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A review on volcanic gas compositions related to volcanic activities and non-volcanological effects

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ABSTRACT: Volcanic gases have been monitored to predict eruptions since their compositions are strongly affected by volcanic activities. Here, volcanic gas geochemistry is reviewed with the main focus placed on the potential use of volcanic gases as eruption precursors. First, volcanic activities are differentiated into four eruptive cycles: eruptive, post-eruptive, inter-eruptive, and pre-eruptive periods. Second, we discuss geochemical factors affecting volcanic gas compositions: magma origin and degassing, hydrothermal interactions, and meteorological variability. Third, volcanic gas data in previous studies are revisited to address the practical aspects in utilizing volcanic gas compositions to predict eruptions. As a magma is depressurized due to its uprising, volcanic gases become progressively exsolved. Generally, the fluxes of volcanic gases (e.g., CO_2, SO_2, H_2S, etc.) increase considerably close to eruptions. Also, CO_2/SO_2, SO_2/HCl, and SO_2/HF ratios in volcanic gases tend to decrease at the imminence of eruptions. Nonetheless, such trends may not be encountered due to both kinetically limited degassing and secondary effects including hydrothermal interactions and meteorological variability. Due to the site specificity of the aforementioned effects, a comprehensive understanding of volcanic gas chemistry requires the knowledge of magma vents and reservoirs as well as hydrothermal systems below volcanoes. Accordingly, the integrated use of volcanic gas data with geophysical and geomorphological information can warrant a better prediction of eruptions.

Key words: volcanic gases, eruption, eruptive cycles, magma degassing, hydrothermal interactions

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1. INTRODUCTION

Volcanic eruptions occur as a result of a number of factors including the injection of a new magma, the collapse of volcanic edifices, earthquakes, and the infiltration of meteoric water (Cañón-Tapia, 2014). A volcano may erupt explosively or quiescently depending on both the type of a magma and its volatile contents (Roggensack et al., 1997). Often, unexpected volcanic eruptions are natural disasters producing not only the direct effects (e.g., the damage of human lives and properties) but also the indirect consequences (e.g., climate changes) (Robock, 2000). If volcanic eruptions are predicted in a timely manner, such adverse effects can be minimized and/or prevented.

To date, volcanologists have tried to predict volcanic eruptions using seismological observation (Ratdomopurbo and Poupinet, 2000; Matthews et al., 2009; Inza et al., 2014), electronic distance measuring (EDM) (Pingue et al., 1998; Young et al., 2000; Ramírez-Ruiz et al., 2002; Saepuloh et al., 2013), magnetic and electric field observation (Sasai et al., 2002; Zlotnicki et al., 2003; Lillis et al., 2008), temperature monitoring (Yamashina and Matsushima, 1999; Cigolini et al., 2009; Feseker et al., 2009), and volcanic gas monitoring (Giggenbach et al., 2001; Aiuppa et al., 2007; Vaselli et al., 2010; Inguaggiato et al., 2011a, 2011b, 2013). Among them, volcanic gas monitoring can provide important clues to volcanic eruptions since the gas compositions are subject to changes in response to volcanic activities. Furthermore, the analysis of volcanic gases can help understand the chemical nature and tectonic environment of a magma (Halmer et al., 2002; Wallace, 2005). Also, since some volcanic gases (e.g., SO_2) may pose serious environmental concerns such as acid rain, volcanic gas data are critical in assessing the aftermaths of eruptions (Parnell and Burke, 1990; Delmelle et al., 2002).

After produced by the (partial) melting at the mantle or the lower crust, a magma ascends to the near surface by buoyancy. As the magma comes close to the surface, the solubility of volatile components in it decreases. Eventually, volatile components are

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exsolved from the magma to accumulate in the upper part of its reservoir, with some of them discharged to the surface as volcanic gases (Proussevitch and Sahagian, 2005). Among volcanic gas components, H_2O is the most abundant component, with CO_2 being usually the second (Bruno et al., 2001). In general, volcanic gases are divided into acidic gases (CO_2, SO_2, H_2S, HCl, and HF), non-condensable gases (H_2, He, N_2, Ar, CH_4, and CO), metallic trace elements, and organic matter (Cadle, 1980; Symonds et al., 1988; Chin and Davis, 1993). As volcanic eruptions become imminent, the emission of volcanic gases becomes stronger due to the increasing degassing from a magma. Often, the concentrations of CO_2, SO_2, and H_2S in volcanic gases have been used to predict volcanic eruptions (Gerlach, 1979, 1980; Bruno et al., 2001; Allard, 2010; Werner et al., 2013). Due to the ranged solubility in magmas, volcanic gases show different tendencies to be exsolved. While less soluble (more volatile) components such as CO_2 show an intense emission from the early stage of eruptions, the emission of more soluble (less volatile) components including HCl and HF is not evident until the later stage. Consequently, the concentrations of less soluble components increase more quickly than those of more soluble components, leading to distinct temporal profiles of volcanic gas compositions. For this reason, the concentration ratios including CO_2/H_2O, CO_2/SO_2, SO_2/HCl, and SO_2/HF have been widely used as eruption precursors (Aiuppa, 2009; Aiuppa et al., 2009; Allard, 2010; Notsu and Mori, 2010; Stremme et al., 2011; López et al., 2013: Werner et al., 2013). Table 1 lists volcanic gas parameters that have been used to predict eruptions. In Table 1, not only the absolute concentrations of individual gas components but also the relative concentrations between them have been employed. Also, changes in the isotopic compositions of specific volcanic gases (e.g., δD and $\delta^{18}O$ of water vapor) have been utilized to assess volcanic activities (Shevenell and Goff, 2000). Although not further discussed, geochemical monitoring can be performed on volcanic ash, snow, subsurface water, hot spring water, and rocks entrapping volatile components of magmas (Allard et al., 2000; Gislason et al., 2002; Toutain et al., 2003; Mackenzie and Canil, 2008).

As aforementioned, volcanic gas compositions undergo significant changes as eruptions become imminent (Fischer et al., 1996). However, such changes are not only due to magmatic processes (e.g., depressurized degassing), but also due to other factors including hydrothermal interactions and meteorological variability (Shimoike and Notsu, 2000; Symonds et al., 2001). Furthermore, even at the same volcano, the gas compositions may vary with the location and method of gas measurements (Stoiber and Rose, 1970). Accordingly, it is not simple and straightforward to foretell volcanic eruptions based on changes in volcanic gas compositions. This study delineates the eruptive cycles of volcanoes in section 2, which is followed by the discussion on the geochemical processes responsible for changes in volcanic gas compositions in section 3. To gain insight into the relationship between volcanic activities and volcanic gas compositions, previous case studies are revisited in section 4.

2. ERUPTIVE CYCLES

Knowledge on the eruptive cycles of volcanoes is required to better understand and predict eruptions. Depending on volcanic gas compositions, temperature, and seismic activities, volcanic activities can be divided into eruptive, post-eruptive, inter-eruptive, and pre-eruptive periods (Menyailov, 1975). Below is the discussion on each period of the cycles.

An eruptive period is the stage which is characterized by magma eruptions along with vigorous volcanic activities. It can contain one or more eruptions, with the relatively quiescent intervals between eruptions lasting for days or months (Menyailov, 1975). During this period, most volcanoes show elevated discharges of CO_2, SO_2, and halogen gases, with the emission sequence generally in the order of CO_2, SO_2, and halogen gases (Aiuppa, 2009; Aiuppa et al., 2009). Consequently, the concentration ratios of CO_2/SO_2, SO_2/HCl, and SO_2/HF are usually higher during the early eruptive period. At Stromboli volcano in Italy, the concentration ratios of CO_2/SO_2 during the eruptive period were twice as high as those during the inter-eruptive period (see Fig. 1). At Redoubt volcano in Alaska, volcanic eruptions occurred in 1902, 1966–1968, 1989–1990, and February to April in 2009, during which

Table 1. Volcanic gas parameters used to predict eruptions

Volcanoes	Volcanic gas parameters	References
Masaya, Nicaragua	SO_2/HCl, SO_2	Duffel et al. (2003)
St. Helens, Washington	HCl, δD-H_2O, $\delta^{18}O$-H_2O	Shevenell and Goff (2000)
Etna, Italy	CO_2, SO_2	Bruno et al. (2001)
Redoubt, Alaska	CO_2, SO_2, H_2S, CO_2/SO_2	Werner et al. (2013)
Stromboli, Italy	CO_2, H_2, He	Carapezza et al. (2004)
Bezymianny, Russia	SO_2, H_2O/CO_2, CO_2/S, S/HCl	López et al. (2013)
Colima, Mexico	SO_2	Zobin et al. (2008)
Izu-Oshima. Japan	CO_2, O_2, H_2	Shimoike and Notsu (2000)
Tatun volcanic group, Taiwan	CO_2/H_2O	Ohba et al. (2010)

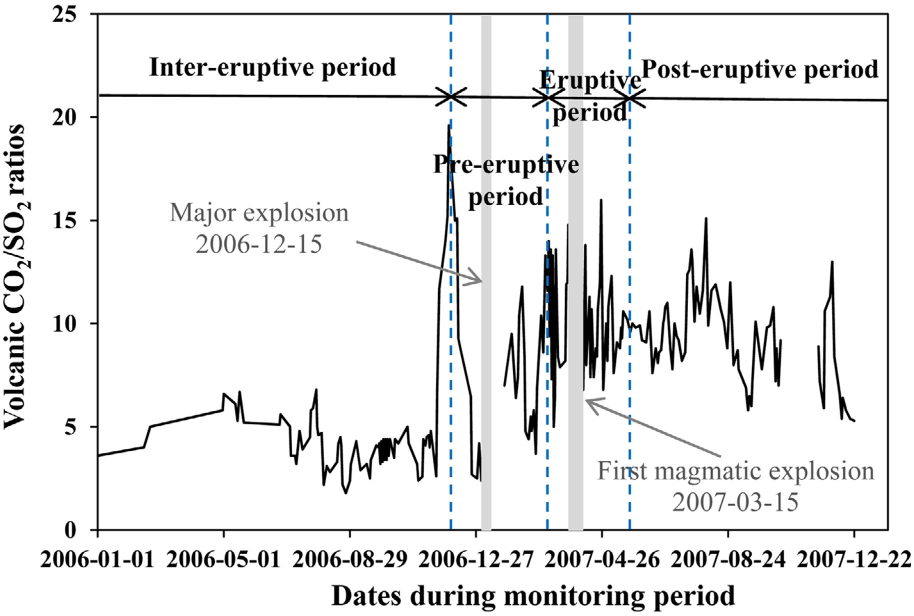

Fig. 1. Eruptive cycles of Stromboli volcano in Italy for the years of 2006–2007 based on volcanic CO_2/SO_2 ratios. This figure is derived from Aiuppa et al. (2009).

elevated CO_2/SO_2 ratios were also observed (Werner et al., 2013).

A post-eruptive period is usually shorter than the other periods. This period is sometimes equivocal since it is lack of distinct features. For example, although a post-eruptive period is generally characterized by high fumarolic temperature, this feature is also noted for the quiescent intervals between eruptions during the eruptive period (Menyailov, 1975). At Stromboli volcano, the post-eruptive period was thought to start from the end of the eruptive period (April 2007) and continued till December 2007 (see Fig. 1). Although magma eruptions ceased during the post-eruptive period, volcanic earthquakes still occurred at Stromboli volcano (Aiuppa et al., 2009).

An inter-eruptive period is much longer than the other periods, and it may last for years to centuries. It is characterized by fumarolic activities such as solfatara, mofettes, and hot springs without magma eruptions (Menyailov, 1975). In case of the eruption of Stromboli volcano in 2007, the inter-eruptive period continued till November 2006, about four months prior to the eruptive period (see Fig. 1). During this period, the ratio of CO_2/SO_2 at Stromboli volcano was found to range from 1.6 to 10.3, which was far lower than the maximum value of 21 on the verge of the eruption. Unlike the post-eruptive period, very few volcanic earthquakes occurred during this period (Menyailov, 1975; Patanè et al., 2007; Aiuppa et al., 2009).

A pre-eruptive period is usually characterized by abrupt changes in fumarolic gas compositions, volcanic earthquakes, and other volcanic activities (Patanè et al., 2007; Aiuppa et al., 2009). This period is of critical importance in predicting volcanic eruptions since it is just ahead of the eruptive period. Sometimes, it is difficult to distinguish this period when volcanic activities become intense during the inter-eruptive period. Unlike the inter-eruptive period, however, this period is accompanied with increases in fumarolic temperature and substantial changes in fumarolic gas compositions (Madonia et al., 2013). During the inter-eruptive period, H_2, HCl, HF, and SO_2 are mainly discharged. On the other hand, the emission of CO_2, SO_2, and H_2 may increase considerably during the pre-eruptive period (Menyailov et al., 1986; Carapezza et al., 2009; Werner et al., 2013). Consequently, the concentrations of sulfur gases and H_2 relative to halogen gases as well as the concentrations of CO_2 and SO_2 become elevated during the pre-eruptive period (Menyailov, 1975). Also, the ratios of CO_2/SO_2 and CO_2/H_2O during this period were found to be higher (Bruno et al., 2001; de Moor et al., 2005; Burton et al., 2007; Carapezza et al., 2009; Giammanco et al., 2013; Werner et al., 2013). Along with the gas compositional changes, volcanic earthquakes during this period become stronger (Bruno et al., 2001; de Moor et al., 2005; Burton et al., 2007; Carapezza et al., 2009; Giammanco et al., 2013; Werner et al., 2013). For the eruption of Stromboli volcano in 2007, the pre-eruptive period spanned from December 2006 to March 2007 (see Fig. 1).

3. FACTORS AFFECTING VOLCANIC GAS COMPOSITIONS

Volcanic gas compositions are influenced by several factors including magma origin and degassing, hydrothermal interactions, and meteorological variability. Thus, all these factors need to be taken into accounts when analyzing volcanic gas data. This section discusses the key factors governing volcanic gas compositions.

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3.1. Magma Origin and Degassing

Volcanic gas compositions from volcanoes with different tectonic environments and magma types are listed in Table 2, in which volcanic gas compositions vary considerably with locality. With some exceptions, volcanic gas temperature at subduction zones (i.e., convergent plate boundaries) is typically below 1,000 °C, but the one at rift zones (i.e., divergent plat boundaries) and oceanic islands (i.e., hotspots) generally exceeds it (see Table 2). Previously, Symonds et al. (1994) found that volcanic gases at subduction zones generally had higher H but lower C and S than those at rift zones and oceanic islands. Consistent with this is the triangular plot of $H_2O/5$-CO_2-S in Figure 2a, in which volcanic gases at subduction zones are typically more enriched with H_2O than CO_2 and S ($SO_2 + H_2S$) compared to those at rift zones and oceanic islands. Also, they reported that volcanic gases at subduction zones were more concentrated with HCl (see Fig. 2b). Accordingly, volcanic gases at subduction zones were characterized by lower volcanic SO_2/HCl ratios (e.g., 0.1–10) than those at rift zones (20–160) (Symonds et al., 1994). To date, no clear relationship between volcanic gas compositions and magma types has been recognized. Nonetheless, while alkali-rich magmas typically have high CO_2 contents, those rich in Mg and Fe contain great amounts of H_2O (Gerlach, 1979). Consistent with this, Etna and Nyiragongo volcanoes, both of which originated from alkali-rich magmas, exhibited relatively high CO_2 contents (see Table 2).

Besides magma compositions, the degassing depth (pressure) is critical for controlling volcanic gas compositions (Edmonds, 2008; Gaillard et al., 2011). In general, magmatic degassing gets stronger with the decreasing depth, thus leading to elevated

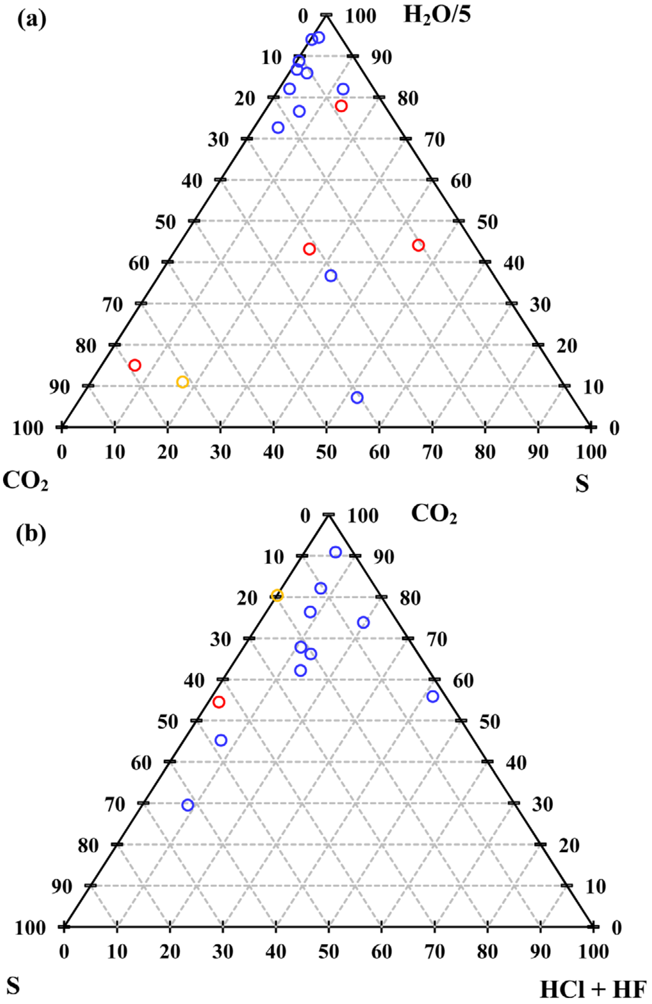

Fig. 2. Triangular plots of $H_2O/5$-CO_2-S (a) and CO_2-S-HCl + HF (b) in volcanic gases. The data are from Table 2. Blue, red, and orange circles correspond to the origins of subduction zones, rift zones, and oceanic islands, respectively.

Table 2. Volcanic gas compositions from various tectonic environments and magma types

Volcanoes	Tectonic environments	Magma types	Temp. (°C)	H_2O	H_2	CO_2	CO	SO_2	H_2S	HCl	HF	References
Etna, Italy	Subduction zone	Hawiite	1000	19.6	–	22.2	–	25.2	3.4	–	–	Gerlach (1979)
St. Helens, USA		Dacite	710	98.9	0.4	0.88	0.003	0.27	–	0.15	0.03	Symonds et al. (1994)
Usu, Japan		Dacite–andesite	690	99.3	0.26	3.85	–	0.23	0.26	0.35	–	Giggenbach et al. (1986)
Showa-Shinzan, Japan		Andesite	800	98.0	0.63	1.2	0.003	0.043	0.0004	0.053	0.024	Giggenbach (1996)
Merapi, Indonesia		Andesite	803	88.7	0.50	5.56	0.024	0.98	0.134	0.608	0.000	Giggenbach et al. (2001)
Ngauruhoe, New Zealand		Andesite	640	96.0	2.6	16.1	–	10.2	6.8	2.5	–	Giggenbach et al. (1986)
Tolbachik, Kamchatka		Basalt	1050	88.2	1.89	2.48	–	0.08	0.14	0.66	–	Menyailov & Nikitina (1980)
Klyuchevskoy, Kamchatka		Calcalkaline basalt	1075	94.8	1.15	2.3	–	0.1	–	1.72	–	Taran et al. (1991)
Poas, Costa Rica		Tholeiitic basalt	940	95.0	0.9	1.35	0.012	2.83	–	0.38	0.015	Rowe et al. (1992)
Momotombo, Nicaragua		Tholeiitic basalt	844	95.1	0.87	2.38	0.025	0.7	0.049	0.35	0.029	Giggenbach (1996)
Nyiragongo, Africa	Rift zone	Melilite-nephelinite	995	44.7	1.4	47.0	2.5	2.2	1.6	–	–	Gerlach (1980)
Erta Ale, Africa		Tholeiitic basalt	1130	77.2	1.39	11.3	0.44	8.34	0.68	0.42	–	Symonds et al. (1994)
Ardoukoba, Africa		Tholeiitic basalt	1070	77.4	1.7	3.7	0.2	14.2	1.7	–	–	Allard (1983)
Surtsey, Iceland		Alkali basalt	1125	92.0	1.65	1.94	0.07	3.27	0.012	–	–	Gerlach (1980)
Kilauea, Hawaii	Ocean island	Tholeiitic basalt	1170	37.1	0.49	48.9	1.51	11.8	0.04	0.08	–	Symonds et al. (1994)

Note: All volcanic gas concentrations are given in mole percent.

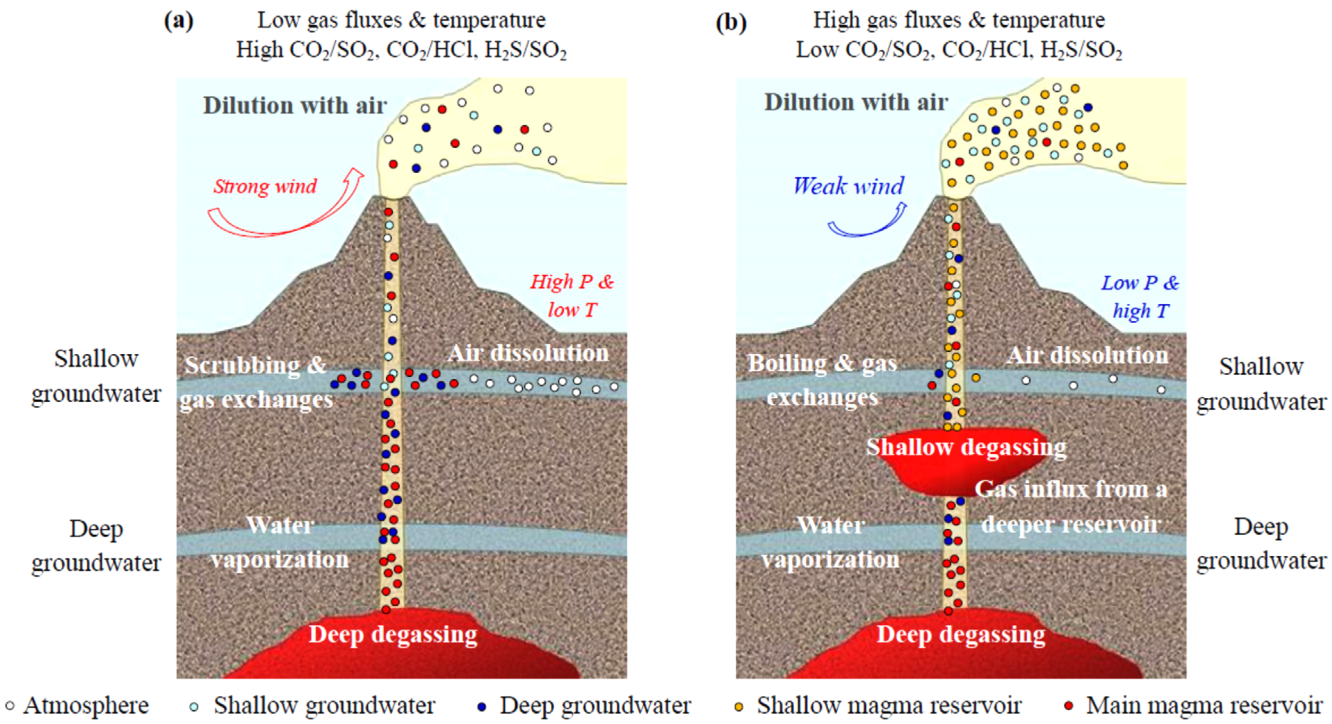

Fig. 3. Evolution of volcanic gases during quiescent periods (a) and strongly passive degassing and eruptive periods (b). In part (a), the deep degassing in the main magma reservoir produces volcanic gases with the composition of CO_2, SO_2, $H_2O >> H_2S$, HCl, HF; the water vaporization of deep groundwater by hot volcanic gases causes the entrainment of water vapors; the scrubbing and gas exchanges of cold volcanic gases at shallow groundwater lead to the intensive dissolution of strong acidic gases (SO_2, HCl, HF); the dissolution of the atmospheric components (O_2 and CO_2) in shallow groundwater is greater at high atmospheric pressure and low temperature; and the dilution of volcanic gas plumes by the air is stronger under windy conditions. In part (b), the volcanic gases from a shallow magma reservoir can be coeluted with those from the main magma reservoir; the shallow degassing in a shallow reservoir produces volcanic gases with the composition of H_2O, SO_2, H_2S, $CO_2 > HCl$, HF; the water boiling and gas exchanges by hot volcanic gases at shallow groundwater cause the entrainment of water vapors and less scrubbing effects; the dissolution of the atmospheric components in shallow groundwater is smaller at low atmospheric pressure and high temperature; and the dilution of volcanic gas plumes by the air is weaker under calm conditions. In the legend, each colored circle represents a specific origin.

volcanic gas fluxes during magma ascents (Bruno et al., 2001; Allard, 2010; Werner et al., 2013). However, such changes do not necessarily occur. If volcanic vents are clogged due to intensive seismic activities, the gas fluxes may decrease. For example, volcanic SO_2 flux decreased from 11 kg/s to 4 kg/s before the eruption of Masaya volcano on April 23, 2001 (Duffell et al., 2003). Also, when multiple magma reservoirs exist beneath a volcano, the gas fluxes may vary depending on which reservoir has a greater impact (see Fig. 3b). If volcanic vents are mainly in the influence of a magma reservoir that is being furnished with a magma with volatile components, the gas discharge will increase (Bruno et al., 2001). On the other hand, if the vents are affected by a magma chamber whose degassing has almost occurred, the emission will decrease (Bruno et al., 2001).

The degassing tendency varies considerably with type of volcanic gases. Under equilibrium degassing conditions, the lower solubility volcanic gases have in magmas, the more readily they are exsolved (Menyailov, 1975). In fact, as a magma ascends, volcanic gases are preferentially evolved in the sequence of CO_2, S, H_2, HCl, and HF (Oskarsson, 1984; Menyailov et al., 1986; Werner et al., 2013). In Figure 4, the compositional changes in the volcanic gases exsolved from a basaltic magma are described as a function of pressure. If the degassing occurs as high as 1,000

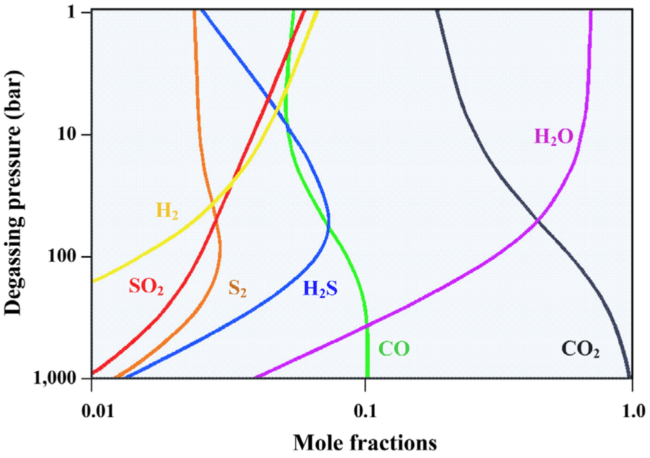

Fig. 4. Simulated volcanic gas compositions (in mole fractions) as a function of degassing pressure. The simulation was performed for a magma with the composition of 0.5 wt% H_2O, 0.3 wt% CO_2, and 0.15 wt% S at 1,300 °C. This figure is modified from Gaillard et al. (2011).

bars, volcanic CO_2 concentration is expected to be 100 times higher than volcanic SO_2 concentration (Edmonds, 2008). As the degassing pressure decreases, volcanic gases are gradually enriched with sulfur species (e.g., $SO_2 + H_2S + S_2$). Notably, SO_2 concentration in volcanic gases continuously increases with the decreasing pressure, whereas H_2S concentration exhibits the maximum at 20–100 bar (Gaillard et al., 2011). Although not included in Figure 4, halogen gases (e.g., HCl and HF) start to be exsolved at the pressure as low as 400 bars (Edmonds, 2008). Consequently, while less soluble gases including CO_2 and sulfuric gases are abundant at the early pre-eruptive period, halogen gases (e.g., HCl and HF) remain mostly in magmas till volcanic eruptions (Aiuppa, 2009).

If the exsolved gases from a magma ascend to the surface without compositional alteration, their composition likely reflects the depth of degassing (López et al., 2013). For example, the concentration ratios of a more insoluble species in magmas to a more soluble one (e.g., CO_2/H_2O, CO_2/SO_2, CO_2/HCl, and SO_2/HCl) are expected to be high if magmatic degassing occurs at a deep depth, whereas these ratios should decrease as a magma ascends. However, these expected patterns are often inconsistent with the gas monitoring data (see section 4.3). There are several possible causes for such inconsistency. Where continued lava fountaining prevails, magmatic degassing may be kinetically limited due to quick ascending magmas (in other words, disequilibrium degassing occurs), causing the measured gas compositions to reflect a deeper degassing signature rather than the present depth (Namiki and Manga, 2008). Also, the degassing of a shallow magma can be accompanied with that of a deeper magma (see Fig. 3b) (López et al., 2013). As discussed below, the gas compositions are also influenced by secondary effects such as hydrothermal interactions and meteorological variability.

3.2. Hydrothermal Interactions

After exsolved from magmas, volcanic gases may interact with hydrothermal systems during their ascents to the ground (Menyailov, 1975; Symonds et al., 2001; Mazot et al., 2008). Figure 3 describes hydrothermal processes leading to changes in volcanic gas compositions. If "hot" volcanic gases come into contact with hydrothermal water, the water becomes vaporized or boiled to be incorporated into volcanic gases (Henley and Ellis, 1983; Federico et al., 2002). Thus, the water vapor in volcanic gases has both magmatic and meteoric origins (Mizutani, 1978; Ohba et al., 2008). The relative contribution of each source can be determined by the isotope analysis of the water vapor; the δD and $\delta^{18}O$ values of the water vapor tend to decrease with the greater extent of water vaporization/boiling (Ohba et al., 2008). To elude the dilution effect of volcanic gases due to water vaporization/boiling, volcanic gas compositions can be expressed on a dry basis (Tassi et al., 2010).

If the gases exsolved at a deep depth ascend all the way to shallow groundwater, the "cooled" gases are likely dissolved (scrubbed) into it (see Fig. 3a). This scenario is true of pre-eruptive period, during which a magma is placed in a deep reservoir. This scrubbing can lower the overall gas fluxes, with the effect varying significantly with type of volcanic gases. While CO_2 and H_2S are less soluble in groundwater, SO_2, HCl, and HF are far more sensitive to the scrubbing (Symonds et al., 2001). Consequently, if the scrubbing is intensive, nearly all of SO_2 and HCl are likely lost from the gas phase. In fact, very low SO_2 fluxes despite vigorous seismic activities were observed before and after the eruption of Mount Spurr in 1992 (Symonds et al., 2001). Thus, unless dry emission paths are developed during the passive degassing period, it is impractical to utilize volcanic SO_2 and HCl fluxes for the prediction of eruptions (Symonds et al., 2001). Compared to CO_2, both He and CH_4 were even less dissolved into groundwater, making them ideal candidates for eruption precursors (Federico et al., 2002). Despite the general characteristics, the scrubbing effect can vary significantly with temperature, pressure, and pH (Henley and Ellis, 1983; Federico et al., 2002). Thus, given the dynamic nature in both magmatic processes and hydrothermal interactions, cares should be taken to assess the scrubbing effect even at a volcano.

Aside from volcanic gases, hydrothermal systems beneath volcanoes also undergo the compositional changes. The dissolution of acidic volcanic gases in groundwater leads to pH decrease and temperature increase, both of which accelerate water-rock interactions such as to increase dissolved salts in groundwater (Henley and Ellis, 1983). For example, Federico et al. (2010) found that the salinity of groundwater was highest close to magma vents. Also, given that shallow groundwater continuously interacts with the atmosphere, the former's composition is influenced by both volcanic gases and the atmosphere (see Fig. 3) (Henley and Ellis, 1983).

3.3. Meteorological Variability

Volcanic gas compositions often take temporal variations in response to seasonal and weather changes (Faber et al., 2003; Kelly et al., 2013). At Izu-Oshima volcano, Shimoike and Notsu (2000) reported seasonal variations in volcanic CO_2 and O_2 concentrations. While volcanic CO_2 at this volcano increased during the summer, it decreased during the winter (Shimoike and Notsu, 2000). On the other hand, the opposite trend was observed for volcanic O_2 (Shimoike and Notsu, 2000). Such patterns were attributed to time-varied interactions among the atmosphere, groundwater, and volcanic gases (Shimoike and

Notsu, 2000). Due to the increasing water solubility of gases at lower temperature, greater amounts of air-driven species in groundwater are available for exchanges with volcanic gases during the winter (see Fig. 3a). Since the ambient atmosphere contains much lower CO_2 but higher O_2 levels than volcanic gases, the increased dissolution of the atmospheric species into groundwater during the winter eventually leads to decreased CO_2 but increased O_2 concentrations in volcanic gases via interactions between groundwater and volcanic gases.

Weather conditions (e.g., the atmospheric temperature and pressure, precipitation, and wind speed) may alter volcanic gas compositions (see Fig. 3). For example, as the atmospheric pressure rose, volcanic CO_2 concentration decreased but volcanic O_2 concentration increased (Shimoike and Notsu, 2000). Also, volcanic H_2 levels were found to increase in a few hours after the atmospheric pressure dropped (Shimoike and Notsu, 2000). At Merapi volcano in Indonesia, Zimmer and Erzinger (2003) found that the radioactivity of ^{222}Rn in volcanic gases increased with the atmospheric pressure. Following heavy rainfalls, they also reported that H_2O content in volcanic gases was elevated along with the decreased fumarolic temperature due to the infiltration of rainwater into the volcano edifice (Zimmer and Erzinger, 2003). Similarly, the fumarolic temperature at Etna volcano was found to drop sharply after rainfalls (Madonia et al., 2013). Wind has also been known to affect volcanic gas compositions (Faber et al., 2003; Kelly et al., 2013). For example, volcanic H_2O content at Galeras volcano changed considerably when a strong wind blew (Faber et al., 2003). Also, volcanic O_3 was found to decrease under strongly windy conditions (Kelly et al., 2013). Importantly, meteorological variability has a pronounced impact on remote gas measurements, which sense volcanic gas plumes, the mixtures of atmospheric and volcanic gases (see Fig. 3). Thus, if remote gas measurements are utilized, it is desirable to remove the atmospheric backgrounds from the monitoring data (Zimmer and Erzinger, 2003).

4. VOLCANIC GASES AS ERUPTION INDICATORS

Several volcanic gases have been utilized to forecast eruptions. The gas components used for this purpose include acidic gases (CO_2, SO_2, H_2S, HCl, and HF), H_2O, rare gases, and metallic trace elements (Sparks, 2003; Johnson and Canil, 2011). Also, the isotopic compositions of certain volcanic gases (e.g., ^{3}He/^{4}He, δ^{13}C of CO_2, and δ^{18}O of H_2O) as well as fumarolic temperature have been measured to aid predict eruptions (Yamashina and Matsushima, 1999). How such gas parameters have been used to evaluate volcanic activities is discussed below.

4.1. Carbon Dioxide

Carbon dioxide (CO_2) has been commonly used for the prediction of eruptions. Due to the low solubility in magmas, CO_2 is readily exsolved to be present at large quantities in volcanic gases (Bruno et al., 2001). Among the major volcanic gases, CO_2 is least affected by the hydrothermal scrubbing (Symonds et al., 2001). When a CO_2-rich magma is newly injected into a magma chamber, or when the degassing of a magma becomes stronger due to its surge, volcanic CO_2 fluxes tend to increase (Carapezza et al., 2004). Thus, volcanic CO_2 fluxes are expected to increase substantially at the imminence of eruptions, making CO_2 an invaluable indicator of eruptions.

At Redoubt volcano, Werner et al. (2013) measured the fluxes of volcanic CO_2 and SO_2 from October 2008 to August 2010 (see Fig. 5). As shown in Figure 5, the CO_2 flux was estimated to

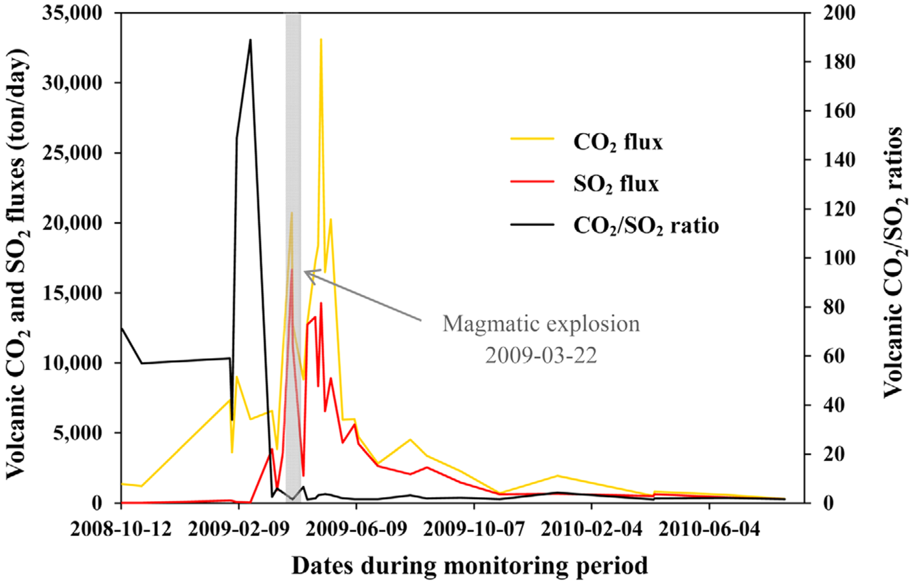

Fig. 5. Discharge fluxes of volcanic CO_2 and SO_2 along with CO_2/SO_2 ratios at Redoubt volcano in Alaska for the years of 2008–2010. This figure is derived from Werner et al. (2013).

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be as low as 1,000 ton/day during the quiescent period, but it increased up to 3,000–9,000 ton/day a month before the phreatic eruption on Mar 15, 2009. Notably, the cumulative CO_2 emission between March 2009 and July 2009, which corresponded to the initiation of the eruption and the subsequent formation of a volcano dome, respectively, accounted for 59% of the total CO_2 emission during the whole monitoring period.

At Etna volcano, Bruno et al. (2001) monitored volcanic CO_2 at two sites between July 1997 and March 1999. At one site, volcanic CO_2 concentration was measured to be 282,000 ppm on September 7, 1998, and then it continuously climbed up to 546,800 ppm about a month before the eruption on February 4, 1999. Right after the eruption, the concentration at this site was recorded as high as 838,400 ppm. Similarly, volcanic CO_2 concentration at the other site increased from 33,300 ppm on January 14, 1999 to 64,100 ppm on February 8, 1999. During the post-eruptive period, CO_2 concentrations at both sites were found to fluctuate a lot due to the ascent of a magma beneath the volcano or the introduction of a new magma into the magma chamber (Bruno et al., 2001). Also, Giammanco et al. (2013) monitored volcanic CO_2 for the eruptions of Etna volcano in 2002–2003 and 2010. As in the case of the eruption in 1999, the CO_2 fluxes at this volcano increased considerably close to the eruptive events. There were other occasions that volcanic CO_2 fluxes increased suddenly on the verge of volcanic eruptions. For example, Jousset et al. (2013) reported sharp increases in volcanic CO_2 and H_2O fluxes right before the eruption of Merapi volcano in 2010. Thus, abruptly increased CO_2 fluxes are generally considered to be indicative of eruptions. Yet, since volcanic CO_2 fluxes do not reach the maxima until eruptions (see Fig. 5), it remains unsolved to determine how close eruptions are.

4.2. Sulfur Gases

Sulfur gases can take various chemical forms including SO_2, H_2S, COS, and elemental sulfur (Ohba et al., 1994). Of all these, SO_2 and H_2S represent the major sulfur species in volcanic gases (Lee et al., 2016). Due to the relative abundance in volcanic gases, SO_2 is regarded as an indicator of eruptions. Since SO_2 exhibits the second lowest solubility in magmas among acidic gases, its discharge rate likely increases as magmatic degassing gets stronger (Oskarsson, 1984). Nonetheless, as discussed earlier, the scrubbing by groundwater can lower volcanic SO_2 concentrations to great extents, sometimes making this species inadequate for the prediction of eruptions (Symonds et al., 2001).

At Redoubt volcano, Werner et al. (2013) observed that both CO_2 and SO_2 fluxes increased substantially during the eruptive period (see Fig. 5). As shown in Figure 5, volcanic SO_2 flux was kept as low as 10 ton/day, but it increased up to 34–177 ton/day a month before the eruption. Indeed, the SO_2 flux between the volcanic eruption and the dome growth period accounted for 66% of the total emission. At Etna volcano, Giammanco et al. (2013) monitored volcanic SO_2 flux (see Fig. 6a). In Figure 6a, the SO_2 flux seemed to increase close to the eruptive events. Yet, the increased SO_2 flux at Etna volcano could be attributed to multiple causes: eruptive events, the ascent of the existing magma, and the injection of a new magma into the magma chamber (Bruno et al., 2001).

At Stromboli volcano, Burton et al. (2009) monitored volcanic SO_2 for the eruptions in 2006 and 2007 (see Fig. 6b). The SO_2 flux ranged between 150 and 200 ton/day before the effusive eruption on February 27, 2007. Then, the SO_2 flux sharply increased at the imminence of this event, with the average flux of 620 ton/day lasting for the next 35 days. During this period, the total SO_2 emission from Stromboli volcano was estimated to be 22,000 ton. At other volcanoes, the increased SO_2 fluxes related to eruptions have also been noticed. At Colima volcano in Mexico, Zobin et al. (2008) reported that volcanic SO_2 flux increased noticeably close to the eruption, which was accompanied with sustained seismic activities. At Kilauea volcano in Hawaii, Chartier et al. (1988) observed the increased SO_2 flux right before the eruption. Similar patterns in volcanic SO_2 flux were also noted at Kilauea volcano (Gerlach and Graeber, 1985) and Etna volcano (Allard et al., 2005). As in case of volcanic CO_2 flux, volcanic SO_2 flux did not to reach the maximum until eruptions (see Figs. 5 and 6), making it difficult to foretell eruptions.

H_2S, another main sulfur species in volcanic gases, can serve as a precursor to eruptions (Werner et al., 2013). As discussed earlier, while the degassing of SO_2 continuously increases with the decreasing depth, the degassing of H_2S initially increases and then decreases (Gaillard et al., 2011) (see Fig. 4). Thus, if a magma ascends sufficiently close to the surface (e.g., as low as ~5 bar), volcanic H_2S flux is expected to be low, making SO_2 the dominant sulfur species (Gaillard et al., 2011). In fact, such features have been noted at field gas measurements (Aiuppa et al., 2005; Gaillard and Scaillet, 2009; Oppenheimer et al., 2011). In contrast, the increased H_2S flux was also observed at the imminence of the eruption of El Hierro submarine volcano (Pérez et al., 2012), thus requiring other accounts for such a discrepancy. After exsolved from magmas, the stability of H_2S and SO_2 in the gas phase is related to each other as follows (Giggenbach, 1987):

$$SO_2 + 3H_2 = H_2S + 2H_2O. \tag{1}$$

According to thermodynamic calculations, Reaction (1) shifts to the right side with the increasing pressure and the decreasing temperature (Giggenbach, 1987; Ohba et al., 1994). Also, as discussed earlier, H_2S is less subject to the scrubbing than SO_2

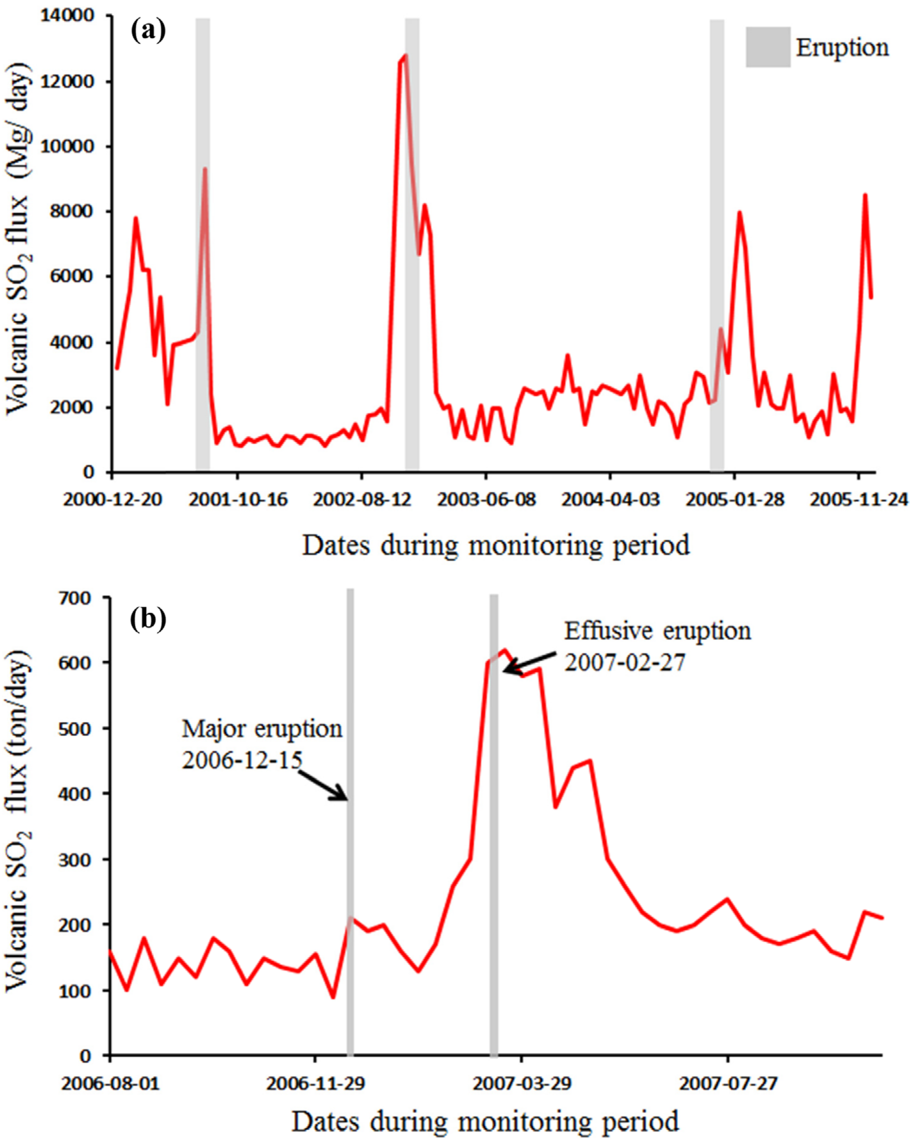

Fig. 6. Volcanic SO_2 fluxes at Etna volcano (a) and Stromboli volcano (b). Parts (a) and (b) are derived from Giammanco et al. (2013) and Burton et al. (2009), respectively.

(Symonds et al., 2001). Taken together, if magmatic degassing occurs at a greater depth (Fig. 3a), H_2S is likely to be the second dominant species after CO_2 as a result of both gas-phase Reaction (1) and preferential scrubbing of SO_2 during volcanic gas rises (Giggenbach, 1980). Thus, high H_2S/SO_2 ratios in volcanic gases may point to the re-equilibrium of volcanic gases at low temperature (Symonds et al., 1994, 2001; Aiuppa et al., 2005). On the other hand, if hot volcanic gases exsolved from a shallow magma reservoir are discharged to the atmosphere without significant scrubbing (Fig. 3b), SO_2 is likely the dominant sulfur gas (Symonds et al., 1994). Consistent with these, an abrupt increase in volcanic SO_2/H_2S ratios was observed on the verge of the eruption of Kusatsu-Shirane volcano in 1976 (Ossaka et al., 1980). Given the potential of both sulfur gases to be eruption precursors, it is necessary to separately quantify them (Montegrossi et al., 2001; Lee et al., 2016).

4.3. Concentration Ratios

Due to the different solubility in magmas, the concentration ratios of relatively insoluble to soluble volcanic gases, which include CO_2/H_2O, CO_2/SO_2, CO_2/HCl, SO_2/HCl, and SO_2/HF, have been commonly engaged to predict eruptions (Duffell et al., 2003; Aiuppa, 2009; Notsu and Mori, 2010; Stremme et al., 2011; López et al., 2013). If exsolved gases are at equilibrium with the remaining magma (i.e., equilibrium degassing) and then ascend to the surface without alterations (e.g., hydrothermal scrubbing), the aforementioned ratios should decrease as volcanic eruptions become imminent (Edmonds, 2008; López et al., 2013). Indeed, the decreasing tendencies during magma ascents were noted for the ratios of CO_2/SO_2 and SO_2/HCl (Aiuppa et al., 2007). However, the contradictory cases have also been documented. As discussed earlier, the discrepancy between thermodynamic presumptions

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and field observations may arise from a number of factors: kinetically limited degassing (disequilibrium degassing), the presence of multiple degassing magmas, hydrothermal interactions, and meteorological variability. Due to the site-specific and time-varied nature, the relative importance of the aforementioned factors should be evaluated on a case-by-case basis. Thus, the knowledge on the configuration of magma vents and hydrothermal systems as well as the number, size, and depth of magma chambers is required to have a comprehensive understanding of volcanic gas chemistry in the context of magmatic degassing and hydrothermal interactions. Unfortunately, no such information is readily available at many volcanoes, often rendering the phenomenological interpretation of the gas monitoring data.

CO_2/SO_2 ratios in volcanic gases is a potential indicator of eruptions. In Figure 5, volcanic CO_2/SO_2 ratios at Redoubt volcano are compared to both CO_2 and SO_2 fluxes. Given that CO_2 is less soluble in magmas than SO_2, volcanic gases are expected to be initially more enriched with the former than the latter (López et al., 2013). Consistently, the CO_2 flux began to rise ahead of the SO_2 flux, such that the CO_2/SO_2 ratios jumped from 50–70 to 190 before the eruption. Even if the CO_2 flux continuously increased through the eruptive period, the CO_2/SO_2 ratios reached the maximum before the eruption due to the more rapidly increasing SO_2 flux. Similar patterns in volcanic CO_2/SO_2 ratios were also observed at Stromboli volcano (see Fig. 1). Therefore, it is more valuable to track CO_2/SO_2 ratios than individual CO_2 and SO_2 fluxes for the prediction of eruptions.

At Kamchatka, Menyailov (1975) found that volcanic S/HCl ratios increased considerably close to eruptions (see Fig. 7). Using volcanic gas data from 28 volcanoes, Aiuppa (2009) noticed a positive correlation between volcanic activities and the ratios of SO_2/HCl and SO_2/HF. In his study, both ratios were shown to increase as volcanic activities became intensive or eruptions were about to occur, whereas the ratios appeared to decrease when volcanic activities became quiescent or eruptions came into an end. Similarly, Duffell et al. (2003) reported that SO_2/HCl ratios increased at the imminence of eruptions. Such trends in the SO_2/HCl and SO_2/HF ratios contrast with the greater solubility of halogen gases (HCl and HF) in magmas than SO_2, which should have led to the decreasing SO_2/HCl and SO_2/HF ratios as volcanic activities became stronger. In fact, volcanic SO_2/HCl ratio at Asama volcano ranged between 6.7 and 9.1 for three to four years before the eruption, but the ratio dropped to 5.0–5.9 on the verge of the eruption (Notsu and Mori, 2010). Even in this case, the lower SO_2/HCl ratio close to the eruptive event was attributed to the scrubbing effect, not the pressure-dependent magmatic degassing. During the quiescent period, the magma of this volcano was placed in a deep reservoir, thus making the dissolution of the exsolved HCl gas into shallow groundwater significant (see Fig. 3a). As the magma ascended near the surface, the loss of HCl to the nearby hydrothermal system became smaller (see Fig. 3b), resulting in lower SO_2/HCl ratios at the imminence of the eruption.

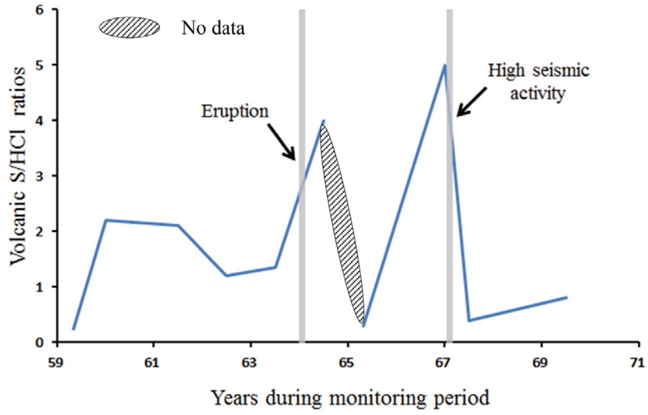

Fig. 7. Volcanic S/HCl ratios at Shiveluch volcano in Kamchatka. This figure is derived from Menyailov (1975).

The CO_2/H_2O ratio in volcanic gases is also considered to be a precursor to eruptions (Ohba et al., 2008, 2010). Given the greater magma solubility of H_2O than CO_2 (Edmonds, 2008), the CO_2/H_2O ratio is expected to decrease as a magma ascends (see Fig. 4). In contrast, this ratio was found to increase when volcanoes were about to erupt (Ohba et al., 2008, 2010). Given that CO_2/H_2O ratio is higher in magmatic gases than in meteoric water, it is expected to increase as volcanic gases are more affected by magmatic degassing than hydrothermal interactions (Holloway and Blank, 1994).

4.4. Metallic Trace Elements

Metallic trace elements in volcanic gases can also be used to predict eruptions since their compositions change as a function of volcanic activities (Quisefit et al., 1988; Toutain et al., 1995). Given that volcanic eruptions are the major inputs of trace metals to the atmosphere, it is of environmental concern to monitor these metals (e.g., Hg, Cd, As, and Cr) in volcanic gases (Johnson and Canil, 2011).

Mambo and Yoshida (1993) conducted a series of lab experiments to examine the geochemical behavior of arsenic (As) as a function of temperature. In their study, gaseous As concentration did not change as long as the temperature was kept above 300 °C. In the meanwhile, this element was readily dissolved in water or reacted with sulfur species to precipitate when the temperature fell below 100 °C. Given that arsenic was rich only in hot volcanic gases, it was proposed as a potential indicator of eruptions (Mambo and Yoshida, 1993). In fact, the similar results were observed at the eruption of Tokachi volcano in Japan (Hirabayashi et al., 1990). Under a range of temperature and pressure, Mackenzie and Canil (2008) examined the volatilization behavior of Cd, Re, Tl, Pb, Sb, and Te from a magmatic melt. Of all these elements, Cd was the most quickly volatilizing metal, with Re being the most sluggishly diffusing one (Mackenzie and Canil, 2008). Due to the significantly different behaviors between Cd and Re, thus, Mackenzie and Canil (2008) have argued that the Cd/Re ratio in volcanic gases is a potential eruption precursor. In agreement, this ratio increased considerably before and after the eruption of Kilauea volcano in 1984 (Crowe et al., 1987).

At Saint Helens volcano, ^{210}Pb concentration in volcanic gases was higher during the eruptive period than the quiescent period, implying that it is useful to monitor volcanic ^{210}Pb to predict volcanic activities (Berlo et al., 2006). Nonetheless, the compositions of metallic trace elements in volcanic gases can also change in response to non-volcanological factors such as the chemical composition of surrounding rocks (Symonds et al., 1992).

4.5. Isotope Compositions

Isotopic compositions of certain volcanic gases can be used to forecast eruptions. The isotope ratio of ^{3}He/^{4}He in volcanic gases was closely related to eruptions (Sano et al., 1988). The primitive isotope of He, ^{3}He, gradually escapes from the Earth due to its lighter mass (Gilfillan et al., 2011). On the other hand, the heavier isotope ^{4}He is continuously generated via radioactive decay of ^{235}U, ^{238}U, and ^{232} Th, and it can be trapped in the gravity of the Earth (Padrón et al., 2012). Consequently, the ratio of ^{3}He/^{4}He is higher in the Earth's mantle than in the crust (Mackintosh and Ballentine, 2012). In this regard, this isotope ratio in volcanic gases should increase as a volcano is about to erupt (Sano et al., 1988; de Moor et al., 2016). In fact, Sano et al. (1991) found that the ^{3}He/^{4}He ratio increased from 4.18 to 4.59 three months before the eruption of Mihara volcano on October 4, 1990. Also, the carbon isotope of CO_2 in volcanic gases or hot spring water is a potential indicator of eruptions (Sano et al., 1995). Due to the isotope fractionation, the δ^{13}C value of CO_2 is lower in volcanic gases than in hydrothermal water (Barry et al., 2013). As volcanic actives get stronger, volcanic gases become less interacting with hydrothermal water, thus resulting in the lower δ^{13}C of volcanic CO_2 on the verge of eruptions (Allard et al., 1991).

Aside from a potential indicator of eruptions, the isotope composition of volcanic gases can be used to track their sources. For example, a condensate of volcanic gases is largely a mixture of magmatic and meteoric waters, both of which have distinct isotope signatures of H_2O (e.g., δD and δ^{18}O) (Kusakabe et al., 1970; Mizutani, 1978). Since meteoric water is less enriched in heavier isotopes (e.g., D and ^{18}O) than magmatic water, lower δD and δ^{18}O values indicate a greater proportion of the former than the latter (Mizutani, 1978). Other isotope compositions including δ^{34}S, ^{4}He/^{20}Ne, ^{38}Ar/^{36}Ar, and ^{84}Kr/^{36}Ar in volcanic gases have been utilized to assess the influence of meteoric water, the injection of a new magma, and the degree of magmatic degassing (Sakai et al., 1982; Sano et al., 1991; Tedesco et al., 1995; Rizzo et al., 2009; Ruzie and Moreira, 2010).

5. CONCLUSIONS

This study discusses the eruptive cycles of volcanoes, the geochemical processes controlling volcanic gas compositions, and the potential of volcanic gas parameters as eruption precursors. In general, the fluxes of volcanic gases (e.g., CO_2, SO_2, H_2S, etc.) as well as gas temperature increase considerably at the imminence of volcanic eruptions. Also, the concentration ratios of CO_2/SO_2, CO_2/HCl, SO_2/HCl, and H_2S/SO_2 tend to decrease (see Fig. 3). However, such trends in volcanic gas compositions can be obscured by kinetically limited degassing as well as secondary effects including hydrothermal interactions and meteorological variability. Consequently, it is often difficult to make a generalization on volcanic gas chemistry for the prediction of eruptions. In such cases, concentration ratios between volcanic gases (e.g., CO_2/H_2O, CO_2/SO_2, SO_2/HCl, and SO_2/HF) may serve better for the prediction of volcanic eruptions than individual gas fluxes. Also, other volcanic gas data such as metallic trace elements and isotope compositions can aid forecast volcanic activities. Furthermore, it is recommended to assess volcanic gas data in conjunction with geophysical and geomorphological data (e.g., seismic observation and electronic distance measurement) for the better prediction of volcanic eruptions.

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