

Soil gas emission of volcanic CO₂ at Satsuma-Iwojima volcano, Japan

Y. Shimoike¹, K. Kazahaya², and H. Shinohara²

¹Research Institute of Materials and Resources, Akita University, 1-1 Tegata Gakuen-cho, Akita 010-8502, Japan

²Geological Survey of Japan, AIST, 1-1-1 Higashi, Tsukuba 305-8567, Japan

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Soil gas surveys were carried out in November 1999 and October 2000 at Satsuma-Iwojima volcano, southwest Japan. The chemical composition of the soil gas was a mixture of CO₂ and air components, with CO₂ concentrations ranging from 0.03 to 59 vol%. The origin of the soil CO₂ was evaluated on the basis of the variation of $\delta^{13}\text{C}_{\text{CO}_2}$ and CO₂ concentration. Although most of the CO₂ is of biogenic origin, large volcanic contributions of greater than 50% were found close to the caldera rim, where soil temperature anomalies were observed. Emission rate of volcanic CO₂ was estimated by assuming simple mixing among volcanic, atmospheric, and biogenic CO₂. High rates of diffuse emission of volcanic CO₂ were also observed at the caldera rim and close to some hot springs, indicating that part of the volcanic-gas discharge ascends through fissures along the caldera rim. The flux of the volcanic CO₂ through the soil is estimated to be 20 t day⁻¹; this is 25% of the total CO₂ flux through soil and 20% of the CO₂ flux from volcanic fumaroles in the main crater of Iwodake.

1. Introduction

The primary emission of volcanic gases released from magma is through fumaroles found in the summit regions of volcanic centers. Additionally, the more soluble magmatic gases such as SO₂ and HCl can dissolve into surface groundwaters and discharge as hot springs. Recent geochemical studies have shown that magmatic gas is also emitted through the soil around volcanoes even during quiescent periods between volcanic activity (Chiodini *et al.*, 1998; Hernandez *et al.*, 1998). Carbon dioxide is the major constituent of the non-condensable component of the magmatic gas emitted from soils (Giammanco *et al.*, 1995), and the amount of CO₂ released by diffuse soil gas emission is comparable to that of fumaroles at some volcanoes (Baubron *et al.*, 1990; Allard *et al.*, 1991). Variation of the diffuse output of CO₂ correlates with changes in volcanic activity in some studied areas, indicating that soil gas surveys can be a useful tool to monitor volcanic activity (Gerlach *et al.*, 1998; Hernandez *et al.*, 2001a). Such CO₂ flux measurements can be carried out at safe distances from active craters, an advantage over fumarole sampling. The measurement of soil gas emission has also been used to detect active geologic structures, because these features are permeable zones and can channel deep gases towards the surface (Sugisaki *et al.*, 1983; Giammanco *et al.*, 1998).

In this study, we conducted a soil CO₂ flux survey on Satsuma-Iwojima volcano, one of the most active degassing volcanoes in Japan (Shinohara *et al.*, 1993). The objective is to define the areal distribution of the CO₂ diffuse degassing, and relate any patterns to geological features. We also analyzed the chemical composition and $\delta^{13}\text{C}_{\text{CO}_2}$ of the soil

gases at each sample point in order to evaluate the contribution of volcanic CO₂ to the soil CO₂ flux. These data allow us to estimate the total amount of diffuse CO₂ flux with volcanic origin and compare it with CO₂ flux from the fumaroles.

2. Geological Setting

Satsuma-Iwojima is a volcanic island located about 40 km south of Kyushu, Japan (Fig. 1). The caldera-forming eruptions occurred 6300 years ago, followed by the eruption of the Iwodake rhyolitic dome (704 m elevation), and the basaltic cone of Inamuradake (236 m). Yahazudake (349 m) is part of the rim of the Kikai caldera. Although the major eruptive activity of Iwodake ceased 1200 years ago (Ono *et al.*, 1982), strong fumarolic activity continued even after the eruption. Many fumaroles with outlet temperature up to 900°C exist inside and around the summit crater, emitting greater than 300 t day⁻¹ of SO₂ and 100 t day⁻¹ of CO₂ (Shinohara *et al.*, 1993, 2002; Kazahaya *et al.*, 2002). Joghara, a lava plateau with an elevation of about 80 m above sea level, lies outside the caldera and forms the western part of the island. Acidic hot springs discharge along the eastern half of the island from Heikenojo eastward to Ketsunohama, and around to the south shore westward as far as Higashi (Fig. 1). White to green discoloration of seawater occurs near the acid hot springs because of precipitates formed by mixing of the Al- and Fe- rich acid water with seawater (Nogami *et al.*, 1993). In contrast, red discolored seawater is present in Nagahama harbor and Akayu, due to the outflow of neutral pH, iron-bicarbonate water, which is related to the basaltic cone of Inamuradake (Kamada, 1964).

3. Sampling and Analytical Methods

The soil gas survey was conducted during 18–22 November 1999 and on 21 October 2000 at Satsuma-Iwojima vol-

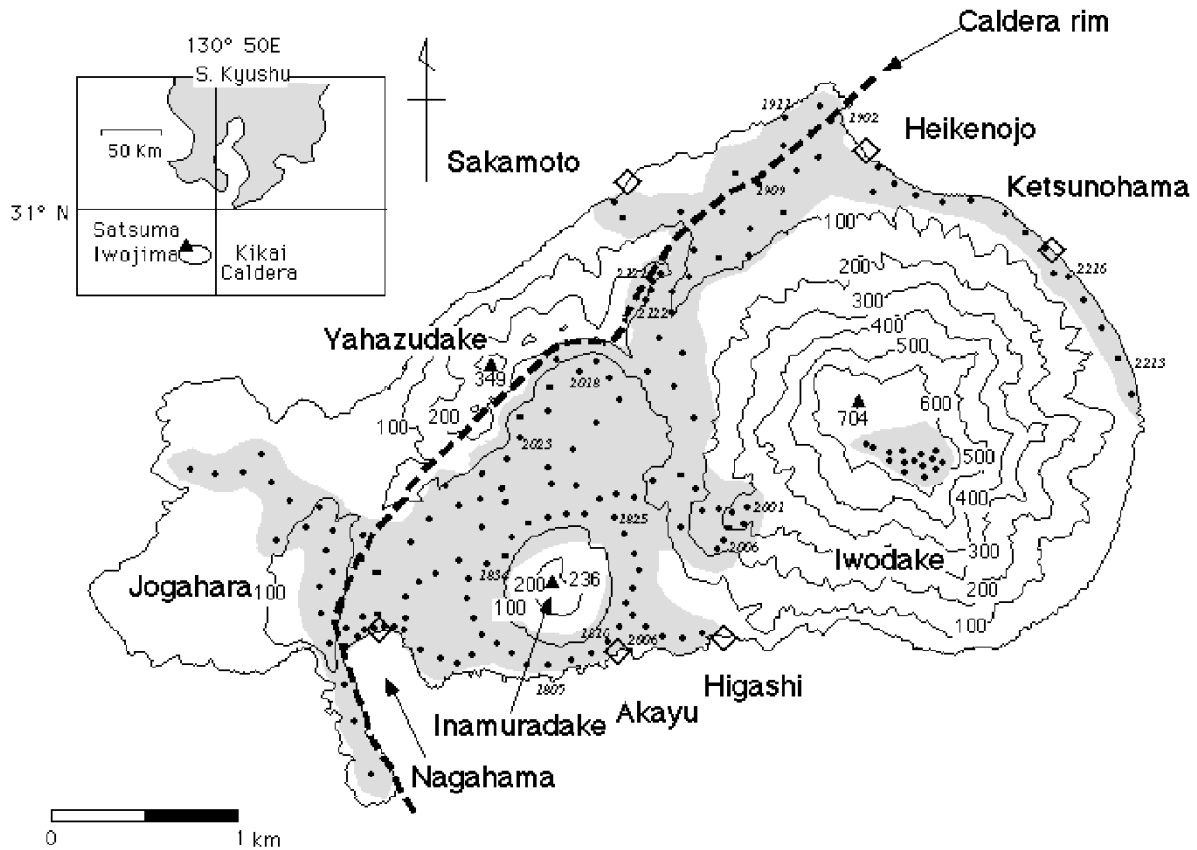


Fig. 1. Geographic map of Satsuma-Iwojima showing sampling locations. (●: measuring point, ○: hotspring, ▲: peak). Dashed line shows the rim of Kikai caldera. The contours are at 100 m intervals. The gray area indicates the survey area. The numbers in italics indicate sample points listed in Table 1.

cano. The 1999 survey covered the whole island except for the Iwodake rhyolitic lava dome, Inamuradake and Yahazudake (Fig. 1). Most of these areas are inaccessible because of steep topography. The survey covered an area of 2.5 km² with 155 sampling points, and the sampling points were selected on the basis of structural features and accessibility, mainly following pathways and roads. Although a road on the southwest flank of Iwodake reaches the summit area, the surveyed area was limited to the lower part of the dome, due to the wide distribution of lava flows on the upper half of the dome. We measured CO₂ flux and soil temperature, and collected gas samples for analysis of the chemical composition of the soil gas and carbon isotope ratio of CO₂. In October 2000, we measured CO₂ flux and soil temperature around the summit area of Iwodake volcano, but soil gas samples were not collected on this occasion.

Carbon dioxide flux was measured using an open-bottomed accumulation chamber (Chiodini *et al.*, 1996). This method has been used in agricultural sciences to determine soil respiration (Parkinson, 1981). For the flux measurement, we selected a flat area to avoid leakage between the ground and bottom of the chamber. Sample locations were in the shade to avoid heating the chamber by sunlight during measurement. The air inside the chamber is mixed by a fan, and introduced into a NDIR Riken Keiki infrared gas analyzer by a pump, and then back to the chamber. A trap filled with Mg(ClO₄)₂ is placed between the chamber and the analyzer to avoid interference from water vapor. In-

crease of CO₂ concentration with time due to the flux from the soil is recorded for several minutes in the data logger. The CO₂ flux (F_{CO_2}) is calculated using the initial rate of increase of CO₂ concentration in the chamber (dC_{CO_2}/dt) and the chamber volume (V), based on the following equation (Chiodini *et al.*, 1996):

$$F_{CO_2} = dC_{CO_2}/dt \times V. \quad (1)$$

Relative error of the calculated flux was estimated to be 10% based on repeated measurements with a controlled CO₂ flux in the laboratory. The detection limit of the system was 0.01 g m⁻² day⁻¹.

Soil gas samples were collected with a syringe from a stainless steel probe inserted into the soil to the depth of 40–50 cm. The samples were analyzed in the laboratory using a gas chromatograph with a thermal conductivity detector. Carbon isotope ratios of CO₂ were measured by mass spectrometry (Finigan MAT delta S). The soil temperature was measured in situ by a thermocouple at 20 cm depth.

4. Results

4.1 Soil gas composition

The chemical composition of the soil gases indicates that all the samples were basically a mixture of CO₂ and air (Table 1). The CO₂ concentration ranges from 0.03 to 59 vol%. High CO₂ concentrations of greater than 10% were found around the Yahazudake area (a in Fig. 2), correlating with the location of the Kikai caldera rim. High CO₂ concentra-

Table 1. Representative composition of the soil gases and high-temperature fumarolic gas at Satsuma-Iwojima volcano.

Sample No.	Date	Temperature (°C)	He (ppm)	H ₂ (ppm)	Ar (%)	N ₂ (%)	O ₂ (%)	CO ₂ (ppm)	$\delta^{13}\text{C}$ (CO ₂) (‰)	Biogenic [#] (%)	Magmatic [#] (%)	Atmospheric [#] (%)	
Soil gas													
1807	1999.11.18	18.5	5.3	10.1	0.91	77.8	20.6	6,829	-23.1	81.6	13.2	5.23	
1810	1999.11.18	19.6	5.1	11.9	0.92	76.8	19.8	9,024	-23.8	84.9	11.1	3.95	
1825	1999.11.18	18.0	4.8	17.5	0.92	77.9	19.6	3,493	-21.5	73.6	16.1	10.2	
1834	1999.11.18	18.2	5.0	9.3	0.93	79.3	20.3	4,570	-22.3	77.6	14.6	7.84	
1902	1999.11.19	18.6	5.1	4.4	0.92	78.3	18.2	47,158	-26.8	99.0	0.3	0.73	
1909	1999.11.19	16.1	5.6	<2	0.92	75.2	19.8	9,873	-24.2	86.8	9.6	3.61	
1911	1999.11.19	21.2	5.1	<2	0.79	69.7	14.3	106,000	-6.1	5.0	94.7	0.30	
2001	1999.11.20	14.1	5.4	6.6	0.95	78.1	20.6	562	-13.8	31.3	4.7	64.0	
2006	1999.11.20	17.5	5.1	<2	0.97	76.3	19.7	6,062	-12.7	34.2	59.9	13.0	
2018	1999.11.20	19.0	4.6	5.4	0.98	78.7	20.6	3,406	-22.8	79.5	10.0	10.5	
2023	1999.11.20	17.1	5.0	555	0.96	72.5	18.9	5,640	-25.4	91.9	1.8	6.35	
2121	1999.11.21	78.6	7.0	18.5	0.42	39.0	1.5	593,753	-7.6	11.8	88.2	0.02	
2122	1999.11.21	81.3	5.5	4.3	0.62	50.0	3.6	454,181	-7.8	12.7	87.2	0.04	
2210	1999.11.22	23.9	4.9	<2	0.95	76.8	20.6	996	-6.2	0.5	63.4	36.1	
2213	1999.11.22	24.1	4.8	6.2	0.95	75.2	20.5	1,674	-11.8	28.0	50.5	21.5	
Fumarolic gas													
		Temperature (°C)	$\mu\text{mol/mol}$					$\delta^{13}\text{C}$ (CO ₂) (‰)					
			He	H ₂	Ar	N ₂	O ₂	CO ₂	H ₂ O				
F1*	1999.11.21	853	n.d.	4,468	0.8	65	n.d.	3,818	974,981	n.a.			
F2*	1999.11.21	853	n.d.	4,133	1.0	64	n.d.	3,679	976,207	n.a.			
F3*	1998.3.18	860	n.d.	4,638	0.9	83	n.d.	3,992	974,149	-4.9			
F4*	1998.3.18	860	n.d.	4,636	1.1	76	n.d.	3,995	974,432	-4.7			

*: from Shinohara *et al.* (2002).

#: Contribution of each source in the soil gases.

n.a.: not analyzed.

n.d.: not detected.

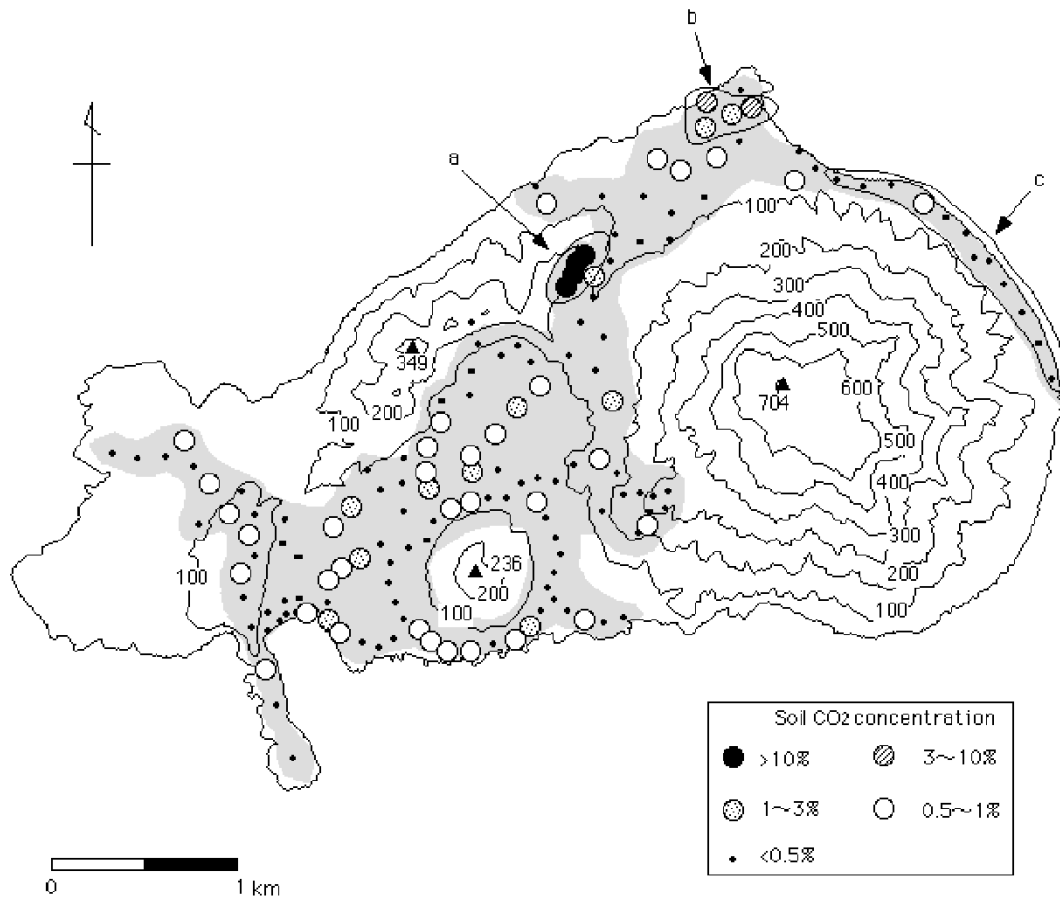


Fig. 2. Distribution of CO₂ concentration in soil gases. The areas indicated with letters are anomalous (see text).

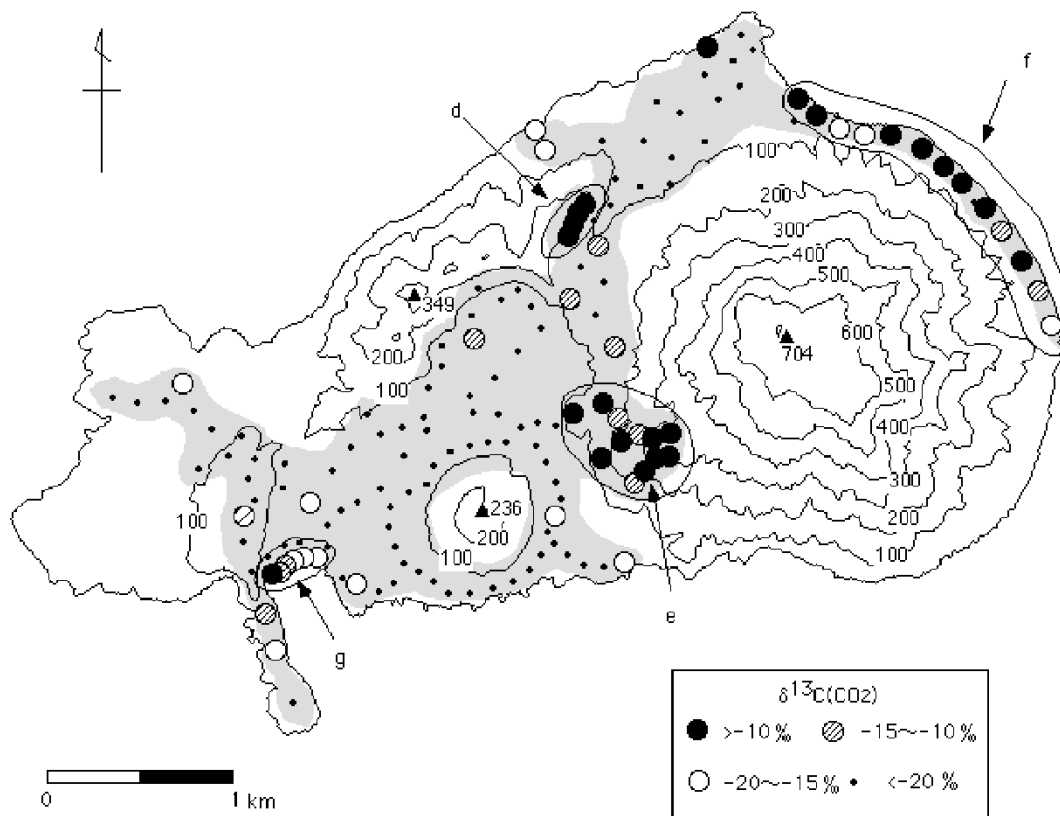


Fig. 3. Distribution of $\delta^{13}\text{C}_{\text{CO}_2}$ values in soil gases. The areas indicated with letters are anomalous (see text).

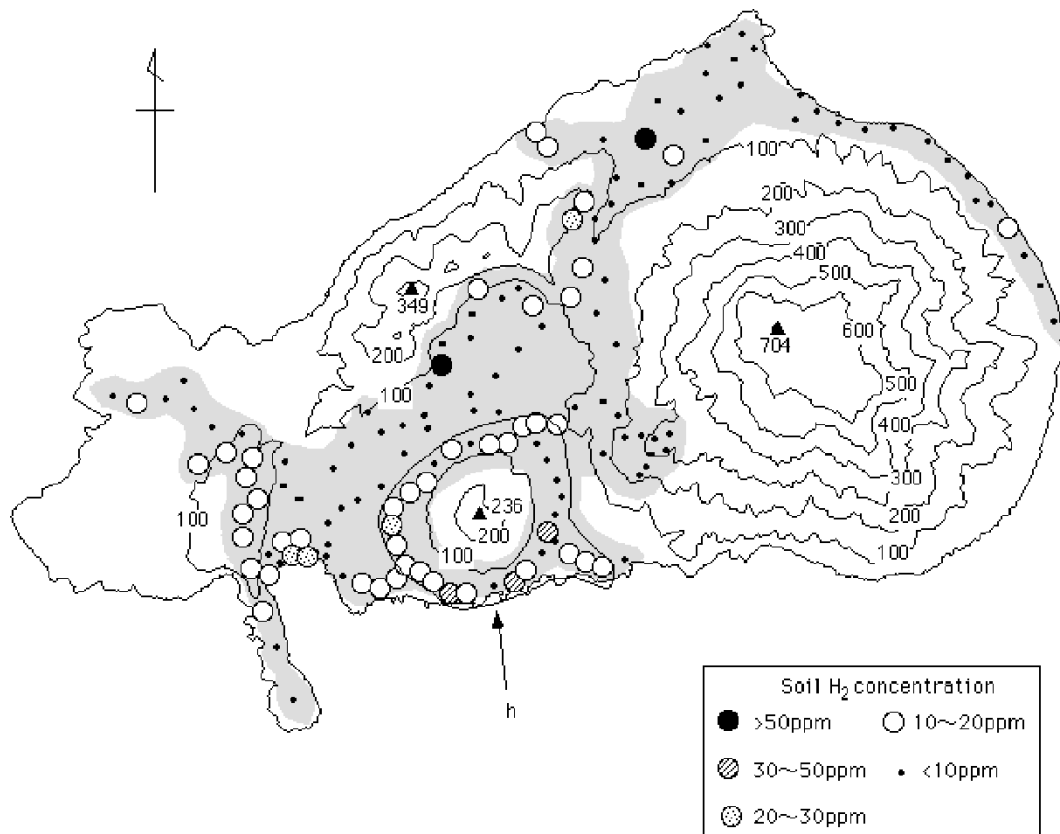


Fig. 4. Distribution of H₂ concentration in soil gases. The areas indicated with letters are anomalous (see text).

tions, up to 10%, were also measured around the Heikenjojo area (b in Fig. 2). In other areas, the CO₂ concentrations were commonly less than 1%, although higher concentrations of up to 3% were also measured at some points.

The $\delta^{13}\text{C}_{\text{CO}_2}$ values were less than -20‰ in most areas, but higher values were obtained from some specific areas (Fig. 3), including the foot of Yahazudake (d), southwest flank of Iwodake (e), Ketsunohama (f), and Nagahama area (g). In the Yahazudake area, the soil temperature was $80\sim 85^\circ\text{C}$, and weak steaming was also observed. In contrast, the temperature was almost the same as the background ($\sim 18^\circ\text{C}$) on the southwest flank of Iwodake volcano. At Ketsunohama, most of the samples had high $\delta^{13}\text{C}_{\text{CO}_2}$ values, above -10‰ , and temperature anomalies up to 28°C were also detected. In the Nagahama area, the $\delta^{13}\text{C}_{\text{CO}_2}$ values were moderately high, ranging from -10 to -20‰ , and the soil temperature was slightly higher ($21\sim 27^\circ\text{C}$) than background. Distribution of the high temperature areas at Ketsunohama and Nagahama correlate with the location of hot-spring sources.

Helium concentration in the soil gas was around 5 ppm, close to the He concentration in air. In contrast, H₂ concentrations greater than air were observed in some areas (Fig. 4). The areas of high H₂ concentration were located mainly around the basaltic volcano, Inamuradake (h), and also along the caldera rim. It is known that H₂ can be generated when an iron pipe is inserted into the ground (Sugisaki *et al.*, 1983). We conducted soil gas surveys with the same stainless steel pipe used at Izu-Oshima and Miyakejima vol-

canoes, Japan (Shimoike, 1999; Hernandez *et al.*, 2001b), but hydrogen was not detected at either volcanoes. Therefore, the high hydrogen detected around Inamuradake was unlikely an artifact of the sampling procedure. The occurrence of high H₂ around Inamuradake and along the caldera rim likely indicates a deep origin of the H₂. In contrast, H₂ can also be created by near-surface phenomena such as rock-water interaction along fault zones (Kita *et al.*, 1982), possibly along the caldera walls.

4.2 Soil CO₂ flux distribution

The CO₂ flux at the sample locations ranged from 0.01 to $5640\text{ g m}^{-2}\text{ day}^{-1}$. An area of high CO₂ flux is located along the caldera rim and around Inamuradake (Fig. 5). The CO₂ flux values do not show any clear correlation with the CO₂ concentration and $\delta^{13}\text{C}_{\text{CO}_2}$ values of the soil gas. In order to quantify the total flux from Satsuma-Iwojima volcano, the survey area was classified into five areas according to the CO₂ flux (Fig. 5). The total CO₂ flux was calculated by multiplying each area by its average CO₂ flux. The total soil CO₂ flux from the survey region, i.e., not including the upper volcano flank, was estimated to be 80 t day^{-1} .

Soil CO₂ flux measurements were also conducted in the summit area of Iwodake during 2000 (Fig. 1). However, we could not detect any diffuse CO₂ flux even in the area surrounding fumaroles, where the CO₂-rich gases discharges. Since the area in and around the crater is covered with thick sulfur deposits, their low permeability may prevent volcanic gas from passing through the soil, concentrating instead in the channels to the fumaroles.

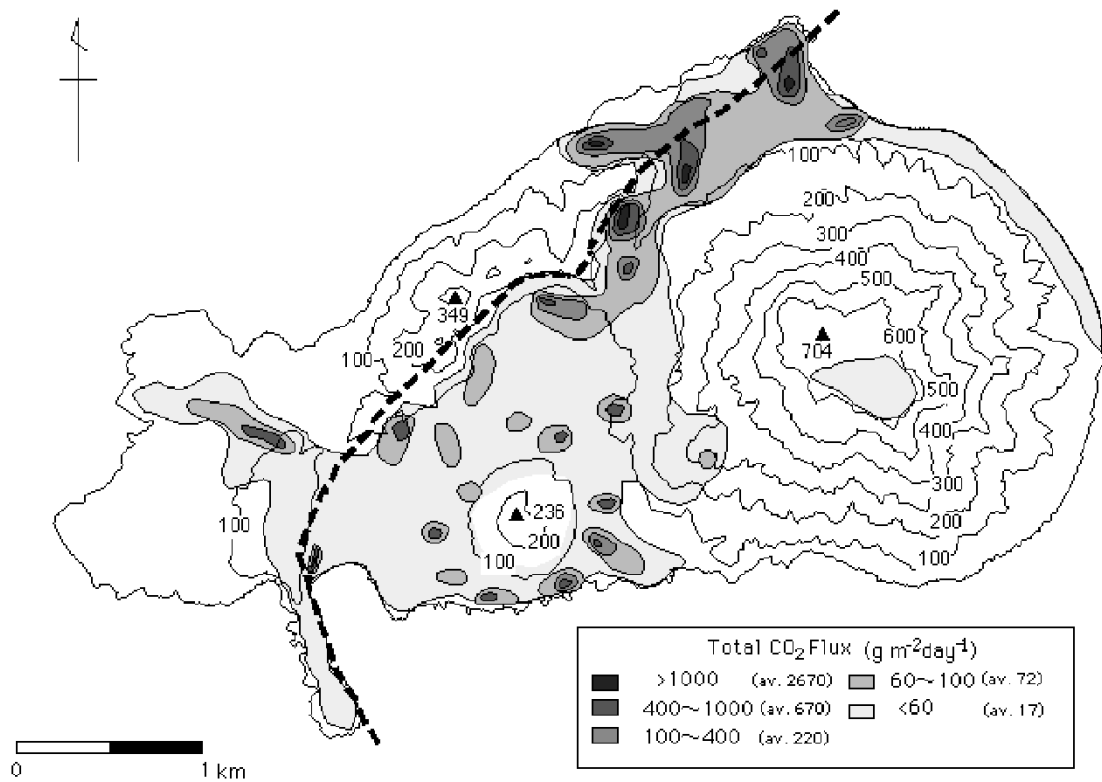


Fig. 5. Distribution of soil CO₂ flux at Satsuma-Iwojima volcano.

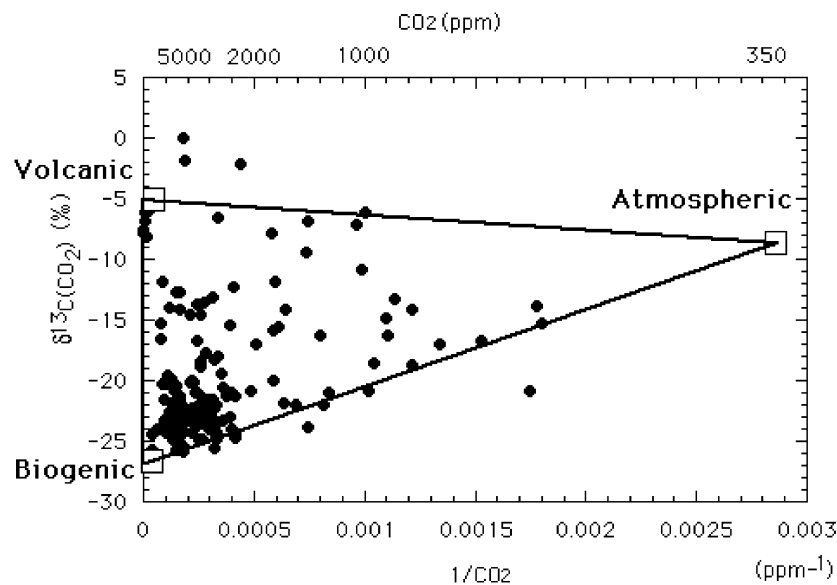


Fig. 6. Relationship between CO₂ concentration and $\delta^{13}\text{C}_{\text{CO}_2}$ of the soil gas. Lines show mixing lines among the three endmembers, CO₂ of volcanic, biogenic and atmospheric origin (see text).

Change in meteorological conditions, such as rainfall, soil humidity, wind speed and atmospheric pressure can affect soil gas fluxes (Chiodini *et al.*, 1996). However, since these meteorological conditions were not monitored on site during the survey, their possible effects were neglected in the following discussion. There was no rainfall during and several days prior to the survey in 1999. The atmospheric pressure

was continuously monitored at a meteorological observatory 50 km from Satsuma-Iwojima. The pressure was relatively stable during the survey in 1999, ranging from 1015.1 to 1019.4 hPa. This small variation in the atmospheric pressure was unlikely to cause a significant difference in the measured CO₂ flux values (Hernandez *et al.*, 2001b).

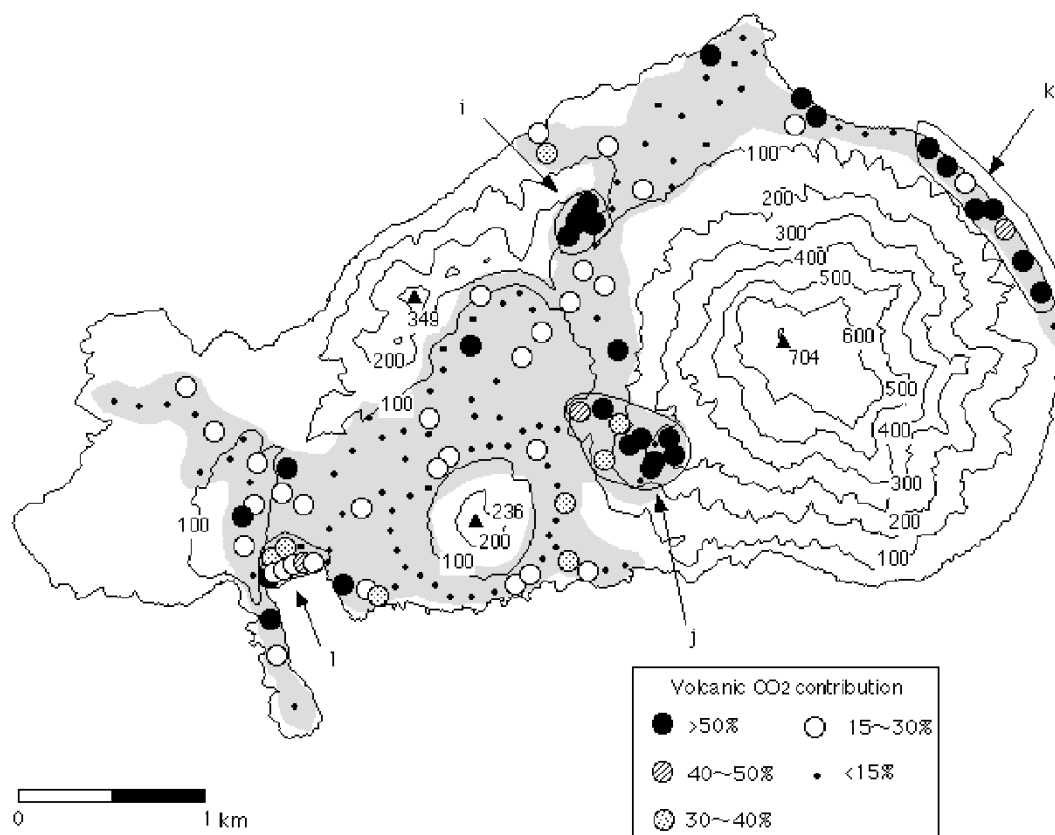


Fig. 7. Relative proportion of the contribution of volcanic CO₂ to soil CO₂ flux. The areas indicated with letters are anomalous (see text).

5. Discussion

5.1 Contribution of volcanic CO₂ to soil gas

The wide range of $\delta^{13}\text{C}_{\text{CO}_2}$ values indicates various origins of CO₂ in the soil gases. Soil CO₂ in geologically inactive areas is commonly considered to be a mixture of CO₂ of atmospheric and biogenic origins (Cerling *et al.*, 1991). The variation of CO₂ concentration and $\delta^{13}\text{C}_{\text{CO}_2}$ values indicated that the soil gas of Satsuma-Iwojima clearly contains a third source of CO₂. The CO₂ concentration and $\delta^{13}\text{C}_{\text{CO}_2}$ of atmospheric and biogenic CO₂ are 0.035%, -8‰ and ~100%, -27‰, respectively. The composition of the third component can be estimated as ~100% CO₂ and $\delta^{13}\text{C}_{\text{CO}_2} = 0 \sim -5\%$ (Fig. 6). The $\delta^{13}\text{C}_{\text{CO}_2}$ of high-temperature volcanic gas is about -5‰ (Shinohara *et al.*, 2002). Therefore, we conclude that the third source of CO₂ is volcanic in origin. Although there are other possible sources of CO₂, such as thermal decomposition of sediments or metamorphism, their contributions are likely to be minor around an active volcano.

We calculated the contribution from each source, assuming that the soil gas CO₂ is a mixture of volcanic, biogenic, and atmospheric CO₂. We assume that the CO₂ concentration and $\delta^{13}\text{C}_{\text{CO}_2}$ of volcanic, biogenic and atmospheric origin to be 100% and -5‰, 100% and -27‰, 0.035% and -8‰, respectively. The concentration of CO₂ in the volcanic and biogenic components in the soil gas is not necessarily 100%. However, because the uncertainty in CO₂ concentration of the high CO₂ end-members has little effect on the mixing calculation given below, the mixing lines among

these CO₂ sources are shown as straight lines (Fig. 6). Most of the data plot inside the three mixing lines, consistent with the three end-member mixing hypothesis. The majority of data plot near the biogenic end-member, indicating an important contribution from a biogenic origin. Less than 5% of the points plot outside these mixing lines, and their $\delta^{13}\text{C}$ values have been changed to fit the nearest mixing line in order to calculate the relative contributions.

For the majority (70%) of samples, the volcanic contribution is less than 15%. A high contribution of volcanic CO₂, more than 50%, is estimated for samples collected from several small areas (Fig. 7); Yahazudake (i), southwest flank of Iwodake volcano (j) Ketsunohama (k), and Nagahama hot spring area (l). The temperature of these areas, except Iwodake, was higher than that of the background level.

5.2 Evaluation of volcanic CO₂ flux

The volcanic CO₂ flux is obtained by the following equation,

$$F(\text{CO}_2)_{\text{vol}} = F_{\text{CO}_2}(\text{obs}) \times C(\text{vol}), \quad (2)$$

where $F(\text{CO}_2)_{\text{vol}}$ is the volcanic CO₂ flux ($\text{g m}^{-2} \text{day}^{-1}$), $F_{\text{CO}_2}(\text{obs})$ is the measured CO₂ flux ($\text{g m}^{-2} \text{day}^{-1}$) and $C(\text{vol})$ is the contribution of the volcanic CO₂(%).

Volcanic CO₂ flux shows a similar distribution to the measured soil CO₂ flux (Figs. 5 and 8). High volcanic CO₂ emission, more than $1000 \text{ g m}^{-2} \text{day}^{-1}$, occurs close to Yahazudake (Fig. 8, A), which is a part of the caldera rim. The soil temperature is about 80~85°C, and there is weak steam venting. The trees were killed by heat and/or high concen-

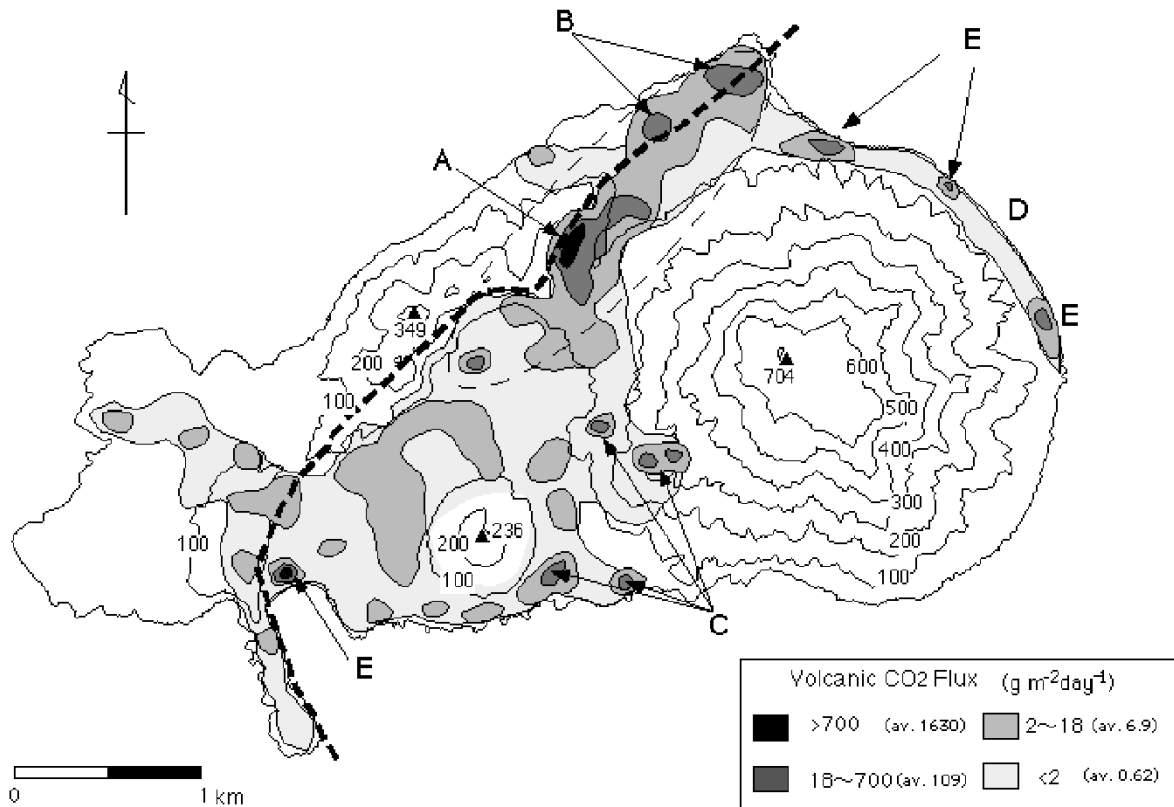


Fig. 8. Areas of anomalous soil CO₂ flux of volcanic origin at Satsuma-Iwojima volcano.

tration of CO₂. Another area of high flux is located further NW along the caldera rim (B). This suggests that a permeable zone may exist along the caldera rim, and serves as a channel for degassing from a deep volcanic source. Around Iwodake volcano (C and D), a high volcanic CO₂ flux occurs in several areas, indicating that volcanic gas similar to that discharging from the summit crater might also diffuse through the soil as far as 2 km from the summit. Although the total CO₂ flux is low at Ketsunohama (D), the volcanic CO₂ flux is relatively high because of the high volcanic contribution. A high volcanic CO₂ flux is also present on the coast around Nagahama hot spring (E).

The total volcanic CO₂ flux is calculated by the same procedure as the total CO₂ flux. The total diffuse volcanic CO₂ flux is estimated to be about 20 t day⁻¹. However, the surveyed region covers only part of the island. We did not investigate a major part of Yahazudake, Inamuradake and Iwodake rhyolitic dome because of the steep topography, heavy vegetation and presence of lava flows. Therefore, the calculated total volcanic CO₂ flux is a minimum estimate for diffuse degassing.

High-temperature volcanic gas discharges from the summit crater of Iwodake. The volcanic CO₂ flux associated with fumarolic discharge can be estimated using the SO₂ flux and CO₂/SO₂ ratio of the fumarolic gas. The SO₂ flux from the volcano was measured by correlation spectrometry (COSPEC), and in October 1999 it was about 320 t day⁻¹ (Kazahaya *et al.*, 2002). Since the average CO₂/SO₂ mole ratio in high-temperature fumaroles was about 0.45 (Shinohara *et al.*, 2002), the volcanic CO₂ from the summit

fumaroles was about 100 t day⁻¹. Therefore, the estimated volcanic CO₂ flux by diffuse degassing was about 20% of the amount of the fumarolic discharge. Considering that this estimate does not include the diffuse CO₂ emission from the major part of Inamuradake and Iwodake, we conclude that diffuse emission contributes significantly to the degassing of the magma beneath Satsuma-Iwojima volcano.

The volcanic CO₂ flux accounts for only 25% of the total soil CO₂ flux (80 t day⁻¹) at Satsuma-Iwojima volcano. Although the soil CO₂ fluxes have been measured at many volcanoes (e.g., Chiodini *et al.*, 1998), the volcanic CO₂ contribution has not been estimated in the previous studies. Therefore, it is likely that the previous estimates of soil CO₂ flux from volcanoes resulted in an overestimate of the volcanic CO₂ flux. This study demonstrates that the contribution from non-volcanic sources can be important for the total soil CO₂ flux at volcanoes. Thus, it is necessary to evaluate the contribution from each source in order to estimate the volcanic CO₂ flux through soils.

6. Conclusions

We conducted a soil gas survey at Satsuma-Iwojima volcano, Japan. The chemical composition of the soil gas was a mixture of CO₂ and air components. We estimated the volcanic contribution at each sampling point by assuming mixing of gas from three origins; volcanic, atmospheric, and biogenic. Most of the CO₂ is of biogenic origin, but a strong volcanic contribution was found close to the caldera rim. We estimated the passive volcanic CO₂ flux through soil by multiplying the volcanic contribution by the measured CO₂ flux.

Although the volcanic CO₂ flux was at a background level over much of the island, high volcanic CO₂ degassing area was found along the caldera rim and around the active rhyolitic dome. Total output of diffuse volcanic CO₂ through soil was estimated to be 20 t day⁻¹, accounting for 25% of the total soil CO₂ flux from Satsuma-Iwojima island. The soil CO₂ flux of volcanic origin is about 20% of the volcanic CO₂ flux from the summit fumaroles. This study indicates that an evaluation of the volcanic contribution to the soil gas is necessary to estimate the total volcanic CO₂ flux from a volcano.

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Y. Shimoike (e-mail: shimo@rimr.akita-u.ac.jp), K. Kazahaya, and H. Shinohara