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SO$_2$ emissions from basaltic eruptions, and the excess sulfur issue

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[1] Volcanic SO$_2$ can affect the Earth's environment. Where no direct measurements of SO$_2$ in the atmosphere are available, a petrologic method of assessing sulfur release from the magma must be used. However, in studies of arc-derived eruptions, satellite-based measurements of SO$_2$ emissions using Total Ozone Mapping Spectrometer (TOMS) data are orders of magnitude greater than those calculated petrologically, implying that a separate S-rich gas phase in the magma chamber may be responsible for the excess sulfur. We test whether this applies in other settings. For Icelandic and Hawaiian basalts we find that petrologic SO$_2$ values are comparable to measurements of SO$_2$ by TOMS. Thus, for non-arc basalts, the petrologic method gives reliable estimates of SO$_2$ released. The implied absence of excess sulfur in non-arc basaltic magmas is a reflection of source magma conditions, notably lower fO$_2$ and volatile contents than arc magmas, inhibiting the exsolution of a S-rich gas prior to eruption. *INDEX TERMS:* 0370 Atmospheric Composition and Structure: Volcanic effects (8409); 8409 Volcanology: Atmospheric effects (0370); 8494 Volcanology: Instruments and techniques; 8499 Volcanology: General or miscellaneous. **Citation:** Sharma, K., S. Blake, S. Self, and A. J. Krueger (2004), SO$_2$ emissions from basaltic eruptions, and the excess sulfur issue, *Geophys. Res. Lett.*, *31*, L13612, doi:10.1029/2004GL019688.

1. Introduction

[2] Volcanic SO$_2$ emissions can have significant effects on the Earth's atmosphere and biosphere. Evaluating the amounts and mechanisms of SO$_2$ release is thus crucial to predicting the environmental consequences of volcanism. The mass of SO$_2$ emitted by volcanic eruptions can be determined using a variety of techniques. Two techniques currently in use are the petrologic method [*Devine et al.*, 1984] and satellite measurements of SO$_2$ releases (TOMS) [*Bluth et al.*, 1997; *Carn et al.*, 2003]. Comparisons between TOMS SO$_2$ measurements and petrologic SO$_2$ estimations for six arc eruptions (listed in Figure 1) of basaltic (e.g., Galunggung) to dacitic (e.g., Pinatubo) magma compositions show that the TOMS SO$_2$ values exceed the petrologic estimations by one to two orders of magnitude. *Westrich and Gerlach* [1992] and *Wallace* [2001; 2003] suggested that this discrepancy can be attributed to a separate S-bearing volatile phase accumulating at depth in the magma system prior to eruption—so-called excess sulfur. Estimates of sulfur release from other basaltic eruptions, such as flood

basalts, in non-arc settings could also perhaps be affected by this same process and be underestimated. Because comparisons between petrologic estimates of sulfur release for non-arc basaltic eruptions with independent SO$_2$ measurements (e.g., satellite data) are lacking, the significance of such petrologic results is uncertain. In this paper, we investigate whether there is evidence for excess sulfur in non-arc basaltic volcanism using improved petrologic estimates and satellite data.

1.1. The Petrologic Method

[3] The petrologic method assumes that glass inclusions in phenocrysts represent volatile-bearing liquid trapped during crystal growth in the magma chamber. If there is no S-bearing gas in the magma chamber, then the sole source of the degassed S is the melt phase of the magma. The mass of sulfur released is then given by the difference in concentration of S between inclusions (C_{inc}) and the degassed matrix glass (C_{matrix}) of the volcanic rock, multiplied by the mass of erupted liquid [*Devine et al.*, 1984]. In the cases to be considered in this paper, SO$_2$ detected by TOMS originated from explosive release of magma that generated scoria with little or no syn- or post-eruptive crystallization, so the mass of erupted liquid is $M_v (1 - W_{xtls})$ where M_v is the mass of volcanic rock and W_{xtls} is the mass fraction of crystals. The petrologic method therefore estimates the mass of S released as

$$M_s = M_v(1 - W_{xtls})[C_{inc} - C_{matrix}] \qquad (1)$$

Taking account of the molecular weights of S and SO$_2$, the mass of SO$_2$ released is $2M_s$.

1.2. Satellite Measurements - Total Ozone Mapping Spectrometer (TOMS)

[4] The TOMS dataset provides a 22-year record of measurements of SO$_2$ released into the atmosphere by over 200 eruptive events [*Carn et al.*, 2003]. The instrument measures back-scattered solar UV radiation in six different wavelength bands; an iterative algorithm is used with radiative transfer tables to retrieve the amount of SO$_2$ [*Krueger et al.*, 1995]. The latest instrument and algorithm in use (version 7) has a minimum detection limit of ∼5 kt and SO$_2$ estimates have errors in the range of ±10–30%. Errors near the lower end of this range are associated with eruption plumes containing little or no volcanic ash.

[5] TOMS measures the total mass of SO$_2$ in entire eruption clouds once per day. This mass is the erupted mass less the amount lost by conversion to sulfate. The erupted mass of SO$_2$ in brief explosive eruptions is determined by
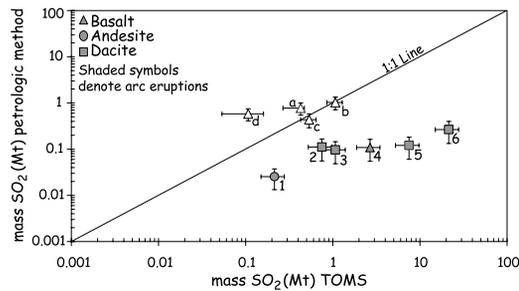
Figure 1. TOMS SO$_2$ measurements vs. calculated petrologic SO$_2$ for the 4 eruptions described here, Krafla 1984 (a), Mauna Loa 1984 (b), Hekla 1980 (c), & Hekla 2000 (d). Also shown are data from 6 arc eruptions (Redoubt 1989 (1), Nevado del Ruiz 1985 (2), Mount St Helens 1980 (3), Galunggung 1982 (4), El Chichon 1982 (5), & Pinatubo 1991 (6)). Petrologic and TOMS SO$_2$ data for the 6 arc eruptions are taken from *Blake* [2003] and references therein. Error bars on the non-arc petrologic SO$_2$ measurements are calculated 2σ errors. Error bars on the arc petrologic SO$_2$ values are ±50%, a maximum estimate.

extrapolating daily cloud totals back to the time of eruption. However, for longer-lasting eruptions, the daily total contains new SO$_2$ released after the previous satellite overpass and old SO$_2$ remaining from earlier days. Summing of daily totals can produce an overestimate of total eruption mass. Winds sometimes make it possible to identify the older SO$_2$ cloud by spatial separation. With low wind speeds the loss rate, which varies by a factor of 30 from the surface to the stratosphere, must be explicitly considered in evaluating total eruption mass [*Krueger et al.*, 1996].

2. Results

[6] Lava and tephra samples were collected from four non-arc mafic eruptions with well-constrained magma volumes and TOMS data: Krafla 1984, Mauna Loa 1984, Hekla 1980, and Hekla 2000. Olivine- and plagioclase - hosted glass inclusions and microlite-free matrix glass shards were analyzed using a Cameca SX-100 electron microprobe (the method is described in the auxiliary material[1]). The glass inclusions range in size from 30 to 100 μm and are crystal-free, homogenous glasses. Care was taken to avoid inclusions containing bubbles or fractures. Histograms of sulfur data for all matrix glasses and glass inclusions are presented in Figure A1, and mean glass chemistry is listed in Table A1 (see auxiliary material[1]). To estimate the mass of SO$_2$ released through melt degassing using the petrologic method, only glass inclusions with the same major element composition as the matrix glass were used, thereby ensuring that the selected inclusions represent non-degassed equivalents of the degassed matrix liquids, as is assumed in the derivation of the petrologic method. Glass inclusions were statistically chosen by calculating the Euclidean distance function, D, between the normalized (100% volatile-free) major element composition of each inclusion and the mean normalized matrix

glass composition. For each inclusion we calculated $D = \sqrt{\sum [(x_i - \mu_i)/\sigma_i]^2}$, where x_i is the wt% of the ith oxide in an inclusion, and μ_i and σ_i are the mean and standard deviation wt% of the ith oxide in the matrix glass. In computing the value of D, we include the 10 major elements; values ≤ 6.3 indicate that the inclusion is statistically identical to the mean matrix glass composition at the 95% confidence level. Only these statistically similar inclusions (D values ≤ 6.3) were averaged to obtain C_{inc} for the petrologic calculations (Table 1).

[7] Density values in Table 1 (for conversion of eruption volume to magma mass) were taken from *Gronvold et al.* [1983] for Hekla (unless otherwise stated). In the case of scoria and lava samples from Krafla and Mauna Loa, volumes were measured using a water displacement method. Dividing the weight by the volume yielded the density which was proportioned according to the ratio of lava to scoria in the eruption products. Crystal mass fractions were measured by point counting of representative back-scatter electron images, except for Hekla 1980 [*Gronvold et al.*, 1983] and Mauna Loa [*Lockwood et al.*, 1987]. Table 1 summarizes the sulfur, magma mass, and crystal fraction data for the studied eruptions.

2.1. Krafla 1984

[8] The 1984 eruption of Krafla began on September 4 with the opening of a fissure segment 6 km north of Leirhnjúkur, within 1 hour the fissure reached its full length of 8.5 km [*GVN*, 1984]. Fire-fountaining characterized the start of the eruption but within 24 hours this activity waned and steady lava flow emplacement began. The eruption lasted 2 weeks and a total bulk volume of 0.134 km^3 of tholeiitic basalt was erupted [*Rossi*, 1997]. TOMS detected a 0.1 Mt SO$_2$-rich plume produced during the first day of eruption; this plume later moved ESE towards Norway. Then, one week into the eruption, TOMS instruments detected additional SO$_2$ in plumes from Krafla. Sulfur dioxide plumes were tracked for 11 days and the total amount of SO$_2$ emitted to the atmosphere as detected by TOMS is 0.4 +0.04/−0.15 Mt; in this case the errors are not symmetric about the mean [*Bluth et al.*, 1993]. Pre-eruptive sulfur concentrations range from 1380–1850 ppm with a mean value of 1590 ppm. Degassed sulfur values are between 400–500 ppm. The petrologic estimate of SO$_2$ released to the atmosphere through melt degassing is 0.64 ± 0.19 Mt.

2.2. Mauna Loa 1984

[9] The 1984 Mauna Loa eruption began on March 25 with a 12-hr summit eruption before activity migrated to the North East Rift Zone where fire fountains 10–50 m high prevailed, forming channel-fed lava flows. Activity lasted 23 days and the total volume of erupted tholeiitic lava was 0.22 km^3 [*Lipman and Banks*, 1987]. TOMS instruments detected an SO$_2$-rich plume from the volcano during 16 days of activity. The total amount of SO$_2$ emitted to the atmosphere was 1.0 ± 0.2 Mt as detected by TOMS and 0.85 ± 0.26 Mt calculated petrologically. The petrologic value is based on pre-eruptive sulfur contents of ~1000 ppm and degassed sulfur values of ~160 ppm.

2.3. Hekla 1980

[10] The 3-day Hekla eruption of 1980 began on August 17 with simultaneous plinian and lava extrusion

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[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2004GL019688.

Table 1. Summary of Petrologic Method Data and Results, TOMS Data and fO₂ Values (ΔNNO)

Eruption	C_{inc} [n]	C_{matrix} [n]	Mass (kg) [Density kg/m³]	W_{xtls}	Petrologic SO₂ (Mt)	TOMS SO₂ (Mt)	ΔNNO
Krafla 1984	0.159 ± 0.017 [8]	0.049 ± 0.007 [13]	2.95 × 10¹¹ [2200]	0.02	0.64 ± 0.19	0.4 + 0.04/−0.15	0.3
Mauna Loa 1984	0.101 ± 0.011 [8]	0.016 ± 0.008 [13]	5.06 × 10¹¹ [2300]	0.01	0.85 ± 0.26	1.0 ± 0.20	−0.1
Hekla 1980	0.091 ± 0.008 [12]	0.034 ± 0.017 [7]	3.22 × 10¹¹ [700 [a], 2400 [b]]	0.03	0.36 ± 0.12	0.50 ± 0.10	−0.4
Hekla 2000	0.090 ± 0.009 [12]	0.031 ± 0.007 [7]	4.14 × 10¹¹ [600 [a], 2400 [b]]	0.01	0.48 ± 0.14	0.10 ± 0.05	−0.3

[a]Indicates tephra/scoria bulk density.

[b]Indicates lava density. Bulk density of Hekla 2000 tephra was provided by G. Larsen (unpublished data, 2002). Errors on C_{inc} and C_{matrix} measurements are 1σ, & errors associated with petrologic SO₂ values are 2σ. [n] = no. of analyses.

phases, producing basaltic-andesite fall deposits and lava flows. A 15-km-high eruption column quickly formed and deposited 0.06 km³ of tephra in 5 to 6 hours, mainly during the first two hours. Lava (0.12 km³) was erupted from an 8-km-long fissure cutting through the Hekla ridge [*Gronvold et al.*, 1983]. Although this was a moderately short Hekla eruption, the SO₂ cloud was relatively long-lived. Satellites tracked the plume for 6 days, with 0.5 ± 0.10 Mt of SO₂ [*Carn et al.*, 2003] measured by TOMS. The average pre-eruptive sulfur concentration is ~900 ppm and degassed sulfur values are ~300 ppm, giving a petrologic estimate of 0.36 ± 0.12 Mt of released SO₂.

2.4. Hekla 2000

[11] The eruption of Hekla in 2000 began on February 26 with the opening of a 6-km-long fissure along the SW flank of the Hekla ridge. Initial activity in the central part of the fissure generated a sub-Plinian eruption column 11−12 km high. Following this, effusive activity began with the emplacement of lava flows interspersed with strombolian fire-fountain activity and phreatomagmatic explosions along the length of the fissure [*Ólafsdóttir et al.*, 2002]. The eruption intensity rapidly decreased after 5 days but activity lasted for 11 days. The total volume of basaltic-andesite magma erupted was 0.18 km³, 0.01 km³ of which was the tephra layer deposited in the first 12 hours of eruption [*Haraldsson et al.*, 2002]. TOMS images from 27 February show a narrow plume arcing away from Hekla before moving north towards Greenland and, finally, east over Norway. At the end of the eruption, the plume drifted east in a band along the Russian and Norwegian coasts of the Barents Sea [*GVN*, 2000]. TOMS instruments tracked an SO₂ plume for 3 days. The total amount of SO₂ released to the atmosphere was 0.1 ± 0.05 Mt. A mean value around 900 ppm for pre-eruptive sulfur and ~300 ppm for the degassed sulfur yields a petrologic estimate of 0.48 ± 0.14 Mt SO₂ released through magma degassing.

[12] Table 1 lists results for all analyzed eruptions, and Figure 1 compares the two types of SO₂ measurements for the eruptions listed here and for 6 arc eruptions (listed in Figure 1) where both data types are available.

3. Discussion

[13] As we see from Figure 1, the TOMS SO₂ measurements for the non-arc eruptions are comparable to the petrologic estimations of SO₂, indicating that there was no excess S-rich gas phase present in the underlying magma systems prior to eruption. This is a possible reflection on the magma source conditions and tectonic setting. Data here suggest that magma composition plays little part in

the separation of sulfur into an excess gas phase. The arc eruptions vary in bulk composition from basaltic (Galunggung) to dacitic (Mt. St Helens, Pinatubo), yet all have excess sulfur. The common factor connecting them is their arc setting.

[14] For Hekla 2000, the petrologic SO₂ value is slightly higher than the SO₂ detected by TOMS. One explanation for this is that a large amount of sulfur was released through lava degassing during the later stages of the eruption, generating low-altitude SO₂ plumes undetectable by TOMS. However, a more likely scenario is that TOMS did not detect part of the SO₂ plume. TOMS estimates are based on the scattering of solar UV radiation, thus the high-latitude location of Hekla combined with the low solar energy during the polar winter may have contributed to the low TOMS value for the 2000 eruption [*Rose et al.*, 2004].

[15] Magma source conditions typically found in arcs (such as higher H₂O contents and fO₂) may be the crucial factor in the sequestering of sulfur from the melt into an excess S-bearing fluid phase. For the arc eruptions listed here, log fO₂ values were obtained from the literature. For the Icelandic and Hawaiian eruptions, log fO₂ values in Table 1 were calculated using the model of *Suguwara* [2001]. This model uses equilibrium plagioclase and olivine compositions together with temperature and pressure values to compute log fO₂, to an accuracy of ±0.8 log units. We express fO₂ in relation to the NNO buffer (Ni-NiO reaction), with ΔNNO defined as the log fO₂ value of the magma minus the log fO₂ value of the NNO buffer at the same temperature [*Carmichael and Ghiorso*, 1990].

[16] When ΔNNO is plotted against the ratio TOMS SO₂/ petrologic SO₂-a measure of excess S, we find that the data form two fields (Figure 2). The arc eruptions plot with high excess sulfur ratios and positive ΔNNO values. The non-arc
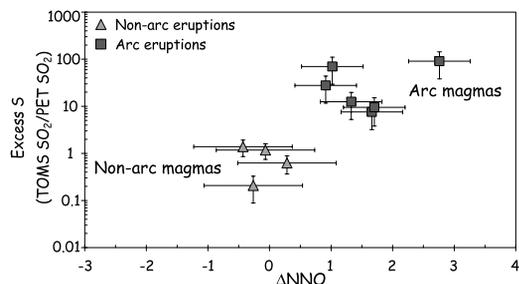

Figure 2. ΔNNO vs. TOMS SO₂/Petrologic SO₂ (excess S).

eruptions fall within the lower left sector of the graph regardless of the magma composition, showing low excess S ratios and low positive and negative ΔNNO values.

[17] Figure 2 indicates that oxygen fugacity (fO$_2$) and thus tectonic setting has a greater influence than magma composition on the presence of excess S. The oxygen fugacity controls the solubility and partitioning of species with different oxidation states, e.g., S. Low fO$_2$ conditions indicate a reducing environment and may stabilize an immiscible sulfide phase. High fO$_2$ conditions indicate oxidizing environments and may stabilize a SO$_2$-bearing gas phase, such that oxidized magmas release pre-existing SO$_2$ gas in addition to SO$_2$ exsolved from the melt during the eruption. Experimental studies of sulfur partitioning between melt and fluid [e.g., *Scaillet et al.*, 1998] demonstrate that oxidized magmas have a greater tendency to sequester sulfur into a fluid phase at depth, whereas reduced magmas have low sulfur partition coefficients, such that nearly all the sulfur remains in the melt and does not get partitioned into a fluid phase. Based on the comparison between TOMS SO$_2$ and petrologic SO$_2$ data, magmas that erupt excess sulfur are more oxidized than log fO$_2$ ~ -9.8 (ΔNNO $\sim$0.5).

4. Conclusions

[18] Using a petrological approach in which we selected inclusions that are statistically similar to the initial matrix composition, we have shown that for the recent eruptions of Mauna Loa, Krafla and Hekla the petrologic estimates of SO$_2$ are within error of the TOMS SO$_2$ measurements, indicating that if there was any fluid phase present in these magma systems prior to eruption it was sulfur-poor. The lack of a separate S-rich gas reflects the links between fO$_2$, melt H$_2$O content, magma source conditions and sulfur solubility. Oxidized, water-rich arc magmas have an excess S-rich gas phase prior to eruption whereas more reduced H$_2$O-poor non-arc magmas do not. One exception to this may be non-arc CO$_2$-rich alkali basalts. As CO$_2$ will inevitably promote fluid saturation at depth, it is quite possible that these magmas could generate a S-rich gas phase depending on their oxidation state.

[19] Our results have shown that a petrologic approach for calculating SO$_2$ released through melt degassing is a reliable method for quantifying SO$_2$ releases in tholeiitic to transitional basaltic volcanism, in contrast to the situation in many arc eruptions. This result is also supported by the similarity between the petrologic estimates of SO$_2$ release from the AD 1783 eruption of Laki (tholeiite with ΔNNO of -0.3) and independent estimates of the mass of sulfate aerosol loading from this eruption [*Thordarson et al.*, 1996]. While the Laki case involves indirect methods of assessing the sulfate burden, the large mass of SO$_2$ released ($\sim$120 Mt) makes the comparison reasonably robust. We therefore conclude that petrologic estimates of SO$_2$ emissions from ancient basaltic eruptions, e.g., flood basalts, are likely to be realistic values.

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