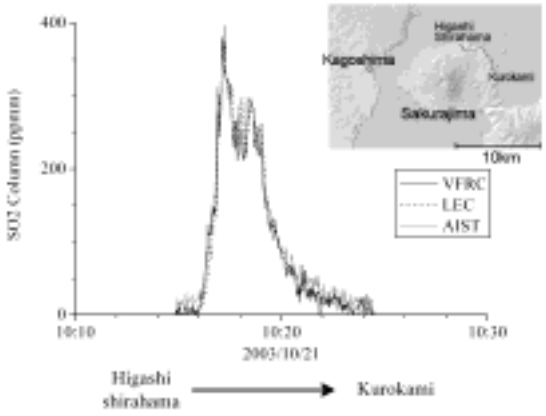

Fig.1 SO2 column concentration that was recorded with the DOASs of three organizations between Higashi-Shirahama and Kurokami on October 21, 2003.

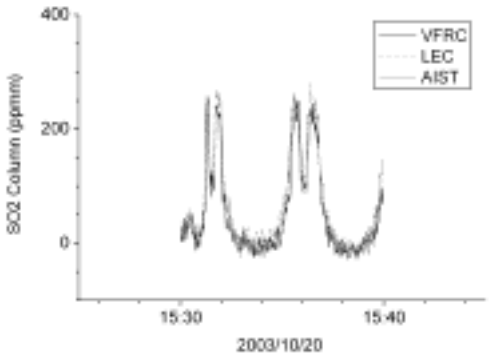

Fig.2 SO2 column concentration that was recorded with the DOASs of three organizations at Hakamagoshi on October 20, 2003.

Table 1 Results of SO2 emission

Organization	SO_2 Flux
VFRC	538 ton/day
LEC	540 ton/day
AIST	584 ton/day

Hirabayashi et al. (1995) designated that the discharge rate of SO2 correlated with the extrusion rate of magma during the prolonged volcanic activity at Unzen-Fugendake. Therefore, monitoring of gas emission can also be a good indicator of volcanic activity.

Though measurement of SO2 emission cannot be performed by the COSPEC at all volcanoes because of less portability of it, a miniaturized, ultraviolet, differential optical absorption spectrometer for remote sensing of SO2 emission has been developed since 2000 (Galle et al., 2003). We also have produced trial instrument for measurement of SO2 emission using differential optical absorption spectrometer and hereafter, the instrument is referred as the "DOAS". Its performance has been tested at several volcanoes in Japan since 2003. In this paper, the performance of the DOAS and its practice measurement at Sakurajima and Suwanosejima are reported.

2. Quality Test for the DOAS

Hirabayashi et al. (2003) carried out side-by-side measurements of SO2 emission using both the DOAS and the COSPEC at Mt. Asama and Miyakejima. They reported that difference in the SO2 emission between these devices was negligible. This result designates that the emission measured by the DOAS is equivalent with those by the COSPEC.

All the DOAS is based on an Ocean Optics USB2000 spectrograph. Though the characteristic value of each USB2000 is specified, it is different from each other and the performance of the DOAS is subject to the characteristics of the spectrometer. Hence, three organizations (Volcanic Fluid Research Center, Tokyo Institute of Technology (VFRC), National Institute of Advanced Industrial Science and Technology (AIST) and Laboratory for Earthquake Chemistry, Graduate School of Science, The University of Tokyo (LEC)) simultaneously measured the SO2 emission from Sakurajima in October, 2003 in order to crosscheck their DOASs performance.

The DOASs were mounted in a vehicle with lens oriented vertically upwards and the vehicle made traverses beneath the gas plume between Higashi-Shirahama and Kurokami on October 21, 2003. The vehicle position was tracked using a handheld GSP receiver and profile of SO2 column concentration in the air was recorded (**Fig.1**).

Table 2 The SO2 emission at Sakurajima Volcano.

Date	Observation Time	Observation Method	Observation Route	SO2 Emission (Average)	SO2 Emission (Minimum)	SO2 Emission (Maximum)
2003/10/18	12:00~13:30	Panning	Arimura	666 ton/day	378 ton/day	1034 ton/day
2003/10/18	12:15~14:15	Traverse	Kurokami-Furusato	761 ton/day	584 ton/day	1010 ton/day
2003/10/20	12:00~12:30	Traverse	HigashiShirahama-Kurokami	481 ton/day	----------	----------
2003/10/21	10:14~13:48	Traverse	HigashiShirahama-Kurokami	827 ton/day	540 ton/day	1302 ton/day

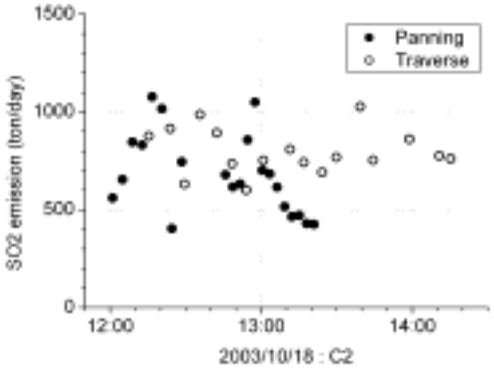

Fig.3 Temporal fluctuation in the SO2 emission. Two measurement methods that is Traverse and Panning were compared.

Sakurajima emitted SO2 gas between 538 and 584 ton/day on that day (**Table 1**). Difference in the SO2 flux between AIST and VFRC was about 40 ton/day, which was not significant taking the error into consideration. Further, the 3 DOASs positioned on a tilting mount and they simultaneously scanned the plume trailing southwestward from stationary position at Hakamagoshi on October 20, 2003. As shown in **Fig.2**, those DOASs recorded a very similar profile of column concentration. These results strongly designates that the performance of the 3 DOASs equivalent to each other.

3. Measurement of the SO2 emission at several volcanoes

3-1. SO2 emission from Sakurajima

Traverses from Arimura area, southeastern Sakurajima on October 18, 20 and 21 were carried out, which yields SO2 emission between 500 and 1000 ton/day (**Table 2**). It was less than half of the emission between 1978 and 1995 ranging 1400 ~ 2800 ton/day by Hirabayashi et al. (1997). Explosions occurred only three times in this month and observation was carried out during a dormant period, which is reasonable that the emission was not so large quantity.

Hirabayashi et al. (1997) pointed out that the emission that the COSPEC measured by traverse was several times as much as that by panning. They estimated the cause of the difference such as hanging of volcanic plume in the air near the volcanic edifice, interference of volcanic edifice with incoming sunlight near skyline, disperse of volcanic plume and so on.

Temporal fluctuation in the SO2 emission is shown in **Fig.3** and difference between the both methods is not significant. This result insists that difference in the emission do not depend on the measurement methods.

3-2. Emission of SO2 from Suwanosejima island

Suwanosejima island, one of the Tokara Islands, located at about 200 km southward from Kagoshima city is one of the most active insular volcanoes in Japan. In December, 2000, eruption resumed, resulting in formation of the new crater inside of the summit crater. The volcanism is

Fig.4 Map showing Suwanosejima. Observation points are A and B. Point A's name is 'Old Crater' and B's name is 'Kiriishi quay'. The measurement line of Tracerse method is C and D. The line C is measured by using a fishing boat. The line D is measured with walking Traverse.

Table 3 The SO2 emission at Suwanosejima Volcano.

Date	Observation Time	Observation Method	Observation Route	SO2 Emission (Average)	SO2 Emission (Minimum)	SO2 Emission (Maximum)
2003/11/4	13:44~14:22	Panning	Old Crater	460 ton/day	104 ton/day	1578 ton/day
2003/11/6	12:10~13:50	Panning	Kiriishi quay	3860 ton/day	2221 ton/day	6608 ton/day
2003/11/8	11:46~12:02	Panning	Old Crater	530 ton/day	396 ton/day	760 ton/day
2003/11/12	11:50~13:45	Panning	Old Crater	1596 ton/day	401 ton/day	2469 ton/day
2003/11/13	14:19~14:41	Traverse	Kiriishi-Sakuchi	890 ton/day	793 ton/day	990 ton/day
2003/11/14	11:00~12:00	Traverse	Crater vicinity	950 ton/day	844 ton/day	1053 ton/day

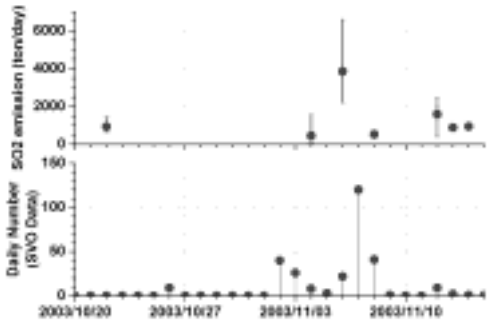

Fig.5 Relationship between the number of explosion and the SO2 emission.

currently active and ash eruption occurs every two and/or three weeks.

There is no road around the volcano and traverse by a vehicle is not available for monitoring of the SO2 emission. Due to less portability of the COSPEC, the DOAS, which is lightweight and suitable for field deployment, was employed for the measurement of the SO2 emission from Suwanosejima from November, 3 to November 15, 2003. The observation point at Suwanosejima Island is plotted in **Fig.4**. The result of the measurement of the SO2 emission is shown in **Table 3**.

Volcanic plume trailing eastward was scanned from the stationary position at Kiriishi quay on November 6. The volcanic gases dispersed and hanged in the air offshore and the wind speed of the scanned column near the surface of the sea was obviously lower than that of the plume near the summit. Then, application of wind speed of the plume estimated by a series of the photographs of the plume to speed for the flux calculation caused the prominence of the SO2 emission on that day. Relationship between the number of explosion and the SO2 emission shown in **Fig.5** clearly indicates the overestimation of the SO2 emission on that day.

In general, ash particles in plume block out

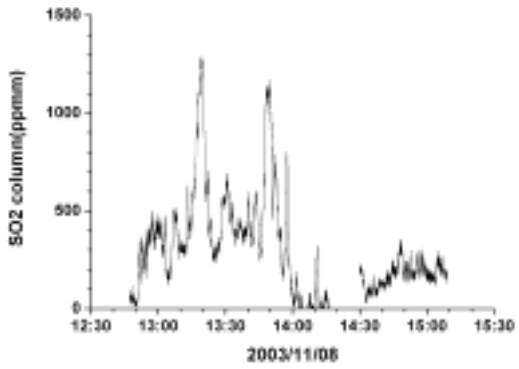

Fig.6 The temporal change in the SO2 column concentration in the plume from the old crater

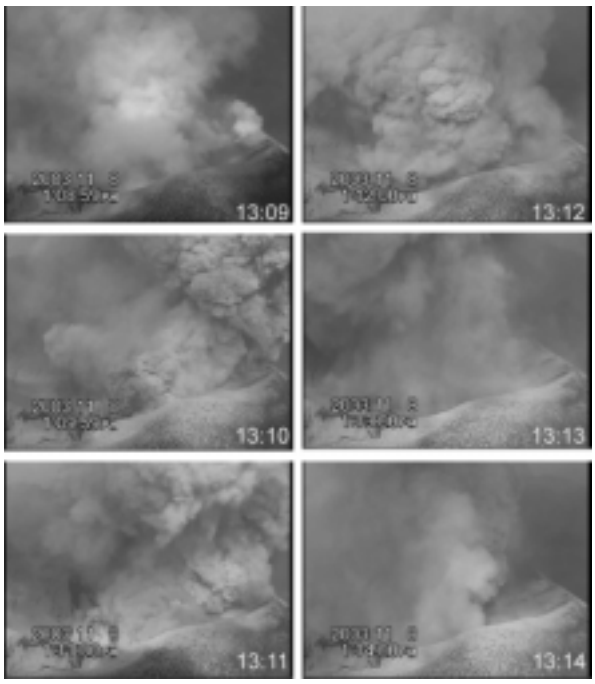

Photo 1 Snap shots from a video picture. A small eruption occurred at about 13:09. The photograph at 13:10 is dark because a volcanic ash is included. However, the photograph of 13:13 is white because a/the volcanic gas became nucleus.

incoming sunlight, which makes the SO2 column concentration downward. Suwanosejima volcano often raise dense ash plume during an active period. Though the average of the SO2 emission discharged from the volcano is estimated at about 1000ton/day, the SO2 emission is actually much more than that average.

4. Detection of temporal variation in SO2 column concentration in plume

Excellent portability of the DOAS enables us to approach close to active crater and the instrument is available to measurement in the vicinity of craters. We carried out measurement of the SO2 column concentration (ppmm) in the plume at Suwanosejima on November 8, 2003.

The temporal change in the SO2 column concentration in the plume from the old crater is shown in **Fig.6**. It is characteristic that the column concentration increased between 13:15 and 13:45. We focus on the change of it at about 13:15, because occurrence of a small-scale eruption was clearly recorded at 13:09 (**Photo 1**). **Fig.7(a)** show a temporal variation from 13:00 to 13:30 in the SO2 column concentration and the temperature measured by an infrared camera.

At 13:09, high temperature volcanic ejecta were discharged from the crater by a small-scale eruption, however, the SO2 column concentration did not change. Increase of the column

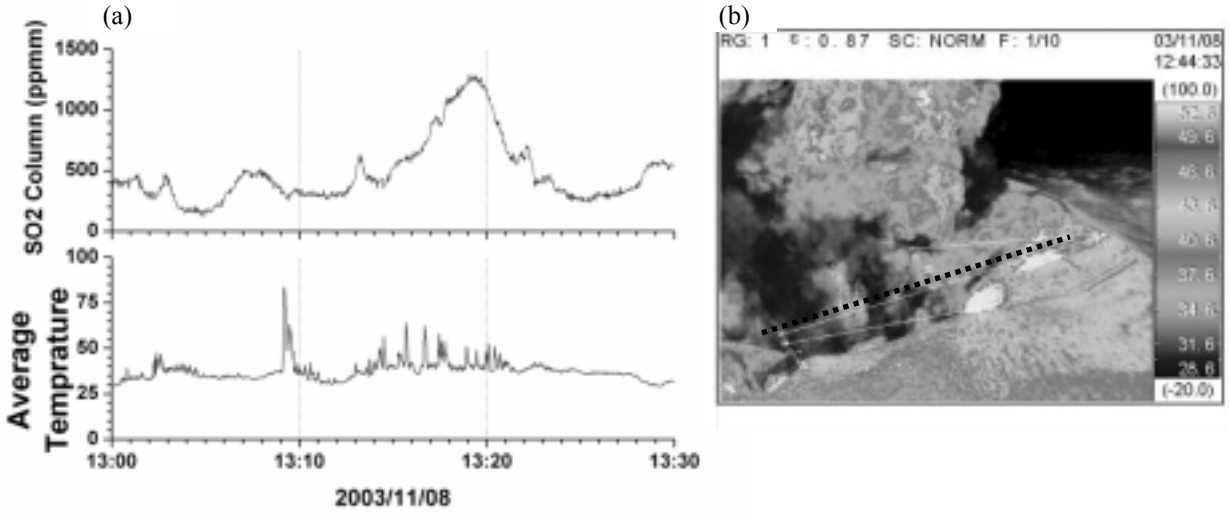

Fig.7 (a): A temporal variation from 13:00 to 13:30 in the SO2 column concentration (above) and the temperature measured by an infrared camera (below). (b): A snapshot from the picture of an infrared camera. The average temperature (shown in (a)) is the temperature of a dotted line.

concentration occurred at 13:13 when the temperature rose again and the ash plume changed into the white plume at that time. This result indicates that discharge of high temperature volcanic gas at 13:13 increased the SO2 column concentration in the plume.

The lightweight DOAS newly enables us to measure the SO2 column concentration in the vicinity of the crater. The column concentration is a new item on monitoring of volcanism and measurement of the SO2 emission by the DOAS contributes to disclose of eruption mechanism at Suwanosejima.

5. Conclusion

1) We have produced trial instrument for measurement of the SO2 emission using differential optical absorption spectrometer, DOAS. It is lightweight and suitable for field deployment, which will take the place of the COSPEC in the near future.
2) The SO2 emission from Sakurajima and Suwanosejima was measured by the DOAS. Sakurajima emitted 500~1000ton/day due to dormancy of the volcanic activity. The excellent portability of the DOAS enables to estimate the SO2 emission rate from Suwanosejima at 1000ton/day.
3) The DOAS enables to monitor temporal variation in the SO2 column concentration in the vicinity of the crater. Measurement of the SO2 emission by the DOAS contributes to disclose of eruption mechanism at active volcanoes.

Acknowledgement

My special thanks are due to Dr. Nogami of Volcanic Fluid Research Center, Tokyo Institute of Technology for reading the manuscript and making a number of helpful suggestions.

I would like to thank Dr. Iguchi, Dr. Tamekuri and all staffs at Sakurajima Volcano Research Center D.P.R.I. for all of this research. I would like also to thank Dr. Yakiwara of Nansei-Touko observatory for Earthquakes and Volcanoes, Faculty of Science, Kagoshima University and thanks to Mr. Oikawa, Mr. Mizuhashi and Mr. Saito of Volcanic Fluid Research Center, Tokyo Institute of Technology and thanks to Mr. Ito and all the islanders of Suwanosejima for supporting my observation.

This research was supported by the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT) 21st Century COE Program for DPRI, Kyoto University (No.14219301, Program Leader: Prof. Y. Kawata) as well as the Grant-in-Aid for Scientific Research (B) (1) (No. 15999999, Principal Investigator: S. Ikebuchi, Kyoto University). In addition, this research was supported in part by grant from the Grant-in-Aid for Scientific Research (B) ('Dynamics of Volcanic Explosion', No.14080205,Project Leader: Y. Ida).

References

Galle, B., Oppenheimer, C., Geyer, A., McGonigle, A. J. S., Edmonds, M. and Horrocks, L. (2002): *J. Volcanol. Geotherm. Res.*, 119, 241-254.

Hirabayashi, J., Ohba, T., Nogami, K. and Kagiyama, T. (1995): Volcanic activity and volcanic gas from 1990 to 1994 at

Unzen-Fugendake, *The report of Grant in Aid for Scientific Research (a) No.06306011*, 22-25 (in Japanese).

Hirabayashi ,J., Ohba, T., Nogami, K., Ohta, K., Shimizu, H., Matsuo, N., Kazahaya, K. and Shinohara, H. (1997): The SO2 emission from Sakurajima Volcano, *The report of Joint Observation of Sakurajima Volcano*, 113-118 (in Japanese).

Hirabayashi, J., Mizuhashi, S., Kazahaya, K., Shinohara, H., Mori, T., Olmos, R. and Mori, T. (2003): Development of a portable SO2 monitor, *Bull. Volcanol. Soc. Japan* (Abstracts), No.2, 14-14 (in Japanese).

Hernandez, P. A., Notsu, K., Salazar, J. M., Mori, T., Natale, G, Okada, H., Virgili, G., Shimoike, Y., Sato, M., and Perez, N. M. (2001): Carbon dioxide degassing by advective flow Usu volcano, Japan. *Science*, 292, 83-86.

Ohta, K., Hirokawa, M., Hirahara, S. and Kagiyama, T. (1984): The change of SO2 gas emission and volcanic activity of Asama volcano. *The report of an unforeseen disaster research, the special research of natural disaster, ministry of Education national science foundation*, No.A59-4, 112-114 (in Japanese).

Ossaka, J., Ozawa, T., Nomura, T., Ossaka, T., Hirabayashi, J., Takaesu, A. and Hayashi, T. (1980): Variation of chemical compositions in volcanic gases and waters at Kusatsu-Shirane Volcano and its activity in 1976. *Bull. Volcanol.*, 43, 207-216.

Saint-Clarie Deville, C. and Leblance, F. (1858): La composition cheniqre des gaz. *Ann. Chem. Phys.*, 3rd Ser., 52, 5―63.

Stoiber, R.E. and Jepsen, A. (1973): Sulfur dioxide contributions to the atmosphere by volcanoes. *Science*, Vol. 182, 577-578.

要 旨

我々は火山からの SO2 放出量測定のために，光学的な吸収分光計を用いた試験的な測定機器（DOAS）を新たに開発した。この機器の基本的な動作試験は浅間山や三宅島などの日本の火山で 2003 年に行われてきた。そして，DOAS の特性試験と実際の計測が桜島と諏訪之瀬島（トカラ列島の火山島）で 2003 年の 10 月から 11 月にかけて行われた。桜島では 500~1000ton/day の SO2 放出量が記録された。活動が最盛期であった桜島からの SO2 放出量の半分以下の計測値であるが，現在の桜島の火山活動の低さから考えると妥当である。また，噴火活動が活発である諏訪之瀬島では初の SO2 放出量の測定が行われ，約 1000ton/day の SO2 放出量が計測された。桜島最盛期と比べると SO2 放出量は低いが，諏訪之瀬島の噴火規模から考えると妥当な放出量と考えられる。一方で，火口近傍に DOAS を運び，SO2 column 濃度の時間変動の測定にも成功した。

キーワード：火山ガス，DOAS と COSPEC，SO2 放出量，SO2 column 濃度

DOAS によって観測された火山ガス放出量の変動について
～桜島・諏訪之瀬島～

○森 健彦（京大理）・石原和弘（京大防災研）・平林順一（東工大火山流体）・
風早康平（産総研）・森 俊哉（東大地殻化学）

はじめに

火山からの SO2 放出量の測定には 1970 年代より紫外線相関スペクトロメター（COSPEC）が使用されてきた。2000 年代に入り，小型の紫外線分光計を用いた SO2 放出量測定器がいくつかのグループにより開発され, 火山での測定が報告されている。我々も同様な小型 SO2 放出量測定装置を試作し, 2003 年度から国内の火山に於いて試験測定を行ってきた。噴煙中の SO2 カラム量の測定原理としては DOAS 法（Differential Optical Absorption Spectroscopy）を採用しており，我々はこの小型 SO2 放出量測定器を ”DOAS ”と呼称している。

DOAS を用いた測定方法も COSPEC 同様にパニング法とトラバース法がある。特にトラバース測定の際には, 測定用の PC に GPS を取り付け, 位置情報も同時に記録できるようになった。さらに, DOAS のサイズが小さく軽量となったため，山頂火口まで容易に運搬でき，火口周縁を歩いてのトラバース測定も可能となった。

本報告では，昨年 10 月に桜島で行った DOAS の機器特性の試験結果を報告する。さらに，DOAS の軽量可搬型の特性を生かして観測した諏訪之瀬島における SO2 の放出量及び火山ガス放出量の時間変化の観測結果を示す。

機器特性の試験

2003 年 10 月に日本国内で DOAS を所持する 3 機関（東工大火山流体・産総研・東大地殻化学）が桜島に集結し，それぞれが所持する DOAS の特性差を検討した。さらに，COSPEC（北大有珠所有）との比較観測を行って, 従来の計測結果との連続性を確かめる試験を行った。3 台の DOAS を用いて，火山ガス濃度を計測した結果，DOAS の機器による特性の差はないことが明らかになった。また，COSPEC で記録された火山ガス濃度も同様な変化を記録していることから，これまでの COSPEC による観測結果と DOAS による観測結果は継続性が保てることが明らかになった。

火山ガス放出量の時間変化

諏訪之瀬島は, 2000 年 12 月にこれまで噴火が発生していた山頂火口内の中央火砕丘の外側に新しい火孔を形成して噴火活動を再開し，現在まで 2～3 週間の間隔で噴火活動を繰り返している。諏訪之瀬島は火口へのアプローチが厳しく，総重量の重い COSPEC での観測は不可能であった。そこで, DOAS を使用することによって，諏訪之瀬島火口から放出される SO2 の放出量を計測した。その結果，日量で約 1000ton の SO2 が放出されていることが明らかになった。

さらに, 火口近傍に持ち込んだ DOAS によって, 噴煙中に含まれる SO2 濃度の時間変化を観測した。その結果，小爆発直後には濃度変化が見られなかったが，火山ガス主体の噴煙へと変化した直後から SO2 column 濃度が上昇する変化が計測された。