Chapter 6

Beyond COSPEC: Recent advances in SO_2 monitoring technology

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1. INTRODUCTION

In the years since its 1971 introduction to the volcanological community, the correlation spectrometer (COSPEC) long maintained its position as the primary tool for the remote sensing of volcanic SO_2 emissions (Stoiber et al., 1983). The technology was first developed by Barringer Research as a means for monitoring industrial output of SO_2 and NO_X to the environment. Volcanologists quickly realized the applicability of such an instrument to volcanic gas monitoring, and began field campaigns around the world (e.g., Stoiber and Jepsen, 1973; Stoiber et al., 1980; Stoiber et al., 1986; Bluth et al., 1994; Casadevall et al., 1994; Elias et al., 1998; Delgado et al., 2001; Rodrîguez et al., 2004).

However innovative the initial application of COSPEC was to volcano monitoring, little was done to improve on the instrumentation itself. It was heavy and cumbersome (~20 kg, 102 cm x 53 cm x 28 cm), and not intended for field work in rough and variable volcanic terrains. Aside from the instrument itself, a power source was necessary, as was a paper chart data recorder and some sort of platform for the instrument. Price was also prohibitive, with a single instrument costing tens of thousands of US dollars (Galle et al., 2002). In fact, the only significant advance of COSPEC technology over the years was the optional integration of a digital data interface, making replacement of the paper chart recorder possible. The size and weight remained essentially the same, while costs climbed as a result of limited demand, and for years, COSPEC remained the sole spectrometer utilized to remotely measure volcanic SO_2 flux.

Beginning in the late 1990s, miniature, low-cost spectrometers, as well as increasingly powerful and compact computer technology led to the advent of alternatives to COSPEC. Based largely on differential optical absorption spectroscopy (DOAS; Platt, 1994) and Ocean Optics USB spectrometers, new systems, such as the mini-DOAS (Galle et al., 2002), RMDI (Wardell et al., 2003), MUSE (Rodríguez et al., 2004), FLYSPEC (Horton et al., 2006) and others (e.g., Mori et al., 2004) offered a smaller, cheaper, more robust alternative to the COSPEC, while also improving several factors in the data collection methodology and offering the opportunity for a wider range of field applications.

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2. INSTRUMENTATION

2.1. ‘Mini-DOAS’-type systems

First described by Galle et al. (2002), ‘mini-DOAS’-type systems utilize an Ocean Optics USB spectrometer, and often comprise a telescope, circular-to-linear optical fiber, a ~50-µm slit, collimating mirror, plane grating, curved mirror, and a linear charge-coupled-device (CCD) array (Fig. 1). The spectral resolution of such instruments is ~0.6 nm in the 245-380 nm wavelength range. Spectra for the ‘mini-DOAS’-type systems are output from the instrument to a computer, which also serves as the power source, via a USB connection. There is also generally a GPS antenna included in the system, which allows for position- and time-stamping of data as they are collected.

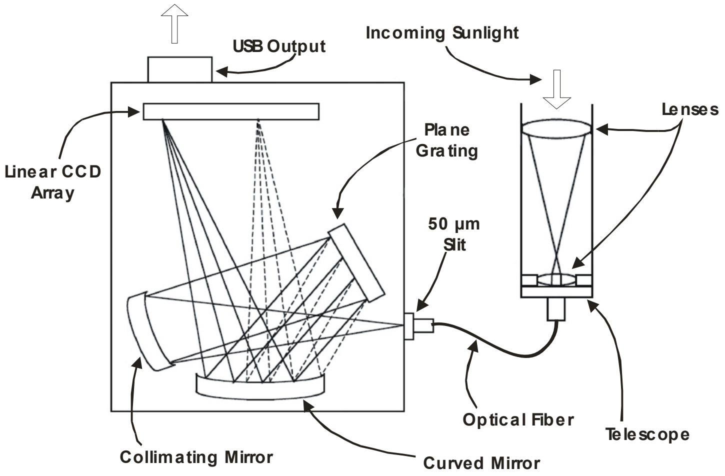

Fig. 1. Sample configuration of an Ocean Optics USB ‘mini-DOAS’-type spectrometer system coupled to a telescope via an optical fiber (figure not to scale). Dashed and solid lines drawn within the spectrometer represent the paths of light at two distinct wavelengths, displaying how the spectrometer disperses radiation on to the CCD array. Modified from Galle et al., 2003.

The entire package, for many of the systems, is considerably more portable than the COSPEC, commonly weighing less than 1 kilogram (without laptop computer). Likewise, the dimensions of the ‘mini-DOAS’-type systems (~90 mm x ~65 mm x ~35 mm) are considerably smaller than the COSPEC dimensions. Compared to COSPEC, less power is required to deploy a ‘mini-DOAS’-type system: 1W via a laptop computer connection versus up to 23W for a COSPEC (Galle et al., 2002). Costs are also significantly lower for a ‘mini-DOAS’-type system, with the entire configuration, including a laptop computer, often attainable for approximately 10% the cost of a COSPEC (McGonigle et al., 2003). Beyond physical specifications, the ‘mini-DOAS’-type systems’ spectroscopic techniques differ from that of the COSPEC. Instead of utilizing a correlation mask, whereby levels of radiation at wavelengths specific to the atmospheric absorption windows of SO_2 are calibrated by means of comparison to internal cells of known SO_2 concentration (see Chapter 1), a ‘mini-DOAS’-type system records full radiation spectra covering a wide range of wavelengths.

Solar radiation spectra are captured at user-defined intervals, which vary depending on optical hardware and ambient atmospheric conditions. Dark spectra are routinely measured during data collection by prohibiting any radiation from entering the telescope. Subtraction of dark spectra from sky spectra is required to account for instrument noise within the data set. Background spectra of SO_2-free sky are also collected for utilization in the data retrievals and isolation of the volcanic SO_2 signals. Spectra are then processed by means of a number of filtering techniques. Final spectra are compared and scaled to a laboratory reference spectrum for SO_2 in order to determine the column amount of SO_2 present at the time of the measurements.

Field trials utilizing the COSPEC and ‘mini-DOAS’-type system in tandem have confirmed comparable results for the two instruments (Galle et al., 2002; Elias et al., 2006).

2.2. FLYSPEC

The FLYSPEC, described in Horton et al. (2006) and Elias et al. (2006), is similar to the mini-DOAS in the respect that the system utilizes an Ocean Optics USB spectrometer. For the FLYSPEC, a 25–μm slit results in a spectral resolution of 0.25 nm over a range of wavelengths from 177-330 nm. Both spectrometer systems utilize a laptop computer for data processing and as a power source, and are significantly smaller, lighter, and cheaper than the COSPEC. The FLYSPEC itself weighs less than two kilograms, including a small, sub-notebook computer.

Aside from differing spectral specifications, there are a number of physical variations between the two instruments. The fiber optic cable is omitted from the FLYSPEC; the telescope is attached directly to the spectrometer in order to reduce light loss. The FLYSPECs often have a larger field of view than the ‘mini-DOAS’-type stystems, at >40 mrad rather than ~20 mrad. Additionally, the FLYSPEC has a UV filter to reduce stray light, and includes SO_2 calibration cells within its configuration. The FLYSPEC components, like many ‘mini-DOAS’-type systems, are also housed within robust, weather-proof cases, well-suited for field deployment (Fig. 2).

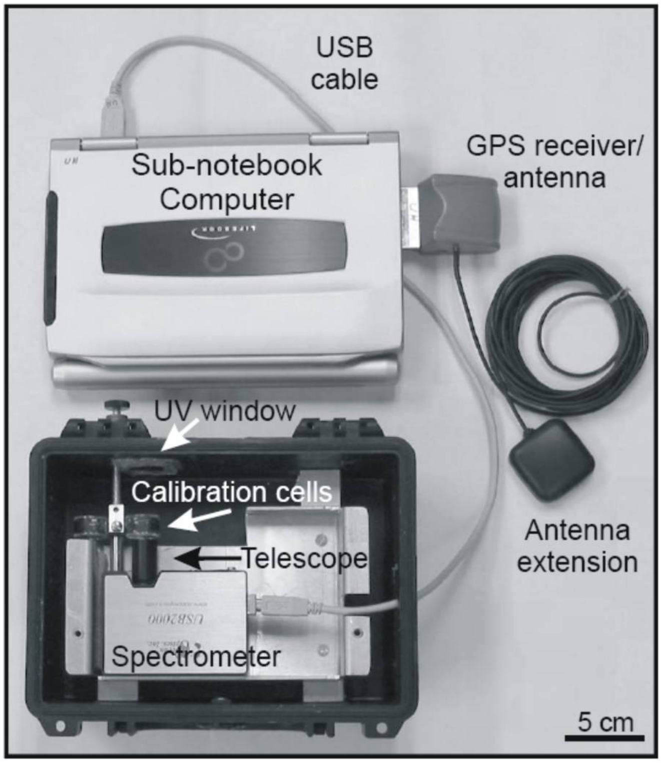

Fig. 2. Components of a FLYSPEC: an Ocean Optics USB spectrometer, sub-notebook computer, GPS with antenna, high and low calibration SO_2 gas cells mounted above the spectrometer, and telescope. The “telescope” is a fiber-optic collimating lens mounted directly to the spectrometer input aperture. The lens, in combination with the UV band-pass filter window mounted on the case, provides a field of view of approximately 2.5°. Power for the spectrometer and GPS is supplied by the computer. Modified from Horton et al., 2006.

Rather than the ‘mini-DOAS’ method of collecting spectra and comparing to a laboratory reference spectrum, the FLYSPEC relies on calibration cells, similar to COSPEC. Dark spectra and background spectra are still utilized, and a procedure similar to the processing of mini-DOAS data takes place. Instead of comparing final spectra to a laboratory reference spectrum, however, the FLYSPEC spectra are calibrated to the reference spectra produced by the insertion of calibration cells of known concentration into the instrument’s field of view. Calibrations are performed in the field, in the same atmospheric conditions as the collection of the sample dataset. By using calibration-cell reference spectra obtained in the field, atmospheric effects such as Fraunhofer lines or ring-effect (which would not be present in model laboratory spectra) will be present in all spectra, eliminating the need for data corrections (Horton et al., 2006). Like the 'mini-DOAS'-type system, the FLYSPEC also records and stores full atmospheric spectra, allowing for re-processing and comparison to library or modeled spectra following the DOAS methodology. FLYSPEC results in field trials are comparable to those of COSPEC and mini-DOAS (Elias et al., 2006; Horton et al., 2006).

Despite differences between the new ground-based tools in the field of remote SO_2 monitoring at volcanoes, each of the spectrometer system types provides a viable alternative to the COSPEC. The compact size, relatively low cost, DOAS techniques, and computer/GPS integration of the new generation of miniature spectrometer systems have allowed for an increased range of possible applications for monitoring volcanic SO_2.

3. FIELD METHODOLOGY

3.1. *Vehicle Traverses*

As with traditional COSPEC methodology (Stoiber et al., 1983; Chapter 2), the new generation of spectrometer systems is well-suited for vehicle-based traverses beneath plumes. Systems utilizing an optical fiber to link the spectrometer and a telescope have increased ease of use in vehicular traverses, as only the telescope requires external mounting, allowing the spectrometer itself to be manipulated as needed within the car, plane, helicopter, or boat. Systems lacking the optical fiber and separate telescope are still very convenient in traverses, as entire instruments are generally small enough to be easily mounted on the exterior of the vehicle (Fig. 3).

Fig. 3. Example of a FLYSPEC field deployment. The spectrometer unit is mounted near the car's side mirror with duct tape, while the GPS antenna is affixed to the roof.

3.2. *Walking Traverses*

At many volcanoes worldwide, automobile traverses using any UV-spectrometer are impossible, as a consequence of remote locations, insufficient infrastructure, or a combination of both. COSPEC has been employed sporadically in walking traverses in situations where plume geometries or limited access prohibited scans or vehicular traverses (Stoiber et al., 1983); however, the weight and size of the instrument, as well as the

Fig. 4. A walking traverse with a backpack-mounted FLYSPEC at Vulcano, Italy, 2002.

requisite constant traverse speed prior to GPS integration kept the walking traverse methodology from becoming commonplace. The newer, smaller, GPS-integrated systems facilitate such walking traverses, as demonstrated at both Kilauea (Horton et al., 2006) and Masaya volcanoes (McGonigle et al., 2002). The small spectrometers are easily mounted in a carrier on the operator's back (Fig. 4), or, in the case of systems with a telescope/optical fiber combination, the telescope may be affixed to a helmet and the body of the spectrometer simply carried by the operator (McGonigle et al., 2002).

3.3. Automated Scanning

While the new spectrometer systems lend themselves to manual scanning of plumes as with COSPEC methodology, their small size and high degree of computer integration make automated scans of plumes reasonably straightforward. The addition of a rotating motor to an individual spectrometer's configuration is relatively simple; a small mirror or prism attached to the motor can be programmed to scan across the breadth of a plume at any desired speed or resolution (Fig. 5; Edmonds et al., 2003).

Such automated scans, with scan duration and angular range more easily constrained than with manual scans, may be especially useful at volcanoes where traverses beneath the plume are not possible. However, as with COSPEC scans, problems and uncertainties in determining the height of and distance to the plume persist when using a single instrument.

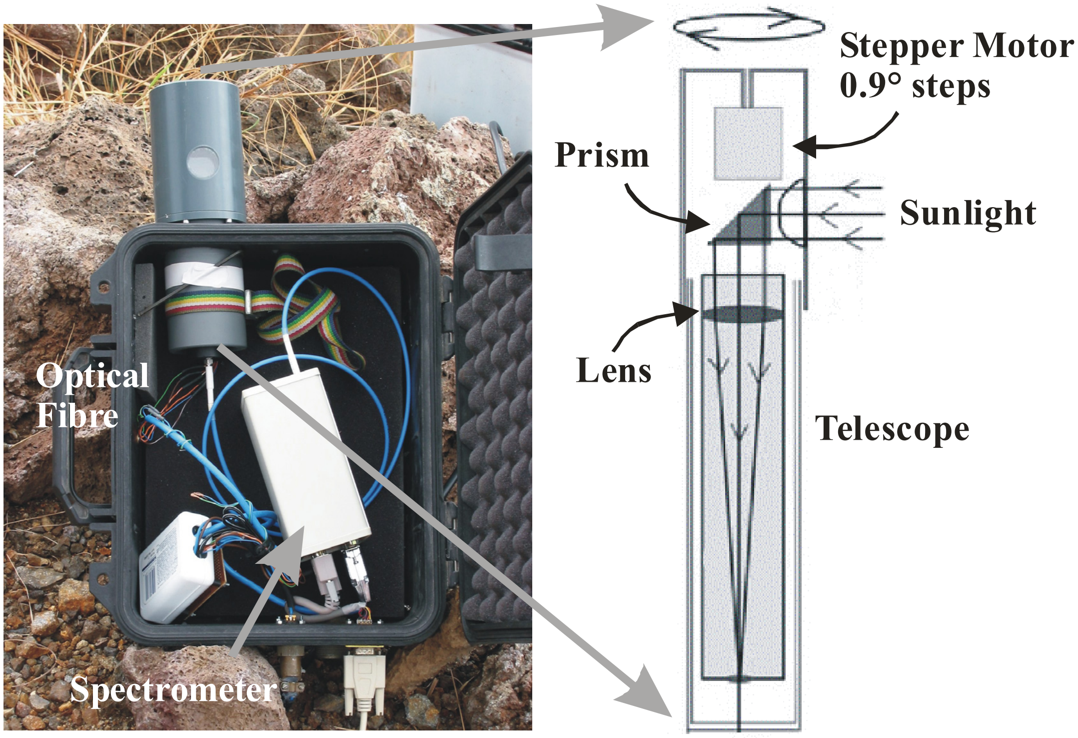

Fig. 5. An example of a scanning DOAS-based spectrometer. Modified from Edmonds et al., 2003.

3.4. Spectrometer Networks

At restless volcanoes, continuous or near-continuous monitoring of the activity is desirable. While instruments such as tiltmeters and seismometers produce high-resolution, real- or near-real-time data and are easily deployed remotely, the COSPEC requires the presence of an operator. COSPEC data are produced only in discrete scans or traverses, and these data also require post-processing (Edmonds et al., 2003). The new spectrometer systems, with their computer integration, real-time data output, and ability to log measurements using telemetry, provide an opportunity to produce high-resolution, continuous SO_2 flux data.

One means by which near-continuous SO_2 measurements can be collected is to modify spectrometers, such that they perform regular scans of a volcano's plume. Automated scanning spectrometer networks have been employed already at Mt. Etna, Italy (McGonigle et al., 2003; Salerno et al., 2004), Stromboli, Italy (Salerno et al., 2004), Soufrière Hills volcano, Montserrat (Edmonds et al., 2003; Young et al., 2003) and White Island, New Zealand (Miller et al., 2006). At Soufrière Hills, two spectrometer systems (Scanspecs) were placed on the downwind slopes of the volcano. Each of the instruments was outfitted with a stepper motor to rotate a prism, which reflects light into the spectrometer's telescope (Fig. 5), allowing for the spectrometers to repeatedly scan the SO_2 plume eight hours per day. As a consequence of the angular scans, data are in the form of SO_2 slant columns rather than vertical columns. For a single angular scan, derived plume size is ambiguous; a high, wide plume of the same concentration as a lower, smaller plume would generate the same signal in the dataset as the smaller plume if they cover the same angular range. Accordingly, accurate SO_2 flux calculations for scanning systems require knowledge of plume height, in addition to other variables such as distance to the plume, plume width, SO_2 slant column concentration, and plume speed. The use of the second, time-synchronized spectrometers allows for determination of plume height; each of the two instruments scans through the plume and the peak concentration pathlength measured is taken to represent the physical center of the plume. For both spectrometers, the angle corresponding to the maximum concentration measurement is extended such that the intersection of the projections from the two spectrometers indicates the position of the plume (Fig. 6). Simple trigonometry yields the plume height, azimuth, and distance from each spectrometer to the plume. Once the distance to the plume is known, the angular range over which an SO_2 signal was obtained can be used to find the width of the plume (perpendicular to the direction of the plume's maximum concentration). Once the plume's apparent width is known, total flux of SO_2 can be calculated as with traditional traverses, substituting the slant column amounts for a traverse's vertical column amounts (Edmonds et al., 2003).

While this type of automated scanning had proven to be successful at Mt. Etna, Stromboli, Soufrière Hills and White Island, it may need to be carefully considered for other types of volcanic terrain. At Kīlauea or Masaya, for example, the generally low topography results in plumes that are quite often very close to ground level. In such cases, the plume's geometry may not permit full scans of the plume; some segments of the plume may appear to be below the horizon, introducing additional error to the technique.

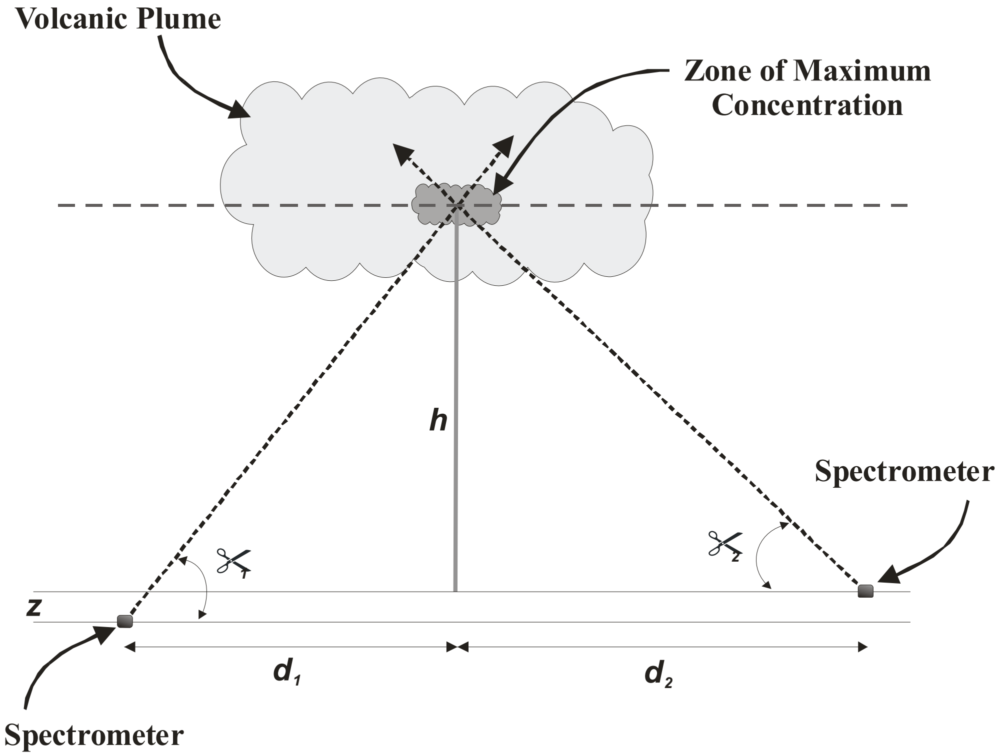

Fig. 6. Calculating plume height and position from the SO_2 concentration and angular data from two fixed scanning spectrometers. Angles α_1 and α_2 are determined from the scan angles coinciding with peaks in SO_2 concentration during one revolution. Tan α_1 = h/d1 and tan $\alpha 2$ = (h+z)/d_2, where h is plume height and z is any elevation difference between the locations of the two spectrometers. The known horizontal distance between the spectrometers is equal to d_1+d_2. Modified from Edmonds et al., 2003.

3.5. Alternative Network Configurations

At volcanoes where plume and topography geometries are not conducive to scanning, but automated, real-time, semi-permanent SO_2 monitoring is desired, other possibilities can be considered. The new generation of instruments, at present, possesses fields of view of ~20-44 mrads (~1.1-2.5°), which limits the instruments to viewing only small segments of plumes at any given time. By making the field of view significantly broader through the use of a 'fish-eye' lens, for example, a small number of stationary instruments aligned perpendicular to the plume's propagation could sense the entire width of the plume (Fig. 7). Each spectrometer would record an average path length concentration for the whole of its respective sector of the plume's cross-section. The multiple spectrometers would yield continuous data across the width of the plume, allowing for real-time flux measurements with the same temporal resolution as the spectral acquisitions, usually 300-1000 ms (Horton et al., 2006). Data from the stationary network would represent integrated instantaneous cross-sections, allowing for the derivation of instantaneous plume fluxes. Alternatively, several stationary spectrometers with a traditional small field of view may be sufficient to continuously monitor volcanic plumes. Arranged in a similar fashion to the proposed 'fish-eye lens' network, the instruments would not measure the entire breadth of the plume, but may provide enough data to closely approximate the plume's true cross- section (Fig. 8). As with any new network, the accuracy of such a measurement configuration relative to the established standards of scanning and traversing the plume should be carefully examined.

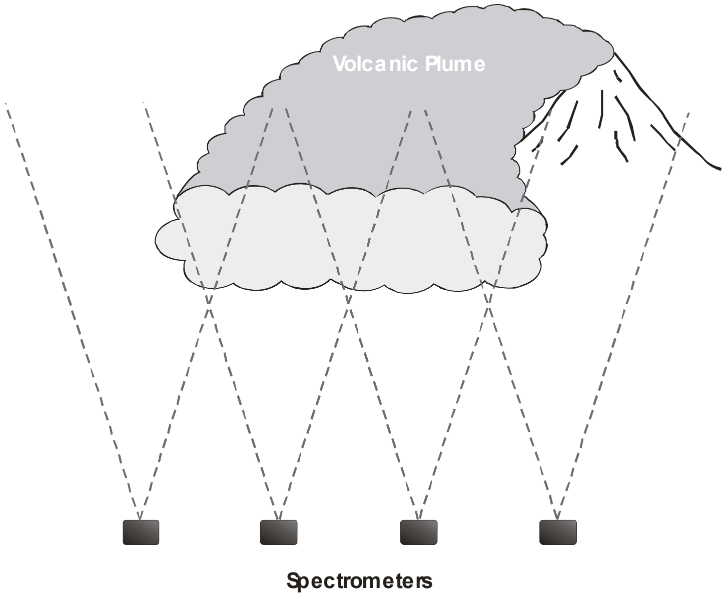

Fig. 7. Schematic diagram of a degassing volcano, plume with cross-section, and a possible configuration for proposed network of stationary wide-angle miniature spectrometers, with their expanded fields of view delineated by dotted lines.

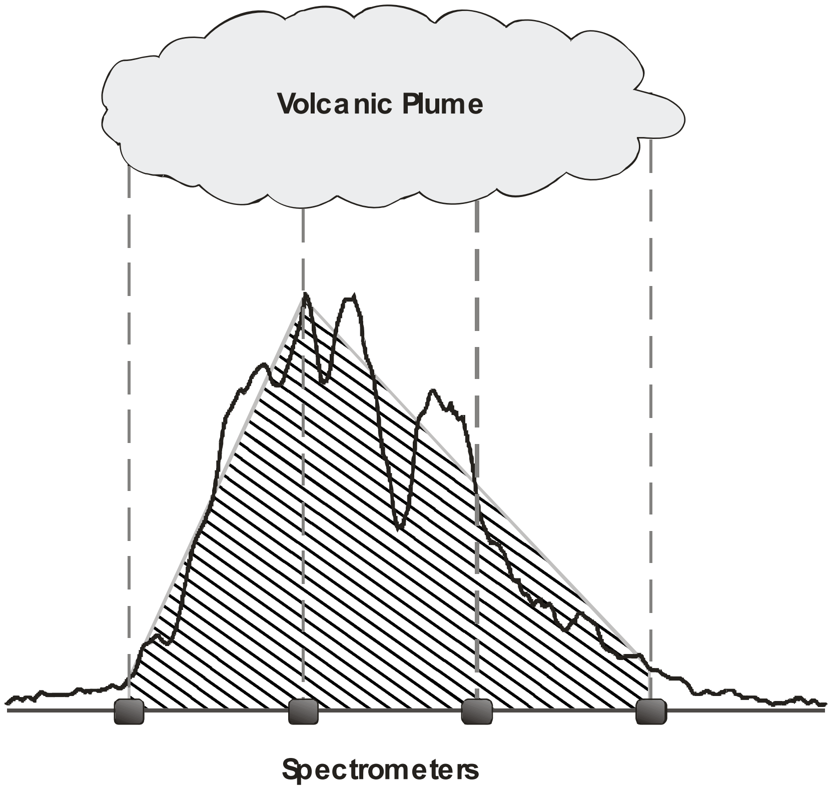

Fig. 8. Example of a network of stationary spectrometers with conventional fields of view, represented by dashed lines. Data from collection points closely approximate the plume shape and average concentration pathlength of a traditional traverse. An example traverse is represented by a dark curve, and the plume approximated from the four spectrometers is marked by the gray-striped area.

3.6. 2D Plume Imaging

Alternatives to multiple one-dimensional gas sensors are recently developed methods by which to image volcanic plumes in two dimensions. Whereas traditional 'mini-DOAS'-type systems utilize a linear CCD array, the IDOAS (Imaging DOAS) employs a 2D CCD array (Bobrowski et al., 2006). The added spatial dimension of the array, combined with the instrument's scanning capabilities, allows for construction of a map of the plume's spatial distribution of SO_2 approximately every 15 minutes. Digital photography has also been used recently as a means by which to obtain images of SO_2 plumes. Bluth et al. (2007) and Mori and Burton (2006) incorporate ultraviolet bandpass filters into digital camera system in order to detect UV absorption by volcanic SO_2. Images can be obtained at rates far exceeding those of an IDOAS (one image every 5-10 s), and from the images, SO_2 amounts may be determined. Though a new technique, UV photography shows promise for volcanic remotes sensing, as it is able to see large portions, if not all, of plumes and plume speed determination requires only visual tracking of structures over a series of images. Possible applications range from traditional SO_2 flux calculations to studies of plume behavior during transport away from the vent (Bluth et al., 2007).

4. ADDITIONAL REMOTE GAS SENSING TECHNIQUES

DOAS technology is most commonly used in the form of ground-based or airborne spectrometers, as with the new generation of miniature spectrometer systems, but recent satellite technology has expanded the applicability of DOAS to volcanic SO_2 sensing. The Global Ozone Monitoring Experiment (GOME) sensor aboard the second European Remote Sensing satellite, while primarily targeting ozone, possesses the capability to detect atmospheric absorption by other trace gases. The same UV absorption signal used to determine quantities of SO_2 from ground-based spectrometers can also be used to derive SO_2 concentrations from GOME data (Bortoli et al., 2000). Other satellite sensors, such as the Total Ozone Mapping Spectrometer (TOMS), also detect volcanic SO_2. However, while TOMS has provided a reliable record of SO_2 output from large eruptions over the past two decades, the technology is limited. TOMS is generally capable of detecting only stratospheric SO_2; smaller eruptions or quiescent degassing that reach only into the troposphere go largely undetected (Carn et al., 2003). A newer tool, the Ozone Monitoring Instrument (OMI), part of the payload on NASA's EOS/Aura satellite, is based on technology similar to that of TOMS. Compared to TOMS, OMI offers a greater degree of spatial and spectral resolution, as well as lower detection limits for SO_2, which translates to the ability to detect smaller eruption clouds than TOMS, and even passive degassing (Carn, 2006). Algorithm development for SO_2 retrievals and validation with ground-based measurements are still underway for OMI.

More recent developments in satellite sensing of volcanic SO_2 have been in the infrared portion of the electromagnetic spectrum. AVHRR, MODIS, ASTER, SEVIRI, and VISSR are all instruments with bands capable of detecting, through manipulation of infrared data, elevated levels of SO_2 into the upper troposphere (Tupper et al., 2004). Recently, Carn et al. (2005) demonstrated the capacity of AIRS to sense volcanic SO_2 from moderate eruptions at altitudes as low as ~6 km. The high spectral resolution of AIRS also may permit detection of volcanic CO_2, but detection of any passively degassed plumes remains problematic as a result of the low spatial resolution (13.5 km/pixel) of the instrument (Carn et al., 2005).

LIDAR, a laser-based technology, and its derivative Differential Absorption LIDAR (DIAL) provide another ground-based option in the remote observation of volcanic gases. Laser pulses are directed at the plume, and based on the return time for back-scattered radiation of specific wavelengths, concentrations of various gases can be determined (Weibring et al., 2002; McGonigle and Oppenheimer, 2003). Plume speed may also be integrated with LIDAR data to determine fluxes of SO_2.

Fourier Transform Infrared spectroscopy (FTIR) has also been employed as an alternative method for monitoring volcanic gases. FTIR yields data in the form of a wide variety of molar ratios, such as SO_2/HCl and HCl/HF, and, in combination with SO_2 flux data from a COSPEC, FLYSPEC, or 'mini-DOAS'-type systems, the data may be used to determine fluxes of trace gases from volcanoes, as at Masaya volcano (Horrocks et al., 2003). While FTIR equipment is quite bulky and difficult to deploy in most field conditions, and may eventually be surpassed by instruments utilizing DOAS technology in the sensing of gases other than SO_2, it is still a key means by which volcanic H_2O and CO_2 may be detected. Because of the high levels of H_2O and CO_2 in the ambient atmosphere, their volcanic components are often quite difficult to detect; the ability of FTIR technology to do so makes possible a fuller quantification of the extent of volcanic degassing.

As with FTIR, detection of H_2O and CO_2 is now also possible with a new development in volcanic gas monitoring: a multi-sensor system (MSS) comprising a temperature/humidity sensor, electrochemical sensors, and an infrared analyzer. The instrument measures H_2O, CO_2, and SO_2, and the addition of specialized sensors and alkaline filter techniques would expand the range of measurable compounds to H_2S, H_2, Cl, and F (Shinohara, 2005). Despite the number of components, the system is portable and weighs only 5 kg. This portability is necessary, as the MSS verges on being a type of direct sampling rather than remote sensing in the manner of instruments like COSPEC or LIDAR. Measurements with the MSS must be taken either very near, or in, the degassing crater, or within the plume itself, as from a helicopter or airplane.

5. AEROSOLS

Regardless of the method utilized in the monitoring of volcanic SO_2, one consideration is the possibility of conversion of SO_2 to sulfate aerosols:

$$OH + SO_2\ (+O_2,\ H_2O) \rightarrow H_2SO_4 + HO_2$$

Should such conversion occur downwind of the degassing volcano, fluxes of SO_2 measured by spectrometers may underestimate the true, original flux from the volcano's vent. To mitigate such underestimates, some SO_2 monitoring campaigns have begun to integrate the use of a sun photometer to measure aerosol quantity along with SO_2 flux (Porter et al., 2002; Mather et al., 2004; Nadeau, 2006). Satellites such as TOMS and AIRS also possess the capability to sense atmospheric aerosols in tandem with volcanic SO_2 measurements (Carn et al., 2005).

6. BEYOND SO_2

The original COSPEC was developed as a means to monitor industrial emissions of both SO_2 and NO_X. This proved to be of limited use in the volcanic sector, as simultaneous measurements of multiple gases with the COSPEC is impossible as a result of the correlation mask spectroscopy methodology. Data concerning nitrogen species within volcanic plumes are of minimal value in terms of volcanic hazard evaluation relative to information on SO_2 fluxes and, therefore, SO_2 data collection took priority during COSPEC field campaigns

The advent of DOAS as a means to study volcanic plumes has introduced the possibility of obtaining, with relative ease, data on gas species other than SO_2. DOAS technology differs from

COSPEC in its ability to record full spectra, thereby obtaining information for wavelengths of ultraviolet radiation in the atmospheric absorption windows of gases other than SO_2. Given calibration cells or laboratory spectra for other species within the range of wavelengths of the instrument, fluxes of a variety of volcanic gases can theoretically be derived (Resonance Ltd., 2006).

Attempts have recently been made to quantify BrO at Soufrière Hills volcano (Bobrowski et al., 2003), NO_2 at a power plant in the U.K. (McGonigle et al., 2004), and H_2S at Solfatara and Vulcano volcanoes in Italy (O'Dwyer et al., 2003) through the use of 'mini-DOAS'-type spectrometer systems and laboratory reference spectra. Similar studies could also theoretically be conducted using calibration cells of different gas species for data retrieval.

7. CONCLUSION

The COSPEC proved invaluable to volcanic SO_2 research for nearly three decades. In recent years, the technology has been surpassed by smaller, cheaper, more versatile instrument systems based on differential optical absorption spectroscopy (DOAS) techniques. The new systems are capable of SO_2 sensing in the manner of traditional COSPEC methods, and also have the potential for more flexible applications in the field of volcano monitoring. Both single scanning instruments and networks of these small spectrometer systems can now be deployed alongside seismometers and GPS stations on volcanoes worldwide, presenting an opportunity for continuous geochemical monitoring. The compact size, computer integration, and low cost of miniature UV-spectrometers are significant improvements over COSPEC attributes, and along with the capability of DOAS methodology to detect a variety of volcanic gas species, will likely lead to miniature spectrometer systems playing an increasingly important role in the future of volcanic gas sensing.

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