Title	Noble gas and carbon isotopic compositions of petit-spot lavas from southeast of Marcus Island
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Citation	Earth and planetary science letters, 497, 139-148 https://doi.org/10.1016/j.epsl.2018.06.020
Issue Date	2018-09-01
Doc URL	http://hdl.handle.net/2115/79162
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Type	article (author version)
File Information	EPSL-D-18-00009R3.pdf

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Noble gas and carbon isotopic compositions of petit-spot lavas from
southeast of Marcus Island

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Keywords: carbon isotopic ratio, crustal assimilation, lithosphere–asthenosphere
boundary, Marcus Island, noble gases, petit-spot

22 Abstract

23

24 We measured noble gas isotopic compositions of quenched lavas sampled from
25 seamounts, so-called petit-spot volcanoes, on the 160-million-year-old northwestern
26 Pacific Plate. The samples $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios were, respectively, 2.5–8.3 Ra
27 and up to 1735, where Ra stands for atmospheric $^3\text{He}/^4\text{He}$, which are analogous to or
28 lower than those of MORB. Considering narrow sampling regions, a secondary effect
29 might be responsible for variation of the data.

30 During ascent and subsequent cooling of magma in the oceanic lithosphere,
31 chemical components in the magma will be assimilated with those in the lithosphere.
32 Correlation between $\text{CO}_2/^3\text{He}$ ratios and carbon isotopic ratios suggests that carbon was
33 affected by the incorporation of seafloor carbonate. The same would be true of noble
34 gases. The mixing of noble gases among a mantle source, an atmospheric component
35 dissolved in seawater and a radiogenic component can explain the data distribution. No
36 $^3\text{He}/^4\text{He}$ ratio exceeds the MORB-like value. The mantle source of the petit-spot magma
37 was likely to have had a MORB-like $^3\text{He}/^4\text{He}$ ratio originally. The eruption of petit-spot
38 magma shows a close relation with the bending of subducting oceanic plates. The
39 MORB-like $^3\text{He}/^4\text{He}$ ratio supports the hypothesis that the petit-spot magma is derived
40 from the lithosphere–asthenosphere boundary.

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44 1. Introduction

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46 The occurrence of partial melts in the lithosphere–asthenosphere boundary (LAB) is
47 an attractive hypothesis to explain a seismic low velocity zone and the high-electric
48 conductive layer in the upper mantle (Kawakatsu et al., 2009; Kumar and Kawakatsu,
49 2011; Schmerr, 2012). Moreover, this hypothesis is supported by results of
50 high-pressure experiments conducted to assess peridotite solidus (Sifré et al., 2014;
51 Wyllie, 1988) and the viscosity of silicate magma (Sakamaki et al., 2013).

52 If partial melt exists in asthenosphere, then it would migrate upward by its own
53 buoyancy and pool beneath the lithosphere (LAB). Later, it might erupt as intraplate
54 volcanoes. Batiza (1982) presents data related to the abundance, size, and crustal age of
55 volcanoes of the Pacific Ocean. The sizes of the largest seamounts are related to the age
56 of the sea floor on which they formed. Large seamounts can form on a thick lithosphere,
57 but only small seamounts form on a thin lithosphere (Hillier and Watts, 2007; Wessel,
58 1997), implying that the maximum volume and the rate at which magma can ascend
59 from the asthenosphere increases with the depth to which lithosphere-traversing
60 fractures must penetrate to reach the ponded melt (Foulger, 2010).

61 As some reports have already suggested (Hirano et al., 2006; Valentine and Hirano,
62 2010), vertical transport of the ponded melt becomes prominent when the stress state at
63 the base of the lithosphere becomes extensional, generating fractures within the
64 lithosphere. The melt will ascend through the fracture and engender volcanism along the
65 concavely flexed zone of the outer rise, outboard of the zone of plate subduction.

66 Additionally, horizontal migration of the ponded melt occurs because of the horizontal
67 pressure gradient induced by the topographic head of the outer rise, sustaining a
68 continuous melt supply to the volcanism (Yamamoto et al., 2014).

69 Young basalts have been discovered beside the outer rise on the northwestern
70 Pacific Plate: so-called petit-spot volcanoes (Hirano et al., 2006). They erupted at the
71 oceanward base of the outer rise of the descending Pacific Plate where flexure of the
72 subducting plate causes fractures in the lithosphere (Hirano et al., 2001, 2004, 2006,
73 2008; Machida et al., 2015; Sato et al., 2018). Based on their geochemical signatures,
74 these lavas presumably originated from partial melts in the asthenosphere, but the
75 original depth of the petit-spot magma remains uncertain.

76 A key question is whether the petit-spot volcanoes represent LAB magma or hot
77 spots fed by a mantle plume. Obayashi et al. (2006) studied the slow seismic velocity
78 anomaly beneath the ocean-ward side of the Japanese subduction zone. Based on their
79 interpretation of the seismological images, they proposed the existence of a hot thermal
80 anomaly at the 410-km discontinuity near the Honshu subduction zone. They concluded
81 that it is mainly an association of occurrence of fractional melt or a thermal anomaly
82 with magnitude of approximately 200°C. The tomographically imaged low-velocity
83 region is interpreted as a remnant of past activity of the Pacific superplume (Honda et
84 al., 2007). However, the possible occurrence of plumes is just one interpretation of the
85 tomographic image. Direct investigation of the magma must be conducted to settle the
86 provenance of petit-spot magmatism.

Applying the $^3\text{He}/^4\text{He}$ ratio is effective for identification of the origin of petit-spot magma. Mid-ocean ridge basalts (MORB) show a uniform $^3\text{He}/^4\text{He}$ ratio of $8.0 \pm 1.5 \text{ Ra}$, where Ra is atmospheric $^3\text{He}/^4\text{He}$ ratio, but $^3\text{He}/^4\text{He}$ ratios of ocean island basalts (OIB) vary widely (Sano and Fischer, 2013). Although values of 20–30 Ra are common on some ocean islands (i.e., Hawaii (e.g., Kaneoka et al., 2002; Valbracht et al., 1996), Iceland (e.g., Moreira et al., 2001; Trieloff et al., 2000), Samoa (e.g., Farley et al., 1992; Jackson et al., 2007), and Galapagos (e.g., Kurz and Geist, 1999; Kurz et al., 2009)), higher $^3\text{He}/^4\text{He}$ values have been obtained for Baffin Island (e.g., up to 50 Ra; Stuart et al., 2003). Moreover, enriched and HIMU-type OIBs show $^3\text{He}/^4\text{He}$ ratios lower than 8 Ra (Hanyu and Kaneoka, 1997; Hanyu et al., 2011). If petit-spot magma were originated from LAB, then it would have a MORB-like $^3\text{He}/^4\text{He}$ ratio because the original depth is comparable to that of MORB.

Although Hirano et al. (2006) once analyzed noble gas isotopic compositions of petit-spot basaltic glasses sampled from the northwestern Pacific Plate, they did not ascertain the $^3\text{He}/^4\text{He}$ ratio because of the samples low He contents and the low sensitivity of the mass spectrometer for He. Recently, swarm petit-spot volcanism was discovered southeast of Marcus (Minamitorishima) Island (Yokosuka YK10-05 Cruise Data). The basaltic glasses have high vesicularities up to 64%, indicating higher amounts of gases including He. For this study, we analyzed the $^3\text{He}/^4\text{He}$ ratios of the glasses using a mass spectrometer with particular sensitivity for $^3\text{He}/^4\text{He}$ ratio. The device enables us to address the origin of petit-spot magma.

2. Samples

During a survey (YK10-05 cruise) of the area on the northwestern Pacific Plate (Fig. 1) by the Japan Agency for Marine–Earth Science and Technology (JAMSTEC), basaltic rocks with glassy rims were recovered from petit-spot volcanoes using the manned research submersible Shinkai 6500 (Table 1). Basalts used for this study were recovered during two dives, 6K#1203 and 6K#1206, at locations about 100 km southeast of Marcus Island. Information related to sampling sites is reported in a cruise report (Yokosuka YK10-05 Cruise Data). Site 6K#1203 is located about 10 km north–northeast of the site 6K#1206. Each site covers a region with a major axis of less than approx. 1 km (Table 1). The lavas have a glassy rim with maximum thickness of approximately 3 mm. Assuming that the rate of palagonitization by seawater is 0.003–0.02 $\mu\text{m}/\text{year}$ (Moore et al., 1985), 0.2–1 Ma is necessary to alter the approx. 3.0-mm thick glassy rim. Therefore the eruption age of the lavas is younger than 1 Ma.

For analyses of noble gases and carbon, only fresh glassy rims were used. Ten samples from two sites were selected based on the glassy rim freshness and thickness. All samples were collected from depths greater than 5,000 m. Figure 2 represents photomicrographs of two samples. We did not make thin sections because we strove to preserve samples to the greatest extent possible for noble gas analyses. We measured the vesicularity of the almost flat plane of the glassy rim using an area analysis option of an image histogram and image software (Photoshop®; Adobe Systems Inc.). The measured area was more than 30 mm^2 for each sample. During sample preparation for the

vesicularity measurement, the obtained vesicularity is somewhat higher than the true value because the probability of generation of fracture within the glassy rim might be biased by the occurrence of vesicles. The vesicularity is greater in the samples of 6K#1203 (37–64%) than in those of 6K#1206 (22–27%). The obtained high vesicularities are not surprising for petit-spot lavas because similar values of 22–43% were reported from the other petit-spot lavas (Okumura and Hirano, 2013). Their chemical compositions have been reported elsewhere.

3. Experimental methods

Glassy rims were chipped carefully from lava samples, yielding chips of 1–7 mm. Only fresh and homogeneous glasses with no visible inclusions were handpicked with tweezers. After the samples were washed with acetone, they were rinsed with ethanol. We did not leach the samples with HNO_3 because leaching effects would be minimal for crushing experiments. After drying, the samples were weighed and loaded into the degassing apparatus.

Gases were extracted from the glassy samples by crushing in vacuum. Before crushing, mineral grains were preheated at 150°C in vacuum for 11 h. During crushing, the extracted gases were exposed to a cold trap at liquid N_2 temperature to remove gases that had adsorbed onto the newly created powder surfaces. Helium analyses were conducted using a sector-type mass spectrometer (Helix SFT; GV Instruments Ltd.) installed at the Atmosphere and Ocean Research Institute (AORI) of The University of

Tokyo, which is equipped with a conventional quadrupole mass spectrometer for analysis of argon isotopic composition. After CO₂ was separated from other components using cold traps held at liquid N₂ and ethanol-dry ice temperature, the amount of CO₂ and ¹³C/¹²C ratio were analyzed, respectively, using a manometer and a GC-IRMS system (IsoPrime100 with vario-EA system; Isoprime Ltd.) installed at AORI. The ¹³C/¹²C ratios were calibrated using a standard CO₂. Before the analysis, sulfur compounds were removed using an adsorbent in EA. Sensitivities and isotopic ratios for He and Ar were calibrated using an air standard. An artificial helium standard gas (HESJ) with ³He/⁴He ratio of 20.63 ± 0.10 Ra (Matsuda et al., 2002) was also applied to verify the stability of the mass spectrometer for ³He/⁴He. Procedural crushing blanks for ⁴He, ⁴⁰Ar, and CO₂ were, respectively, less than 3.0 × 10⁻¹⁰, 4 × 10⁻⁹, and 1.6 × 10⁻³ ccSTP. Uncertainty for all gas abundance is less than 5% in 1 σ. Analytical conditions for noble gases at the AORI were described in an earlier report (Sano et al., 2008).

4. Results

Measurement results are presented in Table 2. The gas abundances vary greatly among samples. Several factors affect gas abundances extracted by crushing, such as the number and size of vesicles, size and quality of glass fragments, and crushing efficiency. For example, sample 1206-R01 has insufficient CO₂ for analysis. This might derive from both weathering and relatively low vesicularity (Table 1). However, sample

1206-R03, which has centimeter-sized tabular cavities in the non-glassy part (Yokosuka YK10-05 Cruise Data), shows the highest gas abundance.

The $^3\text{He}/^4\text{He}$ ratios of the present samples are 2.5–8.3 Ra, which are analogous to or much lower than the average value of Pacific MORB (8.13 ± 0.98 Ra; Graham, 2002). Argon isotopic ratios ($^{40}\text{Ar}/^{36}\text{Ar}$) are higher than those for air (295.5) but much lower than the MORB values, which are often up to 10,000 and which are sometimes as high as 44,000 (Burnard et al., 1997; Moreira et al., 1998).

$\delta^{13}\text{C}$ values vary from -3.7 to +2.9‰, which is higher than that of either MORB or OIB ($-6.5 \pm 1.7\text{‰}$ (White, 2015)). The $\text{CO}_2/^3\text{He}$ ratio is $5.1 \times 10^{10} - 2.5 \times 10^{12}$, which is much higher than that of MORB and OIB ($1 \times 10^9 - 3 \times 10^{10}$ (Jean-Baptiste et al., 2009; Marty and Jambon, 1987)). Overall, no appreciable difference in gas compositions was found between the samples obtained from the two dives.

5. Discussion

The principal purpose of this study was to clarify whether the petit-spot magma originally has a MORB-like $^3\text{He}/^4\text{He}$ ratio. The obtained $^3\text{He}/^4\text{He}$ ratios are 2.5–8.3 Ra. We evaluated them as intrinsic values of petit-spot magma.

5.1. Degassing fractionation

Magmatic degassing might influence the noble gas isotopic ratios of lavas. The $^4\text{He}/^{40}\text{Ar}^*$ ratio is useful as an indicator for examining the degree of degassing of a lava (Burnard et al., 2002; Cartigny et al., 2001; Marty and Zimmerman, 1999; Yamamoto and Burnard, 2005). This is because $^4\text{He}/^{40}\text{Ar}^*$ ratio of the mantle is determined primarily by the mantle (Th+U)/K ratio, where $^{40}\text{Ar}^*$ denotes ^{40}Ar corrected for air contamination (defined as $^{40}\text{Ar}^* = ^{36}\text{Ar} \times [(^{40}\text{Ar}/^{36}\text{Ar})_{\text{sample}} - (^{40}\text{Ar}/^{36}\text{Ar})_{\text{air}}]$). Therefore, one can estimate the degree of magmatic degassing using $^4\text{He}/^{40}\text{Ar}^*$ ratio, over the production ratio of approximately 2 (e.g., Burnard et al., 1998; Jambon et al., 1985).

Figure 3 depicts the degassing fractionation of $^3\text{He}/^4\text{He}$ and $^4\text{He}/^{40}\text{Ar}^*$ ratios. One sample (1206-R03) shows high $^4\text{He}/^{40}\text{Ar}^*$ ratio ($^4\text{He}/^{40}\text{Ar}^* = 3.8$), which is higher than the production ratio. The high $^4\text{He}/^{40}\text{Ar}^*$ ratio might be explainable by kinetic degassing (Aubaud et al., 2004). Several recent reports have described that higher magmatic CO_2 contents enhance bubble growth and engender extensive diffusive loss of He from the magma (Gonnermann and Mukhopadhyay, 2007; Weston et al., 2014). Assuming that the diffusion coefficients are based on the inverse-square-root of the atomic mass, diffusion-induced mass fractionation during degassing engenders preferential diffusive loss of helium into vesicles, resulting in generation of vesicles for which the $^4\text{He}/^{40}\text{Ar}^*$ ratio is higher than that of surrounding magma. The diffusive loss of helium into vesicles is accompanied by an increase in $^3\text{He}/^4\text{He}$ ratio of the vesicles (Fig. 3). Assuming primary magma with MORB-like $^3\text{He}/^4\text{He}$ and $^4\text{He}/^{40}\text{Ar}^*$ ratios, the noble gas isotopic composition of sample 1206-R03 cannot be reconciled with the diffusive

fractionation during kinetic degassing. Another process is necessary to interpret the data.

The kinetic degassing would be effective in interpretation of low $^4\text{He}/^{40}\text{Ar}^*$ ratio. It is noted that most samples show low $^3\text{He}/^4\text{He}$ and $^4\text{He}/^{40}\text{Ar}^*$ ratios accompanied by high $\delta^{13}\text{C}$ value and $\text{CO}_2/{}^3\text{He}$ ratios as compared with the typical mantle values. In principal, it can be explained by diffusive loss of lighter isotopes from magma. We examine the possibility based on a numerical simulation of diffusive fractionation. Figure 4 represents $^3\text{He}/^4\text{He}$ and $\delta^{13}\text{C}$ diagram of the petit-spot lava samples. We simulated the diffusive fractionation based on the model proposed by Yamamoto et al. (2009), which can calculate a fractionated elemental or isotopic ratio of a magma body bounded by two parallel planar diffusion channels. The diffusive fractionation in the magma depends on the diffusion coefficient, timescale and channel spacing. We used a diffusion coefficient for CO_2 in magma (Zhang et al., 2007). Very few studies have been done on He diffusion in silicate melts. Lux (1987) presented a diffusion coefficient for He (D_{He}) in tholeiite basalt at 1350°C as $\sim 5 \times 10^{-9} \text{ m}^2/\text{sec}$, which is roughly consistent with the extrapolated diffusivity at 1350°C proposed by Kurz and Jenkins (1981). A black broken line in Fig. 4 represents the diffusion-induced fractionation in MORB-like magma at 1350°C with channel spacing of 10 cm. It takes only one month to produce one order of magnitude lower $^3\text{He}/^4\text{He}$ ratio. In the interval, $\delta^{13}\text{C}$ value remains unchanged. The constancy in $\delta^{13}\text{C}$ results from a lower diffusion coefficient for CO_2 (D_{CO_2}) in magma. The D_{CO_2} in magma at 1350°C is $\sim 1.0 \times 10^{-11} \text{ m}^2/\text{sec}$ (Zhang et al., 2007), resulting in $D_{\text{He}}/D_{\text{CO}_2} = \sim 500$. The bubble growth driven by the

diffusion-induced kinetic degassing engenders a preferential change in $^3\text{He}/^4\text{He}$ ratio as shown in Fig. 4. Assuming that the magma has a constant isotopic composition, it is clear from the Figure 4 that the kinetic degassing cannot reproduce variation in $\delta^{13}\text{C}$ value. Furthermore it is impossible to explain positive values of $\delta^{13}\text{C}$ by the kinetic degassing of MORB-like magma. Cartigny (1998) defined the mantle $\delta^{13}\text{C}$ value to be from -8 to -2‰. Even if magma had originally had a heavier $\delta^{13}\text{C}$ value such as -2 ‰, the diffusive fractionation cannot produce the positive $\delta^{13}\text{C}$ values. Though there is no direct evidence to deny the influence of kinetic degassing on the present volatile compositions, another mechanism is necessary to explain variation in the $^3\text{He}/^4\text{He}$ and $\delta^{13}\text{C}$ values.

5.2. Air contamination

The petit-spot volcanoes are sea knolls. At the time of eruption, the possibility exists of obtaining atmospheric components dissolved in seawater. To assess the possibility, it is difficult to apply noble gas elemental ratios such as $^3\text{He}/^{36}\text{Ar}$ ratio because the obtained noble gases extracted from vesicles are the result of the degassing fractionation, as described in the previous section. Figure 5 presents He and Ar isotopic ratios of petit-spot samples accompanied by MORB and OIB. Actually, MORB are shown along a mixing line between a MORB source and air. If the $^{40}\text{Ar}/^{36}\text{Ar}$ ratio is higher than approximately 300, then the atmospheric influence on $^3\text{He}/^4\text{He}$ ratio is negligible for MORB (Fig. 5). In the case of OIB, during the ascent of OIB magma, noble gases in the

magma would be assimilated with those in the surrounding lithosphere (MORB source). Therefore, the distribution of OIB in Fig. 5 is explainable by three-component mixing among the OIB source, the MORB source, and air.

Atmospheric contaminants of several types might exist in oceanic basalt glasses that have identical air-like isotopic ratios, but which have different noble gas abundance ratios such as those found for air dissolved in deep-sea water, the surface atmosphere, and fractionated air infiltrated into the pore space of samples (Ballentine and Barfod, 2000; Raquin et al., 2008; Roubinet and Moreira, 2018). However, the mixing lines between the MORB source and the contaminant are mutually indistinguishable in Fig. 5. Samples obtained from each dive were collected from an individual lava flow, which originally has a homogeneous noble gas isotopic composition. Consequently, the atmospheric influence on each lava flow should be represented as a single mixing line with atmospheric contaminants. They are, however, scattered in Fig. 5. Air addition cannot be the cause of the variation of the $^3\text{He}/^4\text{He}$ ratios of the samples.

5.3. Radiogenic ingrowth

For this study, we extracted gases by crushing glass fragments in vacuum. The crushing method is probably effective for extracting gases trapped in vesicles, which contain only small amounts of in-situ generated nuclides (e.g., Graham et al., 1992a,b; Kurz, 1986). Intense crushing, however, presents the possibility of extracting radiogenic

nuclides accumulated within crystal lattices (Yokochi et al., 2005). We examine the effects of in-situ post eruptive addition of ^4He on $^3\text{He}/^4\text{He}$ ratios.

The eruption age and the U and Th contents of the present petit-spot lavas were estimated respectively as 1.1 Ma, 0.6–0.7 ppm, and 2.7–3.1 ppm (Yokosuka YK10-05 Cruise Data). The amount of in-situ generated ^4He during 1 Ma is calculated as approx. 2×10^{-7} ccSTP/g, which is comparable to or higher than the ^4He contents obtained in this study (Table 2). Lava samples from this volcano show uniform U and Th contents (Yokosuka YK10-05 Cruise Data), resulting in uniform ^4He contents (atoms/gram) generated after eruption. In addition, the crushing efficiency might be almost constant throughout the entire crushing because an operator did all crushing using crushing vessels of the same type: radiogenic ^4He contents (atoms/gram) extracted by the crushing would be almost constant for all samples. Lava samples with lower ^4He contents are susceptible to the post eruptive generation of ^4He within the matrix. Assuming that the petit-spot magma originally has a uniform $^3\text{He}/^4\text{He}$ ratio and variable ^4He contents, the effect of the post eruptive accumulation of ^4He results shows a negative correlation between $^3\text{He}/^4\text{He}$ ratios and ^4He contents. No correlation is apparent in Fig. 6.

Correlation of noble gas isotopic compositions with vesicularity is also a useful indicator to evaluate the radiogenic influence. Figure 7 depicts the $^3\text{He}/^4\text{He}$ ratio versus vesicularity. The vesicularity is defined as the proportion of a vesicle volume to a given volume of a lava sample. Radiogenic nuclides accumulated in the vesicles are insignificant when compared to those in the lava matrix. Therefore, radiogenic

influences are expected to be evident on the lava samples, especially with low vesicularity. No such tendency is apparent in Fig. 7. Although we cannot categorically deny the radiogenic influence on $^3\text{He}/^4\text{He}$ ratios, other factors are responsible for the wide distribution of $^3\text{He}/^4\text{He}$ ratios.

5.4. Crustal assimilation

The $^3\text{He}/^4\text{He}$ ratios of the petit-spot samples overlap data distribution of EM-type or HIMU-type OIB rather than MORB (Fig. 5). Hanyu and Kaneoka (1997) reported $^3\text{He}/^4\text{He}$ ratios of HIMU as 6.8 ± 0.9 Ra. Based on the low and uniform $^3\text{He}/^4\text{He}$ ratio, they concluded that it represents a general characteristic of the mantle source region for HIMU lavas. The wide variation in $^3\text{He}/^4\text{He}$ of the present petit-spot lavas dismisses the possibility that the obtained $^3\text{He}/^4\text{He}$ ratios are an inherent characteristic of the petit-spot magma. We next consider the possibility of crustal assimilation. Kumagai et al. (2003) and Moreira et al. (2003) reported noble gas isotopic compositions of oceanic crust gabbro collected from Indian Plate. The $^3\text{He}/^4\text{He}$ ratios analyzed by total melting were, respectively, 0.2–8.1 and 2.9–8.2 Ra. The $^3\text{He}/^4\text{He}$ ratios of the gabbro well overlap the $^3\text{He}/^4\text{He}$ ratios obtained from this study (2.5–8.3 Ra). However, it is unlikely that the wide variation in the present $^3\text{He}/^4\text{He}$ ratio is caused mainly by assimilation with the lower crust. We re-emphasize that, despite the narrow sampling region for each site within approx. 1 km (Table 1), wide diversity of isotopic compositions is apparent. Particularly, samples 1206-R03 and 1206-R04 were recovered from an almost identical

position. The isotopic compositions of the samples mutually differ. It is necessary to consider the possible influence of a local component located near the seafloor.

Although major element compositions are commonly used to assess the assimilation with local components, we applied C-He systematics for evaluation of their direct involvement to volatiles in the samples. Figure 8 shows $\text{CO}_2/{}^3\text{He}$ and $\delta^{13}\text{C}$ of the petit-spot samples together with end-member data for mantle (MORB and OIB), organic sediment and various types of marine limestone including slab carbonate. Assuming the C in a sample is a mixture of mantle, sediment components, and marine carbonate, we were able to make quantitative estimates of carbon contributions from those based on a mass balance calculation using $\text{CO}_2/{}^3\text{He}$ and $\delta^{13}\text{C}$ values as follows.

$$\delta^{13}\text{C}_O = M \cdot \delta^{13}\text{C}_M + L \cdot \delta^{13}\text{C}_L + S \cdot \delta^{13}\text{C}_S$$

$$1/({}^{12}\text{C}/{}^3\text{He})_O = M/({}^{12}\text{C}/{}^3\text{He})_M + L/({}^{12}\text{C}/{}^3\text{He})_L + S/({}^{12}\text{C}/{}^3\text{He})_S$$

$$M + L + S = 1$$

Therein, M, L, and S respectively denote the fractions of mantle, limestone, and sediment. Subscripts O, M, L, and S respectively refer to the obtained data, mantle, limestone, and sediment. Taking values of $\delta^{13}\text{C}_M = -6.5 (\pm 2.5) \text{‰}$ (Sano and Marty, 1995), $\delta^{13}\text{C}_L = 0 (\pm 3) \text{‰}$ (Schidlowski, 1988) and $\delta^{13}\text{C}_S = -30 (\pm 10) \text{‰}$ (Sano and Marty, 1995), and $({}^{12}\text{C}/{}^3\text{He})_M = 1.5 \times 10^9$, $({}^{12}\text{C}/{}^3\text{He})_L = 1.0 \times 10^{13}$ and $({}^{12}\text{C}/{}^3\text{He})_S = 1.0 \times 10^{13}$ (Sano and Marty, 1995), one can calculate the contribution of carbon provided by the three components M, L, and S in the samples. Results indicate that the carbon in the samples is derived mainly from component L, which has a contribution of more than 85%.

However, the sampling depth (5126–5361 m: Table 1) is deeper than the carbonate compensation depth (approx. 4500 m). Probably, the eruption depth is not different from the sampling depth, which implies that the carbon in the samples is not derived from seafloor carbonate. High CaCO_3 production at the shallow mid-ocean ridges produces a calcareous ooze (calcium-carbonate sediments) that has been covered by the seabed. Then the proportion of CaCO_3 can be preserved in bottom sediments as depicted in Fig. 9. When petit-spot magma penetrates through the oceanic plate, it must be assimilated more or less with the ooze. Consequently, carbon isotopic compositions of the petit-spot lava are affected by crustal assimilation, especially for a small lava body. The compositions also affect other components. In the following section, we evaluate the crustal assimilation effects on noble gases.

5.5. Pristine $^3\text{He}/^4\text{He}$ ratio of petit-spot magma

Assimilation with seafloor sediment is not confined to carbon materials. The relation of the isotopic ratios between carbon and helium is shown in Fig. 4. The data are distributed along a mixing line between a mantle component and limestone, as presented in Fig. 8, indicating clearly that the crustal assimilation also affects helium. We examine the original $^3\text{He}/^4\text{He}$ ratio of the samples using a diagram of $^3\text{He}/^4\text{He}$ versus $^{40}\text{Ar}/^{36}\text{Ar}$ ratios (Fig. 10). Noble gases in seafloor sediments can be expressed as a mixture of the atmospheric component dissolved in seawater and radiogenic nuclides. De Vleeschouwer et al. (2017) reported the K contents of seafloor sediments. The

averaged value of the sediments sampled at seafloor deeper than 5,000 m is 1.42 ± 0.92 wt%. Assuming that a sediment originally has an atmospheric $^{40}\text{Ar}/^{36}\text{Ar}$ ratio (295.5), ^{40}Ar of $2-8 \times 10^{-5}$ ccSTP/g (Matsuda and Nagao, 1986) and K content of 0.5–2.3wt%, the $^{40}\text{Ar}/^{36}\text{Ar}$ ratio changes from the atmospheric value to 306–493 for 160 Ma, which is the plate age at which the present petit-spot volcano erupted. We put four seafloor sediments with gradations of radiogenic addition indicated as components A–D in Fig. 10. The mixing lines between MORB source and the components can adequately explain all the data. We cannot exclude the possibility that the data distribution can be interpreted by mixing with OIB magma as a mantle component. However, no $^3\text{He}/^4\text{He}$ ratio exceeds the MORB value. Actually, MORB magma is apparently the more likely candidate for a mantle end-member. This inference is supported by the Ne-Ar isotopic ratios corrected for air-addition that represents values similar to those of MORB (Hirano et al., 2006). Therefore we inferred that the petit-spot magma originated from a MORB-source mantle. The petit-spot magma is regarded as a melt that is extruded passively through fractures of the subducting oceanic plate. Consequently, one can reasonably infer that the petit-spot magma originated from the lithosphere–asthenosphere boundary.

Finally, we consider reasons for which the isotopic compositions of the present petit-spot lavas were affected intensely by those of seafloor sediments. The volume of lava is likely to be interrelated with the degree of crustal assimilation. The petit-spot volcanoes have been discovered as small seamounts, as implied by the name. A small lava body has a large surface area per unit lava volume, thereby increasing the

likelihood of crustal assimilation. Consequently, to elucidate the pristine chemical characteristics of the petit-spot magma, it is better to investigate a petit-spot volcano erupted on a younger oceanic plate, which has a less-radiogenic property. Further discovery of such petit-spot volcanoes is expected to provide a unique perspective to the ongoing debate related to the characteristics of the lithosphere–asthenosphere boundary.

6. Summary

We analyzed noble gas and carbon isotopic compositions of ten basaltic glasses from two submarine lava flows. They are part of the petit-spot volcanoes, which erupted within one million years at the 160-million-year-old northwestern Pacific Plate. The carbon isotopic compositions suggest that the petit-spot lavas were assimilated with calcareous ooze in seafloor sediments. The effect also bears on noble gases. The seafloor sediment on the old oceanic plate accumulates radiogenic nuclides such as ^4He and ^{40}Ar . Assuming radiogenic noble gas isotopic compositions of the seafloor sediment, all the data can be interpreted as a resultant of mixing between a mantle component and the seafloor sediment. The lack of $^3\text{He}/^4\text{He}$ ratio exceeding MORB-like value suggests that the petit-spot magma is genetically related to MORB source mantle rather than OIB source mantle. The petit-spot volcanism has been discovered beside the outer rise on the northwestern Pacific Plate, where the stress state at the base of the lithosphere becomes extensional, engendering the production of fractures. If there is ponded melt at the LAB, then the generation of fractures induces vertical transport of the LAB melt. It then

engenders eruption of magma with MORB-like isotopic composition. Consequently,
petit-spot magmatism with MORB-like isotopic compositions represents the possible
occurrence of a melt-rich layer beneath the oceanic lithosphere.

Acknowledgments

We are extremely grateful to Jens Hopp and to an anonymous reviewer, who
provided thoughtful comments related to the manuscript. We are deeply appreciative of
the cooperation of all crews, marine technicians, and shipboard scientific parties of the
JAMSTEC cruises (YK10-05). We also thank Dr. N. Hirano for access to a valuable
sample from this area. This study was supported by Grants-in-Aid for Scientific
Research (Nos. 26610136 and 25287139) from the Japan Society for the Promotion of
Science.

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Figure captions

Figure 1. Bathymetric map of the study area. The red square represents the sampling area. Topographic data are from Amante and Eakins (2008).

Figure 2. Photomicrographs of thin slabs of two petit-spot lava glasses. Scale bars show 1 mm. The respective vesicularities of samples 1203-R06 and 1203-R17B were 40.3 and 60.2%.

Figure 3. $^3\text{He}/^4\text{He}$ versus $^4\text{He}/^{40}\text{Ar}^*$ of the petit-spot lava samples, where $^{40}\text{Ar}^*$ denotes ^{40}Ar corrected for air contamination (defined as $^{40}\text{Ar}^* = ^{36}\text{Ar} \times [(^{40}\text{Ar}/^{36}\text{Ar})_{\text{sample}} - (^{40}\text{Ar}/^{36}\text{Ar})_{\text{air}}]$). M denotes MORB. The $^3\text{He}/^4\text{He}$ ratio of MORB is 8.2 ± 0.7 (Hilton et al., 1993). The $^4\text{He}/^{40}\text{Ar}^*$ ratio of MORB is 1.6–3.5, which is calculated as follows. The radiogenic $^4\text{He}/^{40}\text{Ar}$ ratio of MORB, accumulated over 4.6 Gyr were calculated, respectively, as 1.6, using the K/U and Th/U ratios for MORB as 12,340 (Gale et al., 2013) and 3.16 (Gale et al., 2013). Possible extensive loss of ancient accumulated gas would occur through strong and widespread magmatism and subsequent continental growth. The accumulated $^4\text{He}/^{40}\text{Ar}^*$ ratio for 1 Gyr was 3.5. Broken lines represent noble gas compositions of vesicles nucleated from magma with a MORB-like value.

Figure 4. $^3\text{He}/^4\text{He}$ and $\delta^{13}\text{C}$ diagram of the samples and end-members. End-members of mantle (MORB), marine limestone (Limestone), and organic sediment (Sediment) are

also shown. The references of the end-members are the following: $\delta^{13}\text{C}_{\text{Mantle}} = -6.5 (\pm 2.5) \text{‰}$ (Sano and Marty, 1995), $\delta^{13}\text{C}_{\text{Limestone}} = 0 (\pm 3) \text{‰}$ (Schidlowski, 1988) and $\delta^{13}\text{C}_{\text{Sediment}} = -30 (\pm 10) \text{‰}$ (Sano and Marty, 1995), and $(^3\text{He}/^4\text{He})_{\text{MORB}} = 8.13 (\pm 0.98) \text{ Ra}$ (Graham, 2002). Helium in limestone and sediment are a mixture of atmospheric (1 Ra) and radiogenic component ($< 1 \text{ Ra}$). Gray broken lines represent mixing lines among the end-members. Error bars for the samples represent 1σ uncertainties. A black broken line represents the diffusion-controlled isotopic fractionation from magma with MORB-like isotopic characteristics.

Figure 5. $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ diagram of the petit-spot lava samples, MORB, and OIB. Error bars for the samples represent 1σ uncertainties. Data sources were the following: MORB (Battani, 1996; Craig and Lupton, 1976; Hiyagon et al., 1992; Jambon et al., 1985; Kumagai and Kaneoka, 2003; 2005; Kyser and Rison, 1982; Macpherson et al., 2005; Marty and Ozima, 1986; Moreira and Allègre, 2002; Moreira et al., 1995; 1996; 1998; Niedermann and Bach, 1998; Niedermann et al., 1997; Ozima and Zashu, 1983; Sarda et al., 2000; Stroncik and Niedermann, 2016a; 2016b; Stroncik et al., 2007; 2008), Loihi (Hiyagon et al., 1992; Honda et al., 1993; Kaneoka et al., 2002; Sarda et al., 1988; Staudacher et al., 1986; Valbracht et al., 1996; 1997), Kilauea (Honda et al., 1993; Kaneoka and Takaoka, 1978; Kaneoka et al., 1986; 2002), Mauna Loa (Valbracht et al., 1996), Hualalai (Valbracht et al., 1996), Koolau (Kaneoka et al., 2002), Maui (Kaneoka and Takaoka, 1980; Kyser and Rison, 1982), Hana Ridge (Hanyu et al., 2007), Iceland (Mukhopadhyay, 2012; Tieloff et al., 2000), Réunion (Hanyu et

al., 2001; Kaneoka et al., 1986; Staudacher et al., 1990), Samoa (Poreda and Farley, 1992), Cook–Austral Archipelago (Hanyu et al., 1999), Society Archipelago (Hanyu et al., 1999; Staudacher and Allègre, 1989), Pitcairn (Honda and Woodhead, 2005; Raquin and Moreira, 2009), Galapagos (Péron et al., 2016; Raquin and Moreira, 2009), and Lau Basin (Petö et al., 2013).

Figure 6. $^3\text{He}/^4\text{He}$ and ^4He diagram of the petit-spot lava samples.

Figure 7. $^3\text{He}/^4\text{He}$ and vesicularity of glassy rim of the petit-spot lava samples.

Figure 8. Correlation diagram for $\delta^{13}\text{C}$ and $\text{CO}_2/^3\text{He}$ ratios of the petit-spot lava samples.

End-members of mantle (MORB and OIB), marine limestone, and organic sediment are

also shown. The end-member values of $\delta^{13}\text{C}$ are the same as those in Fig. 4. The

end-members values of $(^{12}\text{C}/^3\text{He})$ are the following: $(^{12}\text{C}/^3\text{He})_{\text{Mantle}} = 1.5 \times 10^9$,

$(^{12}\text{C}/^3\text{He})_{\text{Limestone}} = 1.0 \times 10^{13}$ and $(^{12}\text{C}/^3\text{He})_{\text{Sediment}} = 1.0 \times 10^{13}$ (Sano and Marty, 1995).

Dotted lines represent mixing lines among the end-members. Error bars for the samples

represent 1σ uncertainties.

Figure 9. Schematic cross-section of the oceanic lithosphere. Petit-spot lava penetrated

the oceanic crust, the surface of which is covered by seafloor mud.

882 Figure 10. $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ diagram of the samples, MORB source, and air. Error
883 bars for the samples represent 1σ uncertainties. A solid line shows the radiogenic
884 change of the seafloor mud, which originally has $^3\text{He}/^4\text{He}$ ratio of 1 Ra and $^{40}\text{Ar}/^{36}\text{Ar}$
885 ratio of 295.5. The radiogenic change is based on an original seafloor mud with K/U of
886 12,340 by mass (Gale et al., 2013) and Th/U ratio of 3.1 by mass (Staudacher et al.,
887 1989). Broken lines are mixing lines between the MORB source, with $^3\text{He}/^4\text{He}$ ratio of
888 8.2 Ra (Hilton et al., 1993) and $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of 44,000 (Burnard et al., 1997; Moreira
889 et al., 1998), and seafloor mud components (A–D), which have $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$
890 of (A) 0.1 Ra and 295.6, (B) 0.01 Ra and 299.3, (C) 0.0073 Ra and 400.0, and (D)
891 0.0072 Ra and 500.0.

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Figure 1
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Table 1

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Table 1. Petit-spot samples used for noble gas analysis

Dive# 6K#1203	Latitude (°)	Longitude (°)	Depth (m)	Vesicularity (%)	Freshness
1203-R06	23.5275	154.5802	5195	40.3	very fresh
1203-R08	23.5273	154.5804	5183	37.1	fresh
1203-R10	23.5263	154.5806	5130	46.1	fresh
1203-R12	23.5263	154.5806	5130	63.5	not fresh
1203-R17B	23.5254	154.5808	5126	60.2	very fresh
Dive# 6K#1206					
1206-R01	23.4630	154.5752	5361	26.1	not fresh
1206-R03	23.4667	154.5765	5252	22.2	very fresh
1206-R04	23.4667	154.5765	5252	27.0	fresh
1206-R05	23.4671	154.5764	5247	22.3	fresh
1206-R09	23.4743	154.5738	5182	23.9	fresh

Table 2

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Table 2. Isotopic compositions of noble gases and carbon extracted from Petit-spot glasses

Dive#	6K#1203	weight (g)	^4He	^{40}Ar	CO_2	$\text{CO}_2 / ^3\text{He}$	$^3\text{He} / ^4\text{He}$ (Ra)	$^{40}\text{Ar} / ^{36}\text{Ar}$	$\delta^{13}\text{C}$ (‰)
	1203-R06	0.9938	4.49E-08	1.74E-07	8.95E-02	5.43E+11	5.974 $\pm$ 0.045	1016.2 $\pm$ 4.2	-2.253 $\pm$ 0.114
	1203-R08	1.3772	1.60E-08	1.74E-07	9.55E-02	3.95E+12	2.455 $\pm$ 0.038	663.0 $\pm$ 2.9	-2.567 $\pm$ 0.112
	1203-R10	0.9530	3.97E-09	2.65E-08	8.11E-03	6.10E+11	5.629 $\pm$ 0.171	1734.9 $\pm$ 14.5	2.871 $\pm$ 0.265
	1203-R12	1.2065	1.67E-08	1.26E-07	3.85E-02	6.82E+11	5.486 $\pm$ 0.056	639.2 $\pm$ 2.8	1.388 $\pm$ 0.132
	1203-R17B	1.0277	5.82E-08	2.00E-07	7.52E-02	3.25E+11	6.465 $\pm$ 0.045	590.5 $\pm$ 2.6	-1.836 $\pm$ 0.115
Dive# 6K#1206									
	1206-R01	0.4809	1.48E-09	5.30E-09	n.d.	n.d.	8.253 $\pm$ 1.699	644.7 $\pm$ 9.7	n.d.
	1206-R03	1.2810	6.55E-07	3.73E-07	1.33E-01	5.07E+10	6.514 $\pm$ 0.030	550.5 $\pm$ 4.5	-3.700 $\pm$ 0.111
	1206-R04	1.1116	1.47E-08	1.36E-07	6.61E-02	2.46E+12	2.982 $\pm$ 0.048	618.3 $\pm$ 2.7	-1.883 $\pm$ 0.116
	1206-R05	0.3737	1.07E-08	1.19E-07	1.03E-01	3.51E+12	4.638 $\pm$ 0.148	835.9 $\pm$ 3.8	-1.288 $\pm$ 0.146
	1206-R09	1.1223	1.20E-08	1.51E-07	6.55E-02	3.05E+12	2.914 $\pm$ 0.055	552.6 $\pm$ 2.4	-2.469 $\pm$ 0.115

N.B., Unit for abundance is cm³STP/g. n.d.: not detected. Errors for isotopic ratios represent 1 σ uncertainties. Experimental uncertainties in amounts are generally within 10%.