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PATHWAYS FOR ESCAPE OF MAGMATIC CARBON DIOXIDE TO SOIL AIR AT UNZEN VOLCANO, SW JAPAN

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ABSTRACT. Estimation of the magmatic contribution to soil air at Unzen Volcano, SW Japan, was carried out using carbon isotopes, both ^{14}C and ^{13}C, and a mixing model of isotopic mass balance in order to assess the spatial variation of magmatic influence from the volcano. The advantage of using soil air samples is that a wide range of gas sampling sites can be selected. Magmatic CO_2 contributed mostly in the eastern region from Unzen Volcano. The high magmatic contribution to soil air appeared along the Akamatsudani fault zone located southeast of the volcano. Our observations across the fault also showed remarkable peaks of CO_2 concentration and $\delta^{13}C$ values, suggesting that magmatic fluid comes up along the fracture zone as for the normal fault system of the graben.

INTRODUCTION

Unzen Volcano is on the western part of Beppu-Shimabara Graben (20 km NS wide and 200 km EW long) in Kyushu Island, SW Japan, and is located at the center of a smaller-scale graben (Unzen Graben) in Shimabara Peninsula. During the latest activity (1990–1995), a lava dome eruption occurred and made the new highest peak (1486 m in elevation). Ohsawa et al. (2002) discussed a magmatic carbon contribution using $\delta^{13}C$ of dissolved inorganic carbon (DIC) in groundwater and estimated a flux of magmatic CO_2 to be 9.1 t/day to the shallow groundwater system in Shimabara Peninsula. However, their study area was limited to the eastern coastal zone of the peninsula, since it was difficult to find a well or springs where samples outside the coastal zone could be taken. We used soil air samples (air occupying pore space among soil grains) instead of groundwater samples in the present study. Hernández (2001) reported on an advective CO_2 degassing from a volcanic body using a soil air survey. Use of soil air allows us to measure CO_2 concentration and flux easily in the field, and to select a large number of observation points.

In this study, we focused on estimating the spatial variation of magmatic CO_2 around Unzen Volcano using soil air samples for comparison with results from a hydrological survey. We also made supplementary observations across the fault to determine the possible path of magmatic carbon to the ground surface.

STUDY SITE AND METHOD

Observation and sampling of soil air at a soil depth of 30 cm were carried out in January 2002 at 51 observation points around Unzen Volcano (Figure 1). In order to discuss the path of magmatic CO_2 to the ground surface, an observation traverse across the fault was also established. The strike of our traverse was in a North-South direction, since the strike of the Akamatsudani fault, located southeast of the volcano, was West-East. Soil gas was sampled using pre-evacuated bottles (150 mL) or aluminized plastic bags (3 L), and the CO_2 was purified cryogenically on a vacuum line within a few days after sampling. At each observation point, CO_2 concentration in the soil air and CO_2 flux from the soil surface were measured by a portable gas detector (GasTec) and a portable CO_2 flux meter (West System), respectively. The CO_2 flux meter was used in association with a dynamic closed-chamber method with NDIR (LI-800, Licor). The $\delta^{13}C$ values of 33 samples were measured by IR-

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MS with a dual inlet system (Delta Plus, Thermoquest) at the Geological Survey of Japan. The $\delta^{14}C$ values of 21 samples were measured by accelerator mass spectrometry (AMS) (model 4130-AMS, HVEE) at the Center for Chronological Research, Nagoya University.

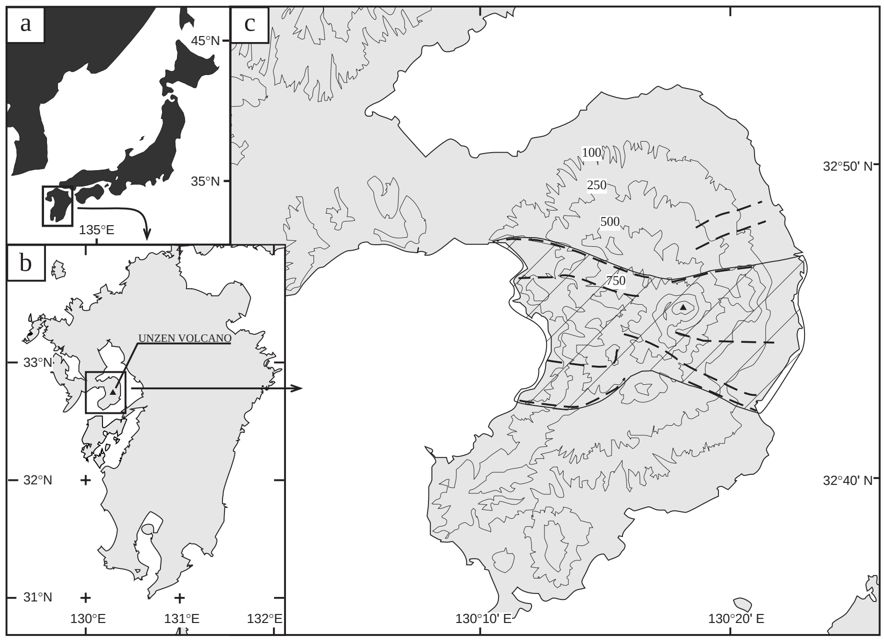

Figure 1 Locality of Unzen Volcano, Kyushu Island, SW Japan. The area of Unzen Graben and the faults are indicated by the striped pattern and dotted lines, respectively (1c, after Hoshizumi et al. 2002).

A mixing model of isotopic mass balance was used to distinguish the magmatic source (m), soil organic matter (s), and atmospheric (a) components in order to estimate the magmatic CO_2 contribution to soil air CO_2. The respective contributions can be computed by the following equations,

$$\delta^{14}C_i = \delta^{14}C_m \cdot F_m + \delta^{14}C_s \cdot F_s + \delta^{14}C_a \cdot F_a \quad (1),$$

$$\delta^{13}C_i = \delta^{13}C_m \cdot F_m + \delta^{13}C_s \cdot F_s + \delta^{13}C_a \cdot F_a \quad (2),$$

$$F_m + F_s + F_a = 1 \quad (3),$$

where F is the contribution of respective components, and $0 \leq F \leq 1$. The subscript i indicates a measured sample. The $\delta^{14}C$ value of the magmatic CO_2 is defined as –1000‰ theoretically, since magmatic carbon was supplied from subducted slab or mantle. The $\delta^{13}C$ value of magmatic CO_2 is –5.3‰ (Takahashi et al., forthcoming), with an error of ±3‰ at 1 σ, which is slightly larger than the $\delta^{13}C$ range of the upper-mantle carbon (Sano and Marty 1995). We assumed the average and error (1 σ) values of carbon isotopic ratios of atmosphere and soil organic matter in the study area because their exact values were not measured. The $\delta^{14}C$ and $\delta^{13}C$ values of atmospheric CO_2 were computed from the observation data of atmospheric CO_2 and their secular trends (Levin and Kromer 1997). Levin and Kromer (1997) reported an exponential trend of $\Delta^{14}C = 417 \cdot \exp(-t/16.0)$; t = years after 1974 and a linear decreasing trend of 0.017‰·yr^{-1} in $\delta^{13}C$. These fitting trends show us the secular decrements of annual mean values from 1996 to 2002. Then, we combined the comparable values from January 1996 (Levin and Kromer 1997) and the secular decrements. The $\delta^{14}C$ and $\delta^{13}C$ values of atmospheric CO_2 obtained in the present study were 106‰ and –8.9‰, respectively. We estimated the error ranges widely to be ±7.5‰ and ±1‰, respectively, since these values were measured in Germany, not in Japan. Comparable values for soil organic matter were 66 ± 7.5‰ and –27 ± 5‰, respectively, since the $\Delta^{14}C$ of vegetation seems to establish equilibrium with the atmosphere, and the $\delta^{13}C$ value is used as the approximate mean value of C3 plant, ranging from –32‰ to –22‰ (Smith and Epstein 1971; Wickman 1952). ^{14}C-dead carbon is not expected from the magmatic contribution because limestone is not present in the Unzen area. Although we have assumed large error ranges in some components, our estimations using Equations 1–3 have a small standard deviation, i.e., better than ±0.02 at 1 σ.

RESULTS AND DISCUSSION

The spatial results of CO_2 concentration, $\delta^{14}C$, $\delta^{13}C$, and flux are shown in Figure 2. The CO_2 concentrations inside Unzen Graben were relatively higher than those outside the graben. The carbon isotopic compositions suggest that the magmatic CO_2 contribution is greater at some of the sample sites inside the graben, shown by relatively lower $\delta^{14}C$ and higher $\delta^{13}C$ values, than at other sample sites outside the graben. The CO_2 flux, however, was higher along the coastal area than inside the graben. The higher CO_2 flux in the coastal area is probably caused by the difference in soil organic matter content between the 2 regions. The outer rim of the graben is a mountain area (Figure 1c) which was mainly covered with pyroclastic deposits (Hoshizumi et al. 2002). Greater amounts of organic matter are formed along the coastal area as compared to inside the graben (mountain area), indicating that there is a greater biological contribution of CO_2 flux along the coastal area. Carbon isotopic values also indicate a greater contribution of biogenic CO_2 in the coastal region.

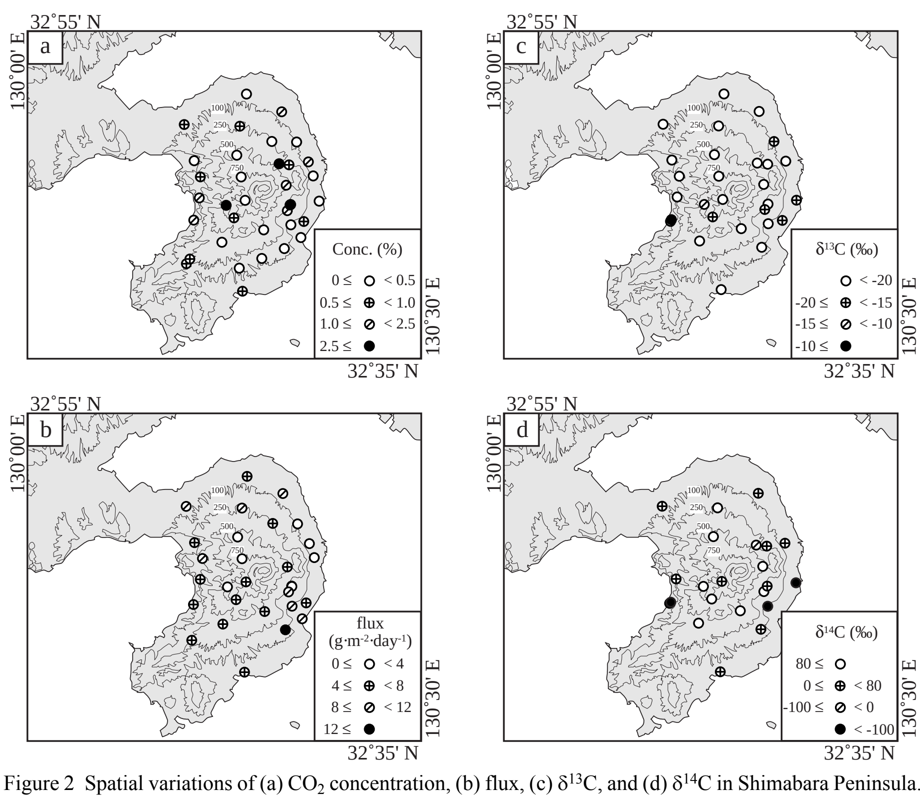

Figure 2 Spatial variations of (a) CO_2 concentration, (b) flux, (c) $\delta^{13}C$, and (d) $\delta^{14}C$ in Shimabara Peninsula.

The spatial variation of the magmatic CO_2 contribution to soil air (Figure 3a) was estimated using Equations 1–3. Using the magmatic contribution and the observed results, magmatic CO_2 concentration in soil air and flux from soil surface can be computed (Figure 3b, c). The highest values of magmatic CO_2 contribution, concentration, and flux were 30%, 0.2%, and 1.7 g $CO_2 \cdot m^{-2} \cdot day^{-1}$, respectively. The magmatic CO_2 is not degassing uniformly across the soil surface, suggesting that degassing from a volcanic body, measured as the soil CO_2 flux, occurred through a specific path to the ground surface. The observed points show that the high magmatic CO_2 flux are located near the fault zones (Figures 1c, 3c). We measured the remarkable peaks of CO_2 concentration and $\delta^{13}C$ in traverse observations across the Akamatsudani fault (Figure 4). The field observation did not allow us to identify whether they were located on the fault or not, since there is no outcrop around this traverse point to confirm the exact location of the fault. However, these peaks suggest that magmatic fluid comes up along the fracture zone, just as in a fault system. It is noteworthy that the spatial variability of CO_2 shows a high magmatic CO_2 contribution, flux, and concentration along the Akamatsudani fault zone located southeast of the volcano.

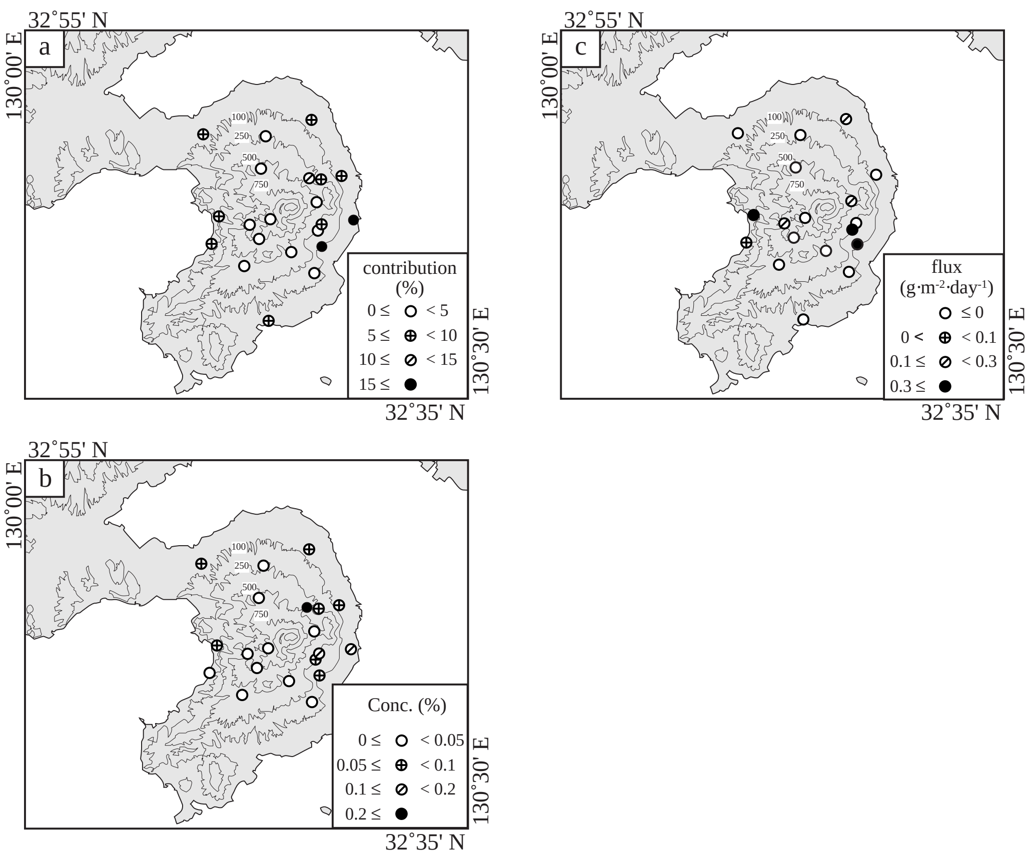

Figure 3 Spatial variations of magmatic CO_2 (a) contribution, (b) concentration, and (c) flux in Shimabara Peninsula.

Even inside the graben, the eastern region showed the relatively high contribution of magmatic CO_2. This agrees well with the groundwater survey (Ohwasa et al. 2002) which suggested that the magmatic fluid in soil air and groundwater might be supplied by the same or a similar system to that of Unzen Graben. Moreover, the approach using soil gas can be used as a tool to substitute for a

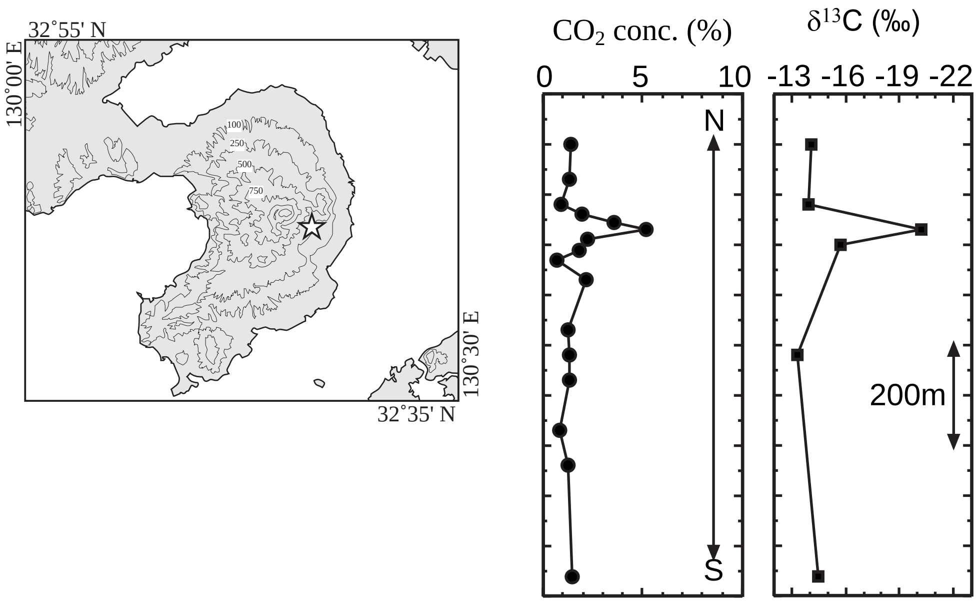

Figure 4 Results of CO_2 concentration and $\delta^{13}C$ along the traverse across the Akamatsudani fault located southeast of Unzen Volcano. The star indicates the observation locality.

groundwater survey to identify magmatic influence. However, we consider that the magmatic contribution may be underestimated because the incorporation of ^{14}C enrichment from nuclear testing is not considered in the present study.

CONCLUSION

The spatial variations of magmatic CO_2 contribution, concentration, and flux were obtained using CO_2 concentration, flux, and carbon isotopic compositions in soil air around Unzen Volcano, SW Japan. The magmatic influence was large inside the Unzen Graben, especially in the eastern region. The spatial variations from soil gas survey were identical to those from the groundwater survey. On the other hand, the traverse across the fault zone suggested that the fracture zone of the fault system played a key role in providing a path for a magmatic fluid from a deep environment to the ground surface. Our study shows that the soil gas survey can provide a convenient tool for the identification of magmatic influences.

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