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Origin of volcanic gases discharging from a cooling lava dome of Unzen volcano, Japan

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ABSTRACT

Chemical and isotopic compositions were measured for volcanic gases discharged from the recently erupted Heisei-shinzan dome of Unzen volcano. The volcanic gases are largely mixed with the air, whose content ranges from 60 to 90%. Chemical and isotopic compositions of the gases are corrected for the air mixing accounting not only for the major air components but also for contribution of the atmospheric moisture and CO_2. The variation of the corrected composition revealed that these gases are derived from various sources including; 1) the air circulating through the lava domes, 2) the local meteoric water at various degrees of evaporation and vapor loss, 3) the magmatic CO_2 likely originated from a deep source, and 4) the gases discharged from the cooling lava dome. The gases discharged from the cooling lava dome are characterized by the oxygen and hydrogen isotopic composition typical of the subduction-type volcanoes but with high Cl content and high Cl/S and F/S ratios. In contrast, discharges of CO_2 with the magmatic $\delta^{13}C$ values are not correlated with discharge of heat or acid gas species, suggesting the ascent of CO_2-rich fluids from a deep source, possibly through the volcanic conduit.

1. Introduction

The essential source of volcanic gases is magma. The origin of the chemical and isotopic composition of volcanic gases has been evaluated to understand the link to the source magma and their degassing conditions (e.g., Giggenbach, 1996). The magmatic link is generally more evident in high-temperature volcanic gases, in particular the gases discharged during eruptions. In contrast, low-temperature volcanic gases are commonly affected by the processes in a hydrothermal system located between the degassing magma and the fumaroles. Although the heat and acid species are believed to have been derived essentially from a magma, the specific links to the source magma is often not clear in the low-temperature gases, such as about magma depth, degassing mechanism or the gas composition degassed from the magma. Source of the volcanic gases discharging from recently erupted lava domes are also not clear. There are two likely source magmas; the lava dome itself and a deep magma chamber which fed the recent eruption.

Relatively high-temperature (~400 °C) acid volcanic gases were discharging from the recently erupted Heiisei-shinzan lava dome of Unzen volcano. The source of these acid volcanic gases was interesting not only from the scientific point of view but also from the safety evaluation of the conduit drilling program. If the high-temperature acid gases originate from a deep source and are still ascending through the recent volcanic conduit, which is the target of the drilling, the safety arrangement against the sudden injection of the high-temperature acid gases are necessary. Therefore, we sampled and analyzed chemical and isotopic compositions of the volcanic gases in order to evaluate their origins prior to the conduit drilling, as a part of the Unzen Scientific Drilling Project.

2. Degassing activity of Unzen volcano

Unzen volcano is a composite volcano located on the Shimabara peninsula, Kyushu, Japan (Fig. 1). Recent eruptive activities started with phreatic eruptions on November 17, 1990 after a dormancy of about 200 years. Before the resurgence of the volcanic activity, there was no appreciable degassing activity or thermal anomaly at the summit area. The eruptive activity changed its style to effusive lava extrusion and continued during 1991–1995, with a total volume of erupted magma of 2.1×10^8 m^3. About half of this amount remained as a lava dome on the summit (Nakada et al., 1999). Emission of high-temperature volcanic gases was associated with the lava effusion. The highest measured temperature at the gas vent was 820 °C and gas equilibrium temperature was estimated to range from 850 °C to 911 °C, that is consistent with petrological estimation of magma temperature (Ohba et al., 1994). Variation of the SO_2 discharge rate monitored with COSPEC correlated closely with the lava extrusion rate (Hirabayashi et al., 1995). Comparison of the SO_2 discharge rate, lava effusion rate and S contents in melt inclusions revealed that the amount of high-temperature volcanic gases is almost the same as that

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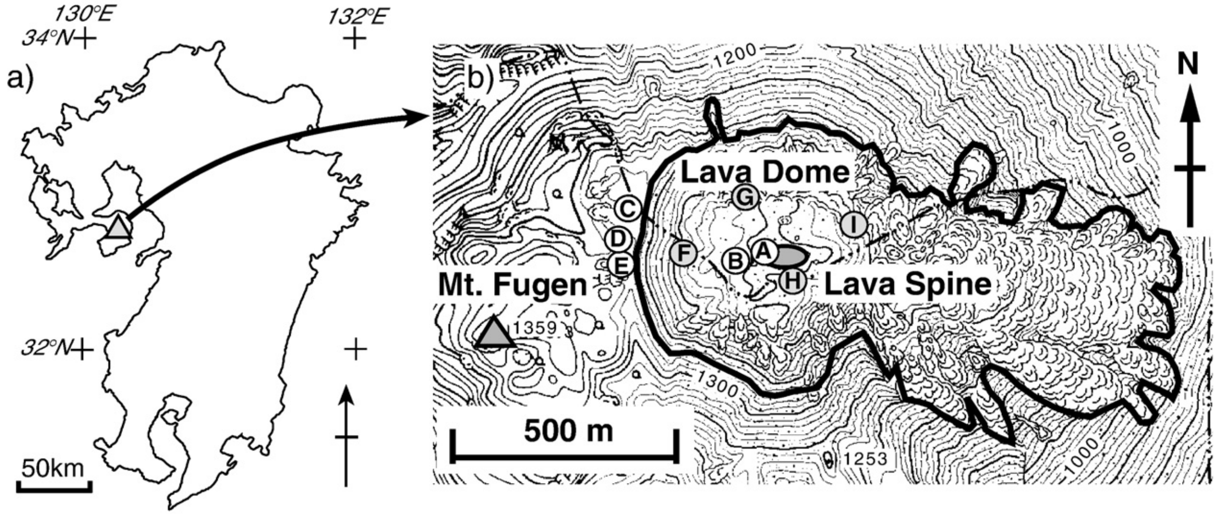

Fig. 1. Location of Unzen volcano in Kyushu, Japan (a) and distribution of fumaroles at the summit area of the volcano (b). The fumaroles where gas samples were collected were shown by open circles, whereas the shadowed circles are other major fumarolic features which were not sampled.

transported by the erupted magma as dissolved volatiles, indicating a simple degassing of the ascending magma to the summit without gas-loss to the volcanic edifice or excess volatile sources (Satoh et al., 2003).

Lava discharge ceased after the formation of a lava spine at the top in 1995. The volcanic gas emission also diminished at the end of the eruption, whereas weak gas emission still continues from several localized sites (Fig. 1). Fumaroles at the foot of the spine were the most active, discharging acidic gases, and the temperature of these vents gradually decreased from >600 °C in 1996, to <400 °C in 2001 and 326 °C in 2003, November (Institute of Seismology and Volcanology, 2004). Temperature of other fumaroles quickly decreased to less than 100 °C by 2001. Fumarolic gas samples were collected from the high-temperature fumarole and from several accessible lower temperature fumaroles, in 1999, 2000 and 2001. Steam discharge rates were also estimated at several accessible sites in 2000 (Fig. 1).

3. Methods

3.1. Volcanic gas sampling

The volcanic gases emitted from the Heisei-shinzan lava dome of Unzen volcano were highly mixed with the air, and could not be sampled with the evacuated bottles containing alkaline solution (e.g., Giggenbach, 1976). Therefore we conducted following series of the sampling procedure; 1) Cold-trap, 2) Alkaline-Trap, 3) Evacuated bottle, and 4) KI–KIO_3 trap. Acid condensates were sampled by two cold traps in a series; the first was cooled with ice–water and the second was cooled with dry ice–ethanol mixture. The sample was sucked by a battery-driven pump with a rate of 1–3 l/min and the amount of the pumped air was measured with a gas flow meter installed after the traps. All of the collected acid condensate was brought back to the laboratory and weighed to obtain the water/air ratio. The oxygen and hydrogen isotopic composition of water was measured with the acid condensate samples. The acid condensates were collected only with ice–water cooled traps at fumarole B in the first year, and collected only with dry ice–ethanol cooled trap at fumarole A. However, cooling only with ice–water was found to be insufficient for complete water condensation for isotope analyses (see below for the results), and it was also found that the gas flow tended to be choked by the trapped ice in the dry ice–ethanol cooled trap. Therefore, a train of the ice–water cooled trap and the dry ice–ethanol trap was applied for the later survey.

Alkaline condensates were collected by bubbling the gases using ball filters through two traps containing 10 ml of 1 N NaOH solution, and measuring the amount of air with the gas flow meter. The alkaline solutions were oxidized with H_2O_2 and analyzed for F, Cl and SO_4 with Ion-Chromatography. The volcanic gas was also directly sampled into an evacuated gas bottles and compositions of the gases were analyzed with two Gas-Chromatographs; one with a 5-m-long molecular sieve 5A column for the analyses of H_2, O_2, N_2, CH_4, CO, and the other with a 2-m-long Porapack Q column for the analyses of CO_2, both with Ar carrier gas and TCD. Carbon dioxide in the gas was separated in a vacuum system by cold traps and measured for its carbon isotopic composition with a mass spectrometer. The SO_2 and H_2S in volcanic gases were trapped separately by the use of KI–KIO_3 mixed solution under acid condition and analyzed to obtain the SO_2/H_2S ratio (Ozawa, 1968).

3.2. Steam discharge rate estimation

Steam discharge rates from each accessible fumarole (Fig. 1) were estimated by the method of Kazahaya et al. (1994). This method estimates the steam flux from volume flux of a white plume, which is assumed as water-saturated air. The difference between the water vapor pressure in the plume and in the ambient air is due to mixing of the volcanic gases. Therefore, we can calculate the volcanic steam discharge rate by the measurement of plume volume flux, and ambient temperature and humidity. Volume flux of the plume is estimated by video image processing with an assumption of circular cross-section of the plume (Kazahaya et al., 1994). This method will give a minimum estimation of the discharge rate as it neglects water contained as droplets in the white plume. Furthermore, we could not measure one of the major fumaroles located at eastern rim of the dome (Fumarole I on Fig. 1). Based on the observation from distance, its plume size was apparently similar or even larger compared to those from fumaroles A, G or H, which have relatively large fluxes.

4. Results

4.1. Chemical composition

Chemical compositions of the fumarolic gases were calculated by combining the results from the cold trap (H_2O/Air ratio), the alkaline trap (ratios among Cl, F, total S and Air), the evacuated bottles (CO_2/Air) and KIO_3–KI trap (SO_2/H_2S ratio). All the fumarolic gases