

Integrating Petrologic and Remote Sensing Perspectives on Magmatic Volatiles and Volcanic Degassing

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The volatile constituents— H_2O , CO_2 , and S, and Cl—play an important role in the generation, evolution, and eruption of magma. Knowledge of the abundance and flux of these volatiles is important for understanding the hazard implications of volcanic activity, explosive eruptive behavior of volcanoes, recycling of volatiles in subduction zones, formation of magmatic-hydrothermal ore deposits, additions of volcanic gases to Earth's atmosphere, and potential climatic impacts of large volcanic eruptions.

Over the past 25 years, new developments in technology have led to major advances in our ability to measure the fluxes of volatiles released from erupting volcanoes. At the same time, new micro-analytical techniques have made it possible to measure the pre-eruptive concentrations of magmatic volatiles through the use of tiny melt inclusions trapped in phenocrysts in volcanic rocks. Comparison of the data acquired from these different perspectives has highlighted some major gaps in our understanding of both the abundance of the different volatiles, and the ways in which they are transported through magmatic systems and released to the atmosphere by degassing.

Remote Sensing Approaches

Sulfur dioxide (SO_2) is the easiest of the main magmatic volatiles to measure in volcanic plumes, because its concentration is relatively high compared to ambient atmospheric values. However, CO_2 and H_2S can also be analyzed using aircraft-based gas analyzers and sensors [e.g., *McGee et al.*, 2001], and H_2O , HCl, HF, CO_2 , and several other species can be measured using Fourier-Transform Infrared (FTIR) spectroscopy [e.g., *Burton et al.*, 2000]. Remote sensing of SO_2 is achieved using absorption bands of SO_2 gas in the ultraviolet (UV) and infrared (IR) regions of the electromagnetic spectrum. Numerous remote measurements of SO_2 fluxes from active volcanoes have been

made, including both ground-based and airborne use of the UV correlation spectrometer (COSPEC) since the early 1970s, and the satellite-based Total Ozone Mapping Spectrometer (TOMS; Figure 1) since 1978 [*Krueger et al.*, 2000; see: <http://skye.gsfc.nasa.gov>].

Building on the measurements of volcanic SO_2 from space pioneered by the UV TOMS instruments, recent years have seen launches of IR sensors that have augmented the SO_2 mapping capabilities available to volcanologists [*Realmuto et al.*, 1997]. The Moderate Resolution Imaging Spectroradiometer (MODIS) carried by NASA's Earth Observing System (EOS) Terra and Aqua satellites, and the Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER) on EOS/Terra offer the potential to quantify the significant non-eruptive or passive SO_2 plumes emitted by many quiescent volcanoes. Measurements of larger SO_2 clouds in the upper troposphere and stratosphere, traditionally the domain of TOMS, can also now be accomplished several times daily using data from the High Resolution Infrared Sounder (HIRS) on NOAA's polar-orbiting satellites [*Prata et al.*, 2003]. Moreover, further developments are anticipated in 2004 with the launch of the Ozone Monitoring Instrument (OMI) on EOS/Aura. OMI will be the successor to TOMS and is expected to be able to routinely measure volcanic SO_2 , including passive plumes in the atmospheric boundary layer.

An advanced multispectral IR sensor (SEVIRI; <http://www.esa.int/msg/pag4.html>) on the European Space Agency's new Meteosat Second Generation (MSG) satellite, expected to be operational very soon, will provide geostationary coverage of volcanic clouds over Europe and Africa. Geostationary platforms stare at the same places and allow for very frequent coverage, which will result in sensing of short-lived eruptions, and the IR bands are ideal for SO_2 and ash sensing. Despite having greater volcanic activity, North and South America have no comparable IR coverage, and from 2004 to ~2012, will lose important geostationary ash detection capability on the GOES satellite. After 2012, a new sensor called ABI (Advanced Baseline Imager), which is currently being

developed, will fly over the Americas and provide robust coverage of volcanic clouds.

Measurement of volcanic clouds by multi-spectral techniques has demonstrated the utility of examining clouds from different perspectives. There is really no one method or spectral range that is universally appropriate for evaluating SO_2 in volcanic plumes. Each sensor is limited by its spatial, temporal, or spectral resolution, and thus, observes only a portion of a plume, or is best suited for evaluating a plume for a short period of time or under a restricted range of meteorological and volcanological conditions. Interference by co-emitted ash, meteorological clouds, background reflectivity, and the presence or generation of aerosols, ice, and other gases in the plumes all affect retrievals to varying degrees, and are unique to each setting and event.

Knowledge of meteorological conditions (relative humidity, temperature, wind vectors) also plays a vital role in evaluating gas fluxes, particularly for effusive emissions. The scientific utility of remotely sensed data is often dependent on the ability to measure or model surrounding environmental conditions. A multi-spectral approach is vital because volcanic clouds evolve, sometimes rapidly, both in physical and chemical character during the first few days of emission into the atmosphere. With the addition of new tools and techniques has come the discovery of many new complexities and processes affecting the fate of volcanic emissions.

Recent Developments in Ground-based and Airborne Methods

Ground-based and airborne techniques for monitoring volcanic emissions are also advancing. One important development is the application of miniature UV spectrometers to SO_2 retrieval [*Galle et al.*, 2002]. These low-cost, lightweight devices will probably supplant the COSPEC as the instrument of choice for proximal surveillance of degassing volcanoes.

Another development is an airborne system devised by the U.S. Geological Survey that directly measures concentrations of CO_2 , SO_2 , and H_2S by extraction sampling of plumes. The system includes an infrared gas analyzer, chemical sensors, a closed-path FTIR instrument, a COSPEC, a high-precision barometer, a temperature probe, and a GPS receiver. Measurements sample a range of altitudes along traverses orthogonal to plume (wind) direction or along orbits circling a volcano (Figure 2A).



access is difficult, and wet climates, snowpack, and glaciers foster hydrothermal and ground waters that can scrub acid magmatic volatiles (SO_2 , HCl, HF), masking degassing at depth during critical early stages of unrest. Campaigns to date have constrained CO_2 emission rates from as low as 200–300 tons/day at Mammoth Mountain and Mount Baker, to as high as 8000–9000 tons/day at Popocatepetl and Kilauea [McGee *et al.*, 2001; Gerlach *et al.*, 2002, and references therein].

Except in rare cases, the gas emission monitoring techniques covered in this article are not likely to measure effectively the diffuse degassing flux of volcanoes. Although tending to be smaller in magnitude than vent degassing, diffuse degassing is an important component of the total gas emission at many volcanoes, and in some instances—Mammoth Mountain being a notable example—it can be the dominant mode of degassing. Eddy covariance methods and soil gas efflux surveys offer alternative monitoring techniques for diffuse degassing.

Petrologic Studies of Magmatic Volatiles

With the techniques summarized above, volcanologists now have at their disposal a toolkit of instruments capable of quantifying emissions of SO_2 and other volatiles at a range of temporal and spatial scales. Used in concert, these techniques have the potential to produce a vastly improved and expanded volcanic degassing data set. Complementary to these developments in remote sensing, new sampling and micro-analytical techniques have made it increasingly possible to get information on magmatic volatiles directly from samples of volcanic material. Because the solubility of most volatile components is strongly pressure-dependent, magmas exsolve gas during ascent to the surface.

To understand eruptive behavior, it is important to know the total volatile content of magma during its pre-eruptive storage within the Earth's crust. An important source of information on dissolved volatiles in magmas is through the analysis of glass inclusions trapped inside of crystals. When melts crystallize, the crystals grow imperfectly, causing small blebs of melt to be trapped inside of the crystals. If the magma erupts and cools rapidly, then these trapped melt inclusions quench to glass. Because the crystalline host for the inclusions is relatively rigid, they act as tiny pressure vessels and prevent the trapped melt from degassing, even though the bulk magma decompresses to surface pressure during eruption. For this reason, trapped melt (glass) inclusions commonly retain their original dissolved volatiles. Melt inclusions are now routinely analyzed for many volatile species (H_2O , CO_2 , S, Cl, F) using infrared and Raman spectroscopy, electron microprobe, and secondary ion mass spectrometry (SIMS).

Comparison of Remote Sensing and Petrologic Perspectives—The Excess Sulfur Problem

It is possible to use data for S in melt inclusions to make an estimate of how much S would

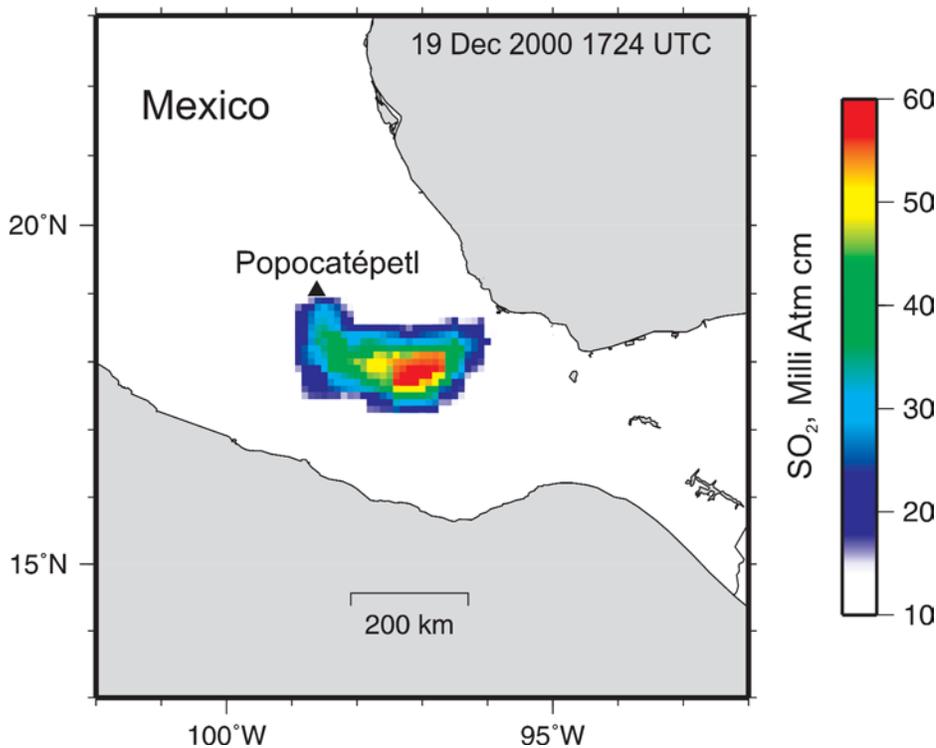


Fig. 1. Photograph of Popocatepetl volcano forcefully venting gases from its summit lava dome on 19 December 2000 (top panel), along with satellite SO_2 data (SO_2 column amount in milli atm cm or Dobson Units) from the Earth Probe TOMS instrument collected approximately 2.5 hours after the photograph was taken (lower panel). The volcanic cloud observed by TOMS contained ~26,000 tons of SO_2 ($\pm 30\%$). Some of the highest SO_2 emission rates and lava dome growth rates recorded during Popo's current eruption were observed in December 2000. (photo courtesy of the Centro Nacional de Prevención de Desastres [CENAPRED].)

Data for all gases are recorded at 1-s intervals and tagged with latitude, longitude, altitude, temperature, and pressure. Gas concentrations in plume cross-sections are contoured with mapping software (Figures 2B, 2C) and used with in-flight wind data to calculate emission rates.

The system is modular and can be deployed on fixed-wing aircraft and helicopters, or mounted on vehicles for ground-level measurements, making it suitable for a wide range of volcanic environments. It is applicable to volcanoes of the western United States and Alaska, where locations are remote, ground

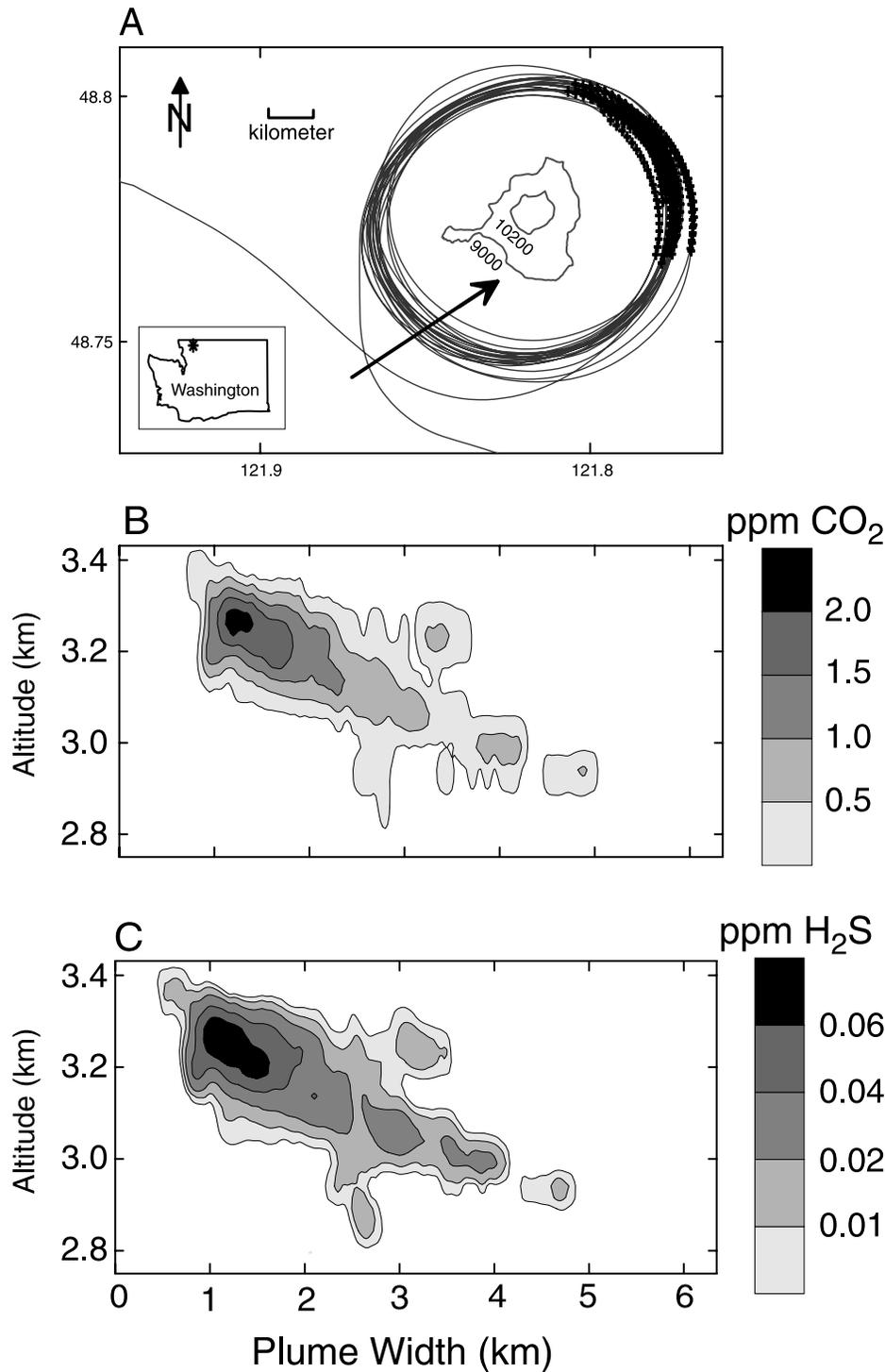


Fig. 2. (A) Location map with GPS flight path for a 21-orbit airborne survey of CO₂ and H₂S at Mount Baker, Washington on 13 September 2000. Upper elevations of Mount Baker are contoured. Solid triangle marks the location of Sherman Crater, a site of several large fumaroles. Arrow shows wind direction. Dark portions of flight paths indicate where the plume was intersected downwind during the survey. Contoured cross-sections of plume CO₂ (B) and H₂S (C) are 3 km downwind of Mount Baker and corrected for orbital curvature. The cross-section views are upwind toward the volcano. Contours indicate ppm concentrations above background, which are approximately 368 ppm for CO₂ and essentially zero for H₂S. The CO₂ emission rate for this survey is about 190 tons/day; that for H₂S is about 6 tons/day [McGee et al., 2001].

be released in a volcanic eruption based on knowledge of the total volume of erupted magma. However, such estimates assume that the only S released during an eruption is S

that was originally dissolved in the silicate liquid (melt) portion of the magma. Comparison of such "petrologic" estimates based on melt inclusion data with SO₂ emissions measured

for recent eruptions by remote sensing techniques has led to a conundrum, known as the "excess" sulfur problem, concerning S mass balance during volcanic eruptions [Andres et al., 1991].

Concentrations of dissolved S in magmas before eruption, as estimated from melt inclusion data, are commonly far too low (by 1 to 2 orders of magnitude) to account for the total mass of SO₂ released during the eruption, as measured by remote sensing techniques (Figure 3). Excess S is observed in most eruptions for which remote sensing and melt inclusion data are available, and in particular, is characteristic of explosive eruptions of intermediate and silicic magma in subduction zone settings [Andres et al., 1991]. In contrast, basaltic eruptions from divergent plate boundaries and hot spots, such as Hawaiian and Icelandic volcanoes, often do not show evidence of excess S emissions (Figure 3).

It has commonly been assumed that magmas of intermediate-to-silicic composition only become vapor-saturated during shallow ascent and emplacement, during eruptive decompression, or during advanced (pegmatitic) stages of plutonic crystallization. However, numerous lines of evidence from petrologic, remote sensing, and volcanic gas data suggest that the "excess" S problem is caused by the presence of an exsolved C-O-H-S vapor phase in the magma before eruption [Luhr et al., 1984; Andres et al., 1991; Westrich and Gerlach, 1992; Wallace, 2003]. That is, the concentrations of dissolved volatiles are sufficient to cause saturation of the magma with a multi-component vapor phase, which would presumably be present as bubbles dispersed throughout magma in crustal storage reservoirs. As a result, eruptions of silicic magma can release large amounts of SO₂ derived from pre-eruptive exsolved vapor, despite the fact that such magmas generally have very low concentrations of dissolved S. Techniques for quantifying the mass fraction of exsolved vapor in magma before eruption are thus important for understanding SO₂ flux data used for forecasting and monitoring eruptions, and for assessing the potential climatic impacts of volcanic eruptions.

The large amounts of volatiles implied by these estimates suggest that exsolved vapor accumulates in the apical regions of magma bodies during repose periods between eruptions, or may get trapped below hydrothermal caps above the body. The presence of an exsolved vapor phase in crystallizing magma bodies may also play an important role in triggering volcanic eruptions. Remote sensing data frequently observe spatial separation of SO₂ and ash in explosive eruptions, and improved frequency of IR satellite data is likely to clarify whether a gas-rich phase is released early in explosive events, an idea consistent with observations [Rose et al., 2003].

Sources of Volatiles in Crustal Magmatic Systems

Isotopic studies of S and CO₂ in intermediate-to-silicic magmas have demonstrated that a large component of these volatiles is

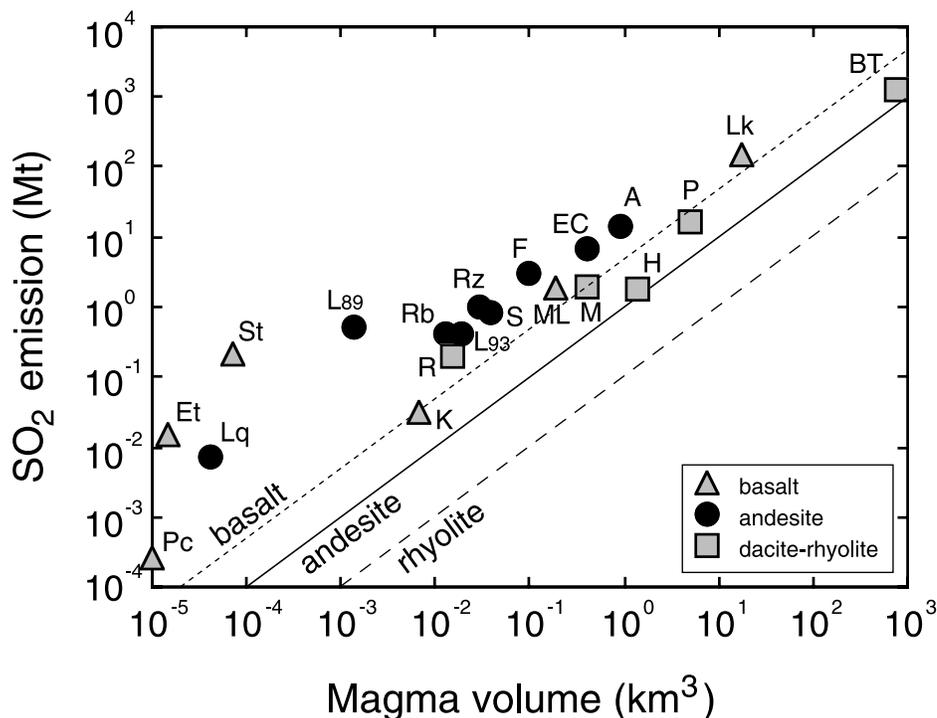


Fig. 3. Volcanic SO_2 emissions, in megatons ($1 \text{ Mt} = 10^9 \text{ g}$), versus total volume of erupted magma. SO_2 emissions were measured using remote sensing methods except for the eruption of the Bishop Tuff (BT; see references in Wallace, 2003). Most of the remote sensing data are from TOMS, except for the data for Fuego (determined by COSPEC), Agung (estimated from stratospheric optical depth measurements), and Laki (estimated from information on atmospheric turbidity). Uncertainties of $\pm 50\%$ for both SO_2 emission and eruptive volume are less than to slightly greater than the size of the symbols. Uncertainties in SO_2 emission data are generally considered to be about $\pm 30\%$ for the TOMS data and $\pm 20\%$ to 50% for COSPEC. Data are shown for the following eruptions: (A) Agung, 1963, (EC) El Chichón, 1982, (Et) Etna, 1975, (F) Fuego, 1974, (H) Cerro Hudson, 1991, (K) Kilauea, annual average, (L89) Láscaz, 1989, (L93) Láscaz, 1993, (Lk) Laki, 1783–1784, (Lq) Lonquimay, 1989, (ML) Mauna Loa, 1984, (M) Mount St. Helens, 1980, (Pc) Pacaya, 1972, (P) Pinatubo, 1991, (Rb) Rabaul, 1994, (R) Redoubt, 1989–1990, (Rz) Ruiz, 1985, (S) Spurr, 1992, and (St) Stromboli, annual average. References to data sources can be found in Wallace [2003]. Shown for comparison are predicted relationships between SO_2 emission and eruptive volume for rhyolitic (dashed line), andesitic (solid line), and basaltic (short dashed line) melts calculated by assuming that the only SO_2 released during the eruption is from S that is originally dissolved in silicate melt. Note that the SO_2 emissions for all eruptions, with the exception of Mauna Loa, Kilauea, and Laki, are at least 1 order of magnitude greater than predicted for the appropriate bulk composition by syneruptive degassing of dissolved S only. This clear discrepancy over many orders of magnitude in eruptive volume indicates that a major additional source of S must be involved in all of these eruptions.

mantle-derived. The ultimate source of S and CO_2 must therefore be from mafic magma, because it is well established that silicic magma reservoirs are created and sustained through long-term intrusion of mantle-derived basaltic magma into the crust. It should be noted, however, that in the case of subduction zones, some portion of the mantle S and C (and almost all H_2O) may ultimately have been recycled from subducted sediments and altered oceanic crust.

Andesitic, dacitic, and rhyolitic magmas in crustal reservoirs are probably vapor-saturated due to recharge and underplating by basaltic magma that is saturated with multi-component C-O-H-S vapor. In this regard, vapor saturation of intermediate to silicic magmas can be viewed as the inevitable consequence of the relatively high CO_2 concentration of basaltic magmas coupled with relatively low CO_2

solubility in silicate melts at crustal pressures. Vapor transfer from underplated basaltic magma into an overlying silicic magma body is probably a slow process that occurs during the long repose periods that separate eruptive episodes. In systems where intrusion of basaltic magma into the lower crust generates silicic magma by partial melting and/or fractional crystallization, transfer of volatiles into mid- to upper-crustal silicic magma chambers may be a very complex process compared to systems in which basaltic magma intrudes directly into a silicic magma reservoir. However, in either case, if the S and CO_2 in crustal magmatic systems are largely derived from the mantle via basaltic magma, then the flux of these volatiles should be directly related to the basaltic magma flux.

The importance of mantle-derived basalt as a source of volatiles is exemplified by mafic

cinder cones surrounding Popocatepetl ("Popo") volcano in central Mexico. Magne- sian andesites erupted explosively from Popo during the current eruption (December 1994 to 2002) have phenocryst assemblages and major- and trace-element systematics that indicate mixing of mafic and silicic magma, and SO_2 and CO_2 emissions have both been very high [see references in Wallace, 2003]. Quaternary mafic cinder cones in the region surrounding Popo provide samples of possible mafic end members involved in mixing. Olivine-hosted melt inclusions from these cones contain 1000–6000 ppm S, 250–2100 ppm CO_2 , and 1.3–5.2 wt% H_2O [Cervantes and Wallace, 2003]. The high S contents suggest that mafic magma recharging into the Popo system provides an abundant source of S that may explain the very large SO_2 emissions from the current eruption. The H_2O and CO_2 contents are sufficiently high that these mafic magmas would be vapor-saturated at the depths of a shallow crustal magma chamber beneath the volcano, so recharging mafic magmas could transfer significant volatiles during recharge events.

Future Directions

Future directions in remote sensing of volcanic clouds feature multi-spectral retrievals to examine the interactions and removal rates of different species in the clouds, and to provide more comprehensive evaluations of emission fluxes throughout various phases (e.g., explosive versus effusive) of volcanic activity. Progress in understanding budgets of the major magmatic volatiles requires integration of remote sensing and volcanic gas data on fluxes of CO_2 , SO_2 , H_2S , H_2O , and HCl with petrologic data on temperature, pressure, oxygen fugacity, and dissolved volatiles in both differentiated magma stored in crustal reservoirs and mafic magma recharging these systems.

Experimental studies of vapor-melt partitioning of volatiles are also needed, particularly for S. Such studies are crucial for developing and calibrating thermodynamic models for predicting vapor saturation and degassing of a multi-component vapor phase over a wide range of conditions. Seismic studies of sub-volcanic magma bodies can also potentially play an important role in the problem of determining volatile budgets, by providing evidence for the presence and abundance of exsolved vapor in magma stored within the Earth's crust.

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Coastal Observatory Investigates Energetic Current Oscillations on Southeast Florida Shelf

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Coastal circulation along the southeast Florida shelf is strongly related to the dynamics of the Florida Current. The current follows the steep bottom terrain along the shelf break separating the deep ocean from the coastal zone (Figure 1 a, b). Energetic and complex flow regimes arise along the current's western edge, causing transient features such as sub-meso-scale eddies [Lee and Mayer, 1977; Shay et al., 2000] and energetic internal oscillations [Moore, 1975; Soloviev et al., 2003], which affect mixing between the shelf and deeper ocean waters. Understanding of these regimes is important for modeling and prediction of the coastal circulation on the southeast Florida shelf and description of western boundary current/continental shelf interactions.

A striking feature of the circulation on the shelf off southeast Florida is the energetic baroclinic supertidal oscillation, which produces 10-hr period current velocity oscillations with a 0.5-m s⁻¹ amplitude. At the same time, current velocity oscillations induced by the barotropic tide usually do not exceed about 0.05-m s⁻¹ amplitude. Spectral analysis of the sea level signal reveals the expected semi-diurnal tidal peak close to the 12-hr time period (Figure 1c). The corresponding peak, however, is not seen in the velocity spectrum; instead, a 10-hr peak is prominent (Figure 1d).

Two years of continuous observations on the southeast Florida shelf reported in Soloviev et al. [2003] suggested that this oscillation is observed primarily during summer months. A 4-year data set collected at the same location, which is now available for analysis, clearly demonstrates that this phenomenon has a seasonal cycle, which is the subject of interannual variability. On average, the kinetic energy

associated with the oscillation appears much higher in the summer season than at other times throughout the year (Figure 2); there is considerable variation between years. Interannual differences can be seen in Figure 3, where the squared current velocity magnitudes within the 8- to 12.5-hour band for each summer from 1999–2002 are shown.

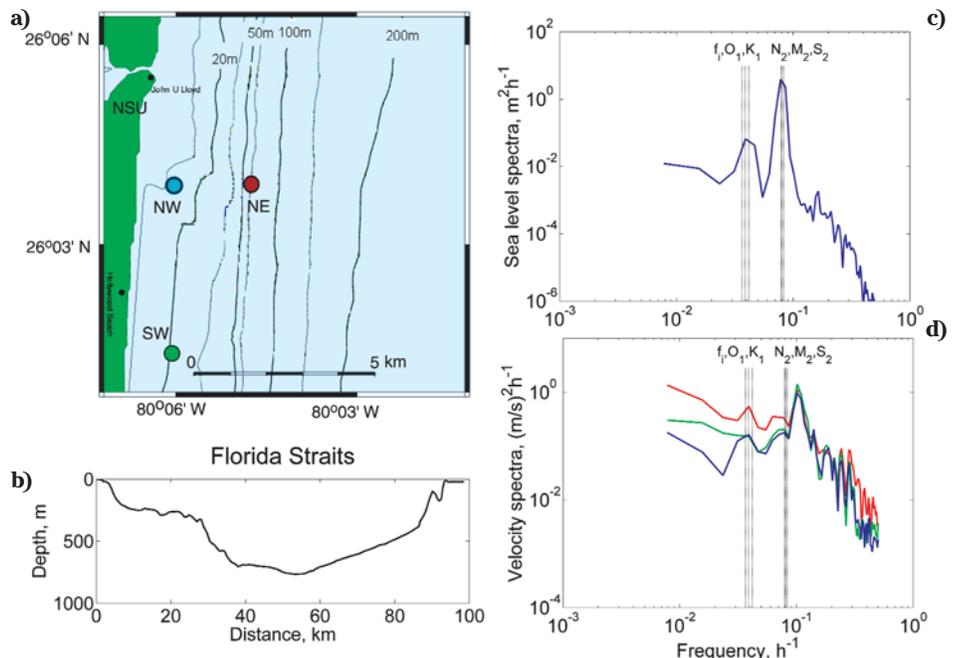


Fig. 1. (a) Map indicating location of three mooring buoys in the SFOMC range. (b) Cross-section of Florida Straits. Sea level (c) and velocity (d) spectra in the SFOMC range. Note a 2-hr “blue” shift of the tidal peak in the velocity spectra compared to the sea-level spectrum. Inertial frequency f_i (27.34 hrs) and tidal constituents O_1 (25.82 hrs), K_1 (23.93 hrs), N_2 (12.66 hrs), M_2 (12.42 hrs), and S_2 (12 hrs) are marked by vertical dashes. The spectral curves in subplots (c) and (d) are color-coded according to the color of the mooring symbols in subplot (a).

Variability of the coastal currents affect societally relevant issues such as navigation, acoustics, search and rescue operations, beach erosion, and coastal pollution from sewage plants, harmful algal blooms, and oil spills [Peters et al., 2002]. These important practical issues could require that the baroclinic current oscillation on the southeast Florida shelf become more fully understood and its frequency and importance in nature be assessed.

Project Background

The South Florida Ocean Measurement Center (SFOMC) was established in 1998 through a partnership involving government and academia. A congressional grant, along with a