

Argon isotope ratio of the plume-source deduced from high-resolution stepwise crushing extraction

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We have carried out noble gas analysis for gases released by a high-resolution stepwise crushing on basaltic glass and olivine separated from a new set of pillow basalts collected from the Loihi seamount, Hawaii. One basaltic glass sample yielded quite consistent $^3\text{He}/^4\text{He}$ ratios of about 35 Ra (Ra denotes the atmospheric ratio) for helium released over six successive crushing steps. This ratio is the highest among those so far reported for Loihi samples, suggesting that this sample represents the most primitive Loihi magma in terms of helium isotope composition. In contrast, the argon isotope ratios ($^{40}\text{Ar}/^{36}\text{Ar}$) observed in this sample appeared to be low (<400) compared with the ratio suggested for the source of Loihi magma (i.e., 8000 by Tieloff *et al.*, 2000), implying a significant contribution from the atmospheric argon. A clear binary mixing trend was defined between $^3\text{He}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios measured during successive crushing steps, which is extending from an atmospheric component to a mantle-derived component. This mixing trend permits us to estimate $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of the mantle by assuming the mantle $^3\text{He}/^{36}\text{Ar}$ ratio of 0.7 (Moreira *et al.*, 1998). For the Loihi basaltic glass sample with 35 Ra, the $^{40}\text{Ar}/^{36}\text{Ar}$ ratio without atmospheric contribution is estimated to be 3000, whereas the ratio estimated for the olivine separates with $^3\text{He}/^4\text{He}$ ratio of 27 Ra appeared to be 11000. We found that $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ (corrected for air addition) ratios determined for the present samples, as well as for other plumes (Reunion and Iceland) and MORB-popping rock, plot on a rough negative trend, suggesting that helium and argon isotope ratios in plume-derived samples might be controlled by a variable contribution from the MORB-type component to the plume endmember. Extrapolation of this trend to $^3\text{He}/^4\text{He}$ of 50 Ra as found in the Baffin Island picrite (Stuart *et al.*, 2003) yielded the plume source $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of about 1000, suggesting that the source region of the high $^3\text{He}/^4\text{He}$ mantle plume might have $^{40}\text{Ar}/^{36}\text{Ar}$ ratio being significantly less radiogenic than has usually been anticipated.

Keywords: $^3\text{He}/^4\text{He}$, $^{40}\text{Ar}/^{36}\text{Ar}$, noble gases, Loihi, mantle plume

INTRODUCTION

Noble gas isotopes are important tracers to construct models of terrestrial degassing and evolution of mantle geochemical reservoirs. Fundamental approach towards this problem is to analyze precisely the elemental and isotope compositions of generally scarce noble gases trapped in natural mantle-derived materials, such as in the basalts erupting at oceanic ridges and intraplate hotspots. Among five noble gases, far more analysis were performed for helium isotopes in oceanic volcanic rocks compared with other heavier noble gases, primarily because there is no atmospheric contamination of helium due to its low concentration in the atmospheric reservoir. It is now well

accepted that relatively narrow range in $^3\text{He}/^4\text{He}$ ratios (7 to 9 Ra, where Ra denotes atmospheric $^3\text{He}/^4\text{He}$ of 1.4×10^{-6}) of mid-oceanic ridge basalts (MORBs) in a global context requires the presence of well-mixed helium reservoir in the convecting or asthenospheric mantle. Helium isotope ratios found in ocean island basalts associated with mantle plumes are generally more variable. Some plumes (Hawaii, Iceland, etc.) yield $^3\text{He}/^4\text{He}$ ratios higher than those for MORBs and are believed to be derived from relatively primitive (or less degassed) part of the mantle. Relatively low $^3\text{He}/^4\text{He}$ ratios (<8 Ra) are also found in some islands (HIMU lavas) whose sources might contain recycled oceanic crust (e.g., Hanyu and Kaneoka, 1997). The origin and processes responsible for producing the helium isotope variability among mantle plumes are not fully understood and need further studies.

In terms of heavier noble gas isotope signatures in the mantle, the presence of a component with solar-like $^{20}\text{Ne}/^{22}\text{Ne}$ ratio helps to quantify the air contamination to sam-

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Table 1. Helium and argon analytical results on Loihi glass and olivine

Clushing strokes	⁴ He [10 ⁻⁸ cm ³ STP/g]	R/Ra	³⁶ Ar [10 ⁻¹⁰ cm ³ STP/g]	⁴⁰ Ar/ ³⁶ Ar	⁴ He/ ⁴⁰ Ar*
698-4B Glass 2.35 gram					
2	3.7 ± 0.1	36.3 ± 0.8	5.06 ± 0.1	317.7 ± 4.5	3.3 ± 0.7
10	3.8 ± 0.1	34.7 ± 0.7	2.55 ± 0.04	333.1 ± 4.8	4.0 ± 0.5
50	4.2 ± 0.1	34.3 ± 0.4	1.58 ± 0.03	349.6 ± 5.3	4.9 ± 0.5
100	1.42 ± 0.03	34.4 ± 0.6	0.33 ± 0.03	378.9 ± 9.9	5.2 ± 0.8
300	1.15 ± 0.02	37.1 ± 0.6	0.94 ± 0.03	307.0 ± 4.8	10.7 ± 4.5
600	1.10 ± 0.02	36.2 ± 0.5	0.75 ± 0.03	326.6 ± 5.4	4.7 ± 0.8
Total	15.4 ± 0.13	35.3 ± 0.3	11.4 ± 0.1	327.6 ± 2.5	7.2 ± 0.3
698-6B Glass 2.35 gram					
20	32 ± 3	25.8 ± 0.5	30.73 ± 3.1	373.7 ± 2.4	1.3 ± 0.2
696-6B [#26 >> #36] Olivine, 0.657 gram					
200	14 ± 1	25.8 ± 0.6	1.98 ± 0.20	920 ± 9	1.1 ± 0.2
1000	4.8 ± 0.5	28.5 ± 0.6	0.38 ± 0.04	1525 ± 49	1.0 ± 0.2
1500	2.5 ± 0.2	29.7 ± 0.9	0.12 ± 0.02	2347 ± 241	1.0 ± 0.2
Total	21 ± 1	26.8 ± 0.5	2.48 ± 0.20	1083 ± 24	1.1 ± 0.1
696-6B [#22 >> #26] Olivine, 0.710 gram					
400			3.75 ± 0.38	819 ± 7	
1000	7 ± 1	26.6 ± 0.4	0.42 ± 0.04	1630 ± 45	1.2 ± 0.2
1500	3.7 ± 0.4	28.3 ± 0.6	0.23 ± 0.03	1590 ± 75	1.2 ± 0.2
Total	10 ± 1	27.2 ± 0.4	4.41 ± 0.38	937 ± 15	
696-6B [#14 >> #22] Olivine, 0.557 gram					
200	22 ± 2	27.5 ± 0.3	3.12 ± 0.31	801 ± 8	1.4 ± 0.2
1000	16 ± 2	27.7 ± 0.4	0.90 ± 0.09	1741 ± 31	1.3 ± 0.2
212	2.4 ± 0.2	25.0 ± 1.2	0.34 ± 0.04	873 ± 29	1.2 ± 0.2
Total	40 ± 3	27.5 ± 0.2	4.36 ± 0.33	1002 ± 23	1.3 ± 0.1
696-6B [#10 >> #14] Olivine, 0.609 gram					
100	44 ± 4	27.1 ± 0.3	6.99 ± 0.70	697 ± 6	1.6 ± 0.2
1000	22 ± 2	26.7 ± 0.3	1.27 ± 0.13	1390 ± 18	1.6 ± 0.2
1500	11 ± 1	27.1 ± 0.4	0.28 ± 0.03	2531 ± 126	1.8 ± 0.3
Total	77 ± 5	27.0 ± 0.2	8.53 ± 0.71	860 ± 18	1.6 ± 0.2
696-6B [>#10] Olivine, 0.230 gram					
100	68 ± 7	27.4 ± 0.4	6.05 ± 0.61	1089 ± 10	1.4 ± 0.2
1000	21 ± 2	26.8 ± 0.6	1.11 ± 0.12	1526 ± 47	1.5 ± 0.2
1500	15 ± 2	27.5 ± 0.5	0.51 ± 0.07	2092 ± 146	1.7 ± 0.3
Total	104 ± 7	27.3 ± 0.3	7.66 ± 0.62	1218 ± 20	1.5 ± 0.2

ples (Honda *et al.*, 1991), enabling the determination of ²¹Ne/²²Ne ratios of mantle reservoirs. However, presence of three stable isotopes of argon (³⁶Ar, ³⁸Ar and ⁴⁰Ar) is not as advantageous as the case of neon, because the occurrence in the mantle of ³⁸Ar/³⁶Ar ratio being different from the atmospheric value has been anticipated (Pepin, 1998), but it has yet to be proven. Thus, it is difficult to determine ⁴⁰Ar/³⁶Ar ratios of the mantle reservoirs without ambiguity. Likewise, apart from a rather consolidated

occurrence of the isotopically non-atmospheric xenon component with excesses in ^{129,131-136}Xe in mantle source region of MORBs, isotope ratios of xenon associated with the high ³He/⁴He-mantle plumes are still a matter of debate (e.g., Trieloff and Kunz, 2005). Identification of ⁴⁰Ar/³⁶Ar and ^{129,131-136}Xe/¹³⁰Xe ratios indigenous to mantle plumes should provide important constraints on models of early and long-term history of earth's differentiation processes (e.g., Porcelli and Wasserburg, 1995; Kamijo

et al., 1998; Seta *et al.*, 2001; Tolstikhin and Hofmann, 2005), but so far precise heavy noble gas isotopic ratios are not well constrained because of the difficulty in estimating the contribution from the ubiquitous atmospheric component in any analyses. A recent idea that atmospheric heavier noble gas can be recycled into the mantle (e.g., Sarda *et al.*, 1999; Matsumoto *et al.*, 2001, 2002a, 2005; Matsumoto, 2006; Holland and Ballentine, 2006) necessitates additional consideration when assessing the atmospheric contamination to samples (Matsumoto, 2006).

One possible approach to assess pristine mantle $^{40}\text{Ar}/^{36}\text{Ar}$ ratio is to extract and analyze noble gases from mantle-derived samples by a multi-step crushing; because it would yield elemental and isotope ratios showing coherent and simple mixing trend if only two components were involved (i.e., air and mantle) (e.g., Hopp and Trieloff, 2005; Trieloff *et al.*, 2000; Moreira *et al.*, 1998). This approach had successfully been applied for constraining noble gas isotope composition of the MORB source mantle (Moreira *et al.*, 1998). For samples derived from the hotspots, only a few reports of high-resolution stepwise crushing are available (two from Loihi dunites and from Icelandic glasses by Trieloff *et al.*, 2000). Here we report our new helium and argon data obtained by high-resolution stepwise heating (six steps) on basaltic glass and olivine separated from relatively large pillow basalts. The purpose of this paper is to demonstrate possibility to utilize argon isotope information to discuss isotope systematics of noble gases by estimating $^{40}\text{Ar}/^{36}\text{Ar}$ ratios in a less crude fashion based on data taken by the multiple step-crushing from our new samples from Loihi seamount and those available in the literature.

SAMPLES AND ANALYSIS

The basaltic glass and olivine separates used for noble gas analyses in this study were collected from Loihi seamount by a submersible dive (SHINKAI 6500) during the 2002 JAMSTEC Hawaii cruises (YK02-05 Leg 1B). Two sampling dives (S696 and S698) had been targeted to steep eastern and western foothills of Loihi where some interiors of the volcano (thus of magma erupted earlier stages) are expected to be exposed by masswasting. We have collected more than 20 pillow basalts from depths range from 2500 to 4100 meters. In this paper, we report results we taken from two particular pillow lava samples (698-4B from 18°54.7' N, 155°12.6' W, 2815 m depth; 696-6B from 18°48.6' N, 155°14.2' W, 3883 m depth). The major element compositions for the whole rock are shown in the Appendix. The former sample is sparsely olivine phyric basalt with relatively thick quenched glasses and the latter one is picrobasalt. The both compositions are potted within those of alkalic to transitional lavas in Loihi reported by Garcia *et al.* (1995). Glassy

rinds had been carefully chipped off from rock samples. Only fresh and homogeneous glasses were handpicked under a binocular microscope. Olivine phenocrysts were prepared by handpicking. Because there were enough olivine separates were available for preparing several size fractions, we sieved obtained olivine phenocrysts into five different sizes and subjected for separate analyses. These samples had been cleaned up in an ultrasonic bath with demi-water, acetone, and ethanol at least for 30 minutes each. After drying them in an oven at temperature >100°C, samples were weighted and loaded into the crushing apparatus.

Noble gas analyses by step-crushing had been carried out with VG5400 noble gas mass spectrometer and dedicated gas extraction and purification system. The type of crusher used and gas handling, purification procedures are the same as those described in our previous publications (e.g., Wada and Matsuda, 1998; Matsumoto *et al.*, 2001). Sensitivity of noble gases and isotope discrimination factors are calibrated against HESJ standard for helium (Matsuda *et al.*, 2002) and atmospheric gases for other heavier noble gases.

This paper will focus on helium and argon isotope ratios we obtained from 696-6B and 698-4B (Table 1). Results of neon and xenon isotope analysis from these samples as well as a full set of noble gas data from other samples we collected during the 2002 cruise from Loihi will be presented in a separate publication.

RESULTS AND DISCUSSION

He and Ar in Loihi Glass and olivine

698-4B glass: This sample yielded $^3\text{He}/^4\text{He}$ ratios between 34.3 to 37.1 Ra in fractions released during stepwise crushing. These ratios agreed each other within respective two sigma uncertainty levels. The total $^3\text{He}/^4\text{He}$ ratio of 35.3 ± 0.3 coincides with the highest ratio previously reported for Loihi basalts by Valbracht *et al.* (1997). As far as helium isotope is concern, the 698-4B can be regarded as a sample with the most primitive signature among available Loihi samples. $^{40}\text{Ar}/^{36}\text{Ar}$ ratio is, however, significantly lower than the highest ratio of 8000 found in the dunites from Loihi and glasses of 6300 (Valbracht *et al.*, 1997) and 6600 (Trieloff *et al.*, 2003). Thus, this sample have most primitive helium signature but at the same time significantly more affected by an addition of an atmospheric component, if $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of the Loihi source is 6000~8000. This issue will be addressed further in the following sections.

696-6B glass and olivine: 696-6B glass and olivine phenocrysts yielded consistent $^3\text{He}/^4\text{He}$ ratios of about 27 Ra, indicating glass and olivine both acquired the same helium component. $^{40}\text{Ar}/^{36}\text{Ar}$ ratios appeared to be low in glass (~370) in contrast to relatively higher ratios found

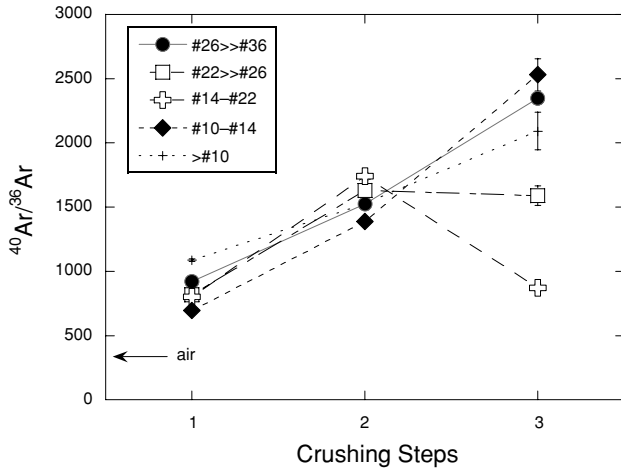


Fig. 1. $^{40}\text{Ar}/^{36}\text{Ar}$ ratios measured during three-step crushing on several size fractions of olivine separated from Loihi picrite (696-6B). Atmospheric ratio of 295.5 is indicated. In all cases, $^{40}\text{Ar}/^{36}\text{Ar}$ ratio observed in the first crushing step is closer to the atmospheric ratio compared to subsequent steps.

in the latter crushing steps on the olivine phenocrysts (Fig. 1). There is a general trend found all olivine fractions that $^{40}\text{Ar}/^{36}\text{Ar}$ ratios measured for argon released at the first crushing steps are lower than those of second and third fractions. Similar trend can also be found for the case of 698-4B glass especially for the first 4 crushing steps. This is a commonly found feature of stepcrushing on basaltic glasses and phenocrysts and was attributed to entrapment of atmospheric gas in microfractures created and resealed during sample preparations (Ballentine and Barfod, 2000). Although we performed grain size separation for olivine phenocrysts to see if there is systematic variation in elemental and isotope compositions, it appeared that the size of grains only affects the gas concentrations (Table 1). Smaller grains in this sample are likely to have lost their vesicles by pulverization of larger grains.

$^3\text{He}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ correlation

As noted earlier, isotope ratios measured for multiple fractions released from a single specimen often display clear mixing trends. If a pair of isotope ratios with a common denominator is chosen for axis of a three-isotope diagram, these ratios form a mixing line extending from one endmember to another. Figure 2a and b plot $^3\text{He}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios measured from six steps from sample 698-4B glass and 696-6B olivine. For the olivine, results of all five samples are included, as there seems no systematic variation found in elemental and isotope ratios among different grain sizes. As shown in Fig. 2, both samples define clear linear trends both extending from a component with low $^{40}\text{Ar}/^{36}\text{Ar}$ ratio which must be greatly

influenced by air-Ar ($^{40}\text{Ar}/^{36}\text{Ar} = 295.5$), but the slopes of these two correlation lines are clearly different each other.

Elemental fractionation between helium and argon cannot be a viable explanation for apparently different slopes defined by these samples. Ratio of radiogenic isotopes of helium and argon can be used as indicator of recent elemental fractionation. Unfractionated value expected from mantle K, U, Th composition is estimated to be 1.7 for an accumulation time of 4.5 Ga (Honda and Patterson, 1999). The production ratios may vary by a factor of 2 or 3 because of the variations in the K/U and Th/U ratio and integration times (e.g., shorter integration times lead to higher values up to a maximum of about 5 for the present-day instantaneous production ratio; Honda and Patterson, 1999). It is known that the MORB popping rock show $^4\text{He}/^{40}\text{Ar}^*$ ratios ($^{40}\text{Ar}^*$ is radiogenic ^{40}Ar , corrected for atmospheric argon) consistent with the mantle production ratios (1.2 to 2.8; Staudasher *et al.*, 1989; Javoy and Pineau, 1991). As shown in Table 1, 698-4B glass yielded $^4\text{He}/^{40}\text{Ar}^*$ ratios of about 4 which is within the range of production ratios (1.7~5) expected for the mantle. The $^4\text{He}/^{40}\text{Ar}^*$ ratios of 1.0~1.5 found in the olivine samples (696-6B) might be somewhat low compared with the production ratio, as commonly found in the crystalline phases (Honda and Patterson, 1999). Several models predict that different solubility of helium and argon in erupting magma would result in significant change in He/Ar ratio by a factor of ten or more through the formation and loss of vesicles (e.g., Honda and Patterson, 1999; Sarda and Guillot, 2005; Yamamoto and Burnard, 2005). In this respect, the degree of fractionation possibly affected the olivine samples is clearly small and will not critically affect the following discussion. Therefore, helium and argon are safely regarded not to be fractionated each other to a significant extent in 698-4B glass and 696-6B olivine, suggesting that the correlation lines shown in Figs. 2a and b are most likely to be the binary mixing lines between an atmospheric component as a common endmember and mantle endmembers having different helium-argon isotope compositions.

Estimation of indigenous $^{40}\text{Ar}/^{36}\text{Ar}$ ratios based on the multi-step crushing data

To further utilize the well-defined mixing trends between $^3\text{He}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios measured by multi-step crushing, attempt will be made to constrain $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of each endmember by assuming $^3\text{He}/^{36}\text{Ar}$ ratios of the mantle. As for the $^3\text{He}/^{36}\text{Ar}$ ratios of the mantle, it has been suggested that they are considered to be uniform in the whole mantle scale (e.g., Honda *et al.*, 1991; Ozima and Zahnle, 1993; Porcelli and Wasserburg, 1995; Moreira *et al.*, 1998; Sasaki, 1999; Trierloff and Kunz, 2005). As shown in Fig. 3a, $^3\text{He}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ar-

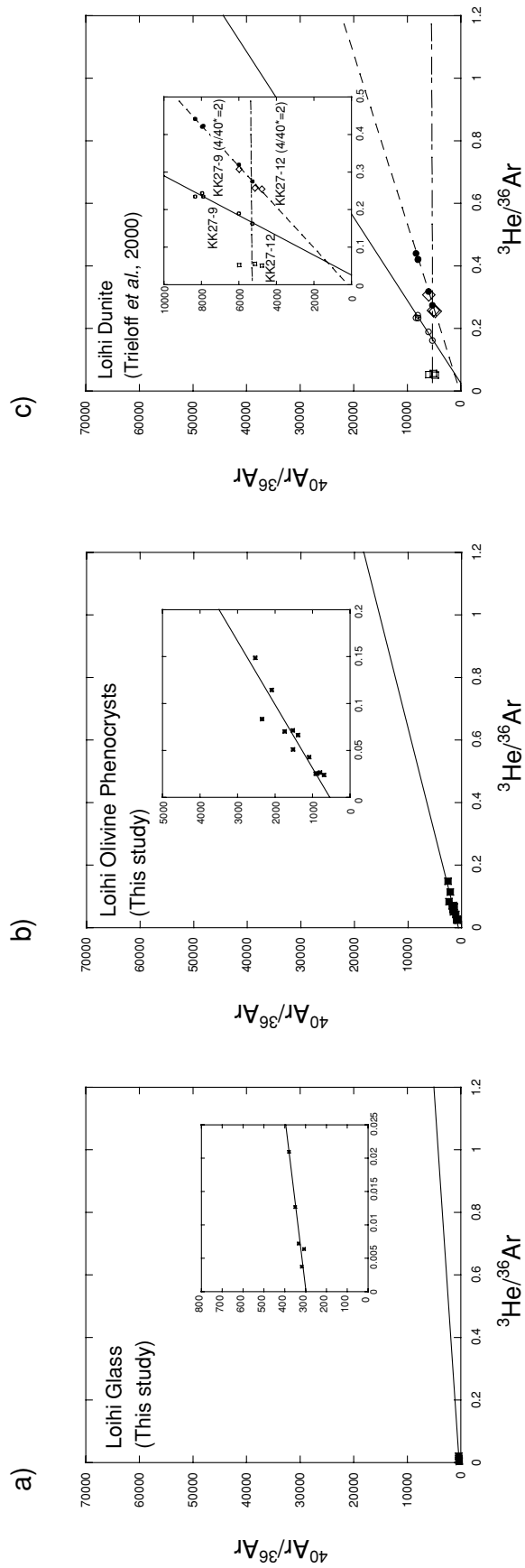


Fig. 2. $^3\text{He}/^{36}\text{Ar}$ versus $^{40}\text{Ar}/^{36}\text{Ar}$ ratios measured by sequential step crushing for a) Loihi glass (698-4B) and b) olivine separates from Loihi pillow basalt (696-6B). Data from Loihi Dunites (KK27-9 and 27-12) are also shown in c). A regression line labeled as $(4/40^* = 2)$ is that determined for data corrected for helium-argon elemental fractionation and adjusted for assumed unfractiionated $^4\text{He}/^{40}\text{Ar}^*$ of 2 (see text for more details).

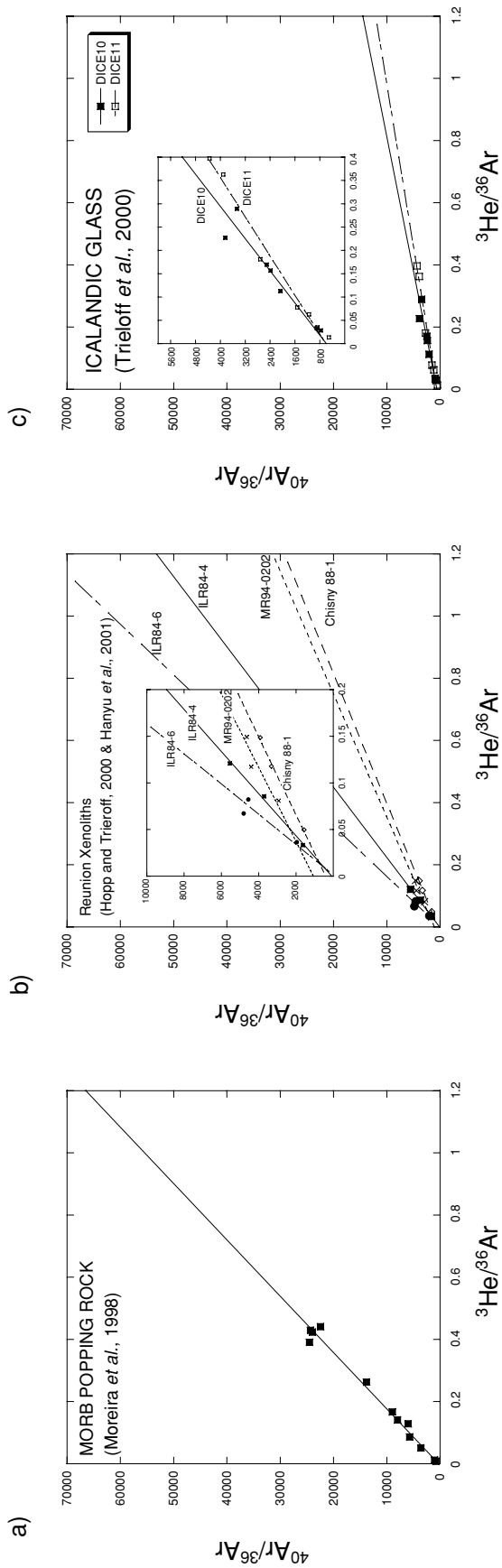


Fig. 3. $^3\text{He}/^{36}\text{Ar}$ versus $^{40}\text{Ar}/^{36}\text{Ar}$ ratios measured by sequential step crushing (>3 steps) for a) MORB popping rock by Moreira et al. (1998), b) Reunion Xenoliths (Hopp and Trieroff, 2005; Hanyu et al., 2001) and c) Icelandic glasses (Trieroff et al., 2000). Simple linear regression was applied to all suites and their Y-intercepts and slopes of regression with standard errors are followings: MORB popping rock: 246 ± 602 and 55200 ± 2300 , Reunion xenoliths: 89 ± 279 and 44300 ± 3200 (ILR84-4), -74 ± 1730 and 61700 ± 26600 (ILR84-6), 411 ± 168 and 24000 ± 1500 (Chisny 88-1), 1065 ± 1149 and 25300 ± 9700 (MR94-0202), Icelandic glass: 593 ± 263 and 11600 ± 1500 (Dice 10), 608 ± 127 and 9500 ± 600 (Dice 11). Atmospheric $^3\text{He}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios are 2.3×10^{-7} and 295.5 , respectively.

tios in vesicles in the volatile-rich basaltic glass “popping rock” yields a very good linear trend extending from atmospheric composition. This sample is known to have unfractionated $^4\text{He}/^{40}\text{Ar}^*$ ratio, thus its $^3\text{He}/^{36}\text{Ar}$ ratio would be a good representative of the mantle. $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of the popping rock is estimated to be about 44000 based on its He–Ne–Ar isotope systematics (Moreira *et al.*, 1998). $^3\text{He}/^{36}\text{Ar}$ ratio of the MORB source constrained from $^{40}\text{Ar}/^{36}\text{Ar} = 44000$ is about 0.7 (Moreira *et al.*, 1998). Note that Moreira’s $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of 44000 roughly agrees with the highest measured $^{40}\text{Ar}/^{36}\text{Ar}$ of 64000 (+8000/–15000) by laser extraction on vesicles in the popping rock (Burnard *et al.*, 1997). The ratio also agrees with equally high $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of 42000 ± 10000 for a different MORB sample reported in Marty and Humbert (1997). Thus, we expediently assumed the mantle $^3\text{He}/^{36}\text{Ar}$ ratio to be 0.7 for the following discussion, as this would be a good compromise among the various estimates.

With the assumption that a whole mantle shares the same primordial $^3\text{He}/^{36}\text{Ar}$ ratio of 0.7 and with the series of data taken from a single specimen by multiple stepwise crushing, it is possible to estimate the $^{40}\text{Ar}/^{36}\text{Ar}$ ratio without the contribution from atmospheric argon. In the case of two Loihi samples reported above, $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of the 698-4B glass is about 3000 whereas the olivine needs to have much higher ratio of 11000. Note that the endmember ratio suggested for the olivine separates are significantly higher than that inferred from the 698-4B glass and than any previous estimate of $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of the Hawaiian plume source which vary from 2000 (based on Kilauea and Loihi basalts, Valbracht *et al.*, 1997; Hiyagon *et al.*, 1992) to 8000 (found in Loihi dunites, Trieloff *et al.*, 2000).

Although the two Loihi dunites by Trieloff *et al.* (2000) were measured by multistep crushing extraction, the $^{40}\text{Ar}/^{36}\text{Ar}$ and $^3\text{He}/^{36}\text{Ar}$ ratios of those Loihi dunite samples (KK27-9 and KK27-12) did not form clear mixing trends extending from air. In these samples, the expected mixing trends had most likely been disturbed by some recent elemental fractionation between helium and argon and its effect had apparently been not uniform because $^4\text{He}/^{40}\text{Ar}^*$ ratios vary by a factor of 5 in KK27 samples. If $^3\text{He}/^{36}\text{Ar}$ ratios are corrected for this elemental fractionation by assuming unfractionated $^4\text{He}/^{40}\text{Ar}^*$, then we observe a clear linear trend extending from air–Ar as shown in Fig. 2c. Thus, at least we can regard argon in these Loihi dunites are a mixture of air and mantle components. However, because the extrapolated $^{40}\text{Ar}/^{36}\text{Ar}$ ratios vary depending on the choice of unfractionated $^4\text{He}/^{40}\text{Ar}^*$ which is difficult to speculate upon (for example, the extrapolated $^{40}\text{Ar}/^{36}\text{Ar} = \sim 9000$ at $^4\text{He}/^{40}\text{Ar}^* = 3$ whereas ~ 17000 at $^4\text{He}/^{40}\text{Ar}^* = 1.5$), further attempt to determine their $^{40}\text{Ar}/^{36}\text{Ar}$ ratio without atmospheric contribution is not

possible. Nonetheless, it is important to note that the nominal $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of 8000 found in the Loihi dunite might better to be regarded as the lower limit for the trapped mantle component.

In Fig. 3, we also plot $^3\text{He}/^{36}\text{Ar}$ ratios versus $^{40}\text{Ar}/^{36}\text{Ar}$ ratios measured by more than three stepwise crushing on samples from Iceland (2 glass samples from Trieloff *et al.*, 2000) and Reunion (1 xenolith by Hanyu *et al.*, 2001; 3 xenoliths by Hopp and Trieloff, 2005). The ratios from these samples form relatively good correlation lines extending from air-like $^{40}\text{Ar}/^{36}\text{Ar}$ ratios as expected from their consistent and rather uniform $^4\text{He}/^{40}\text{Ar}^*$ ratios. Extrapolated $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of four xenoliths from Reunion appeared to be considerably variable (17000–43000) and consistently higher than those by Loihi and Iceland. This is in general agreement with He and Ne isotope signatures found in those samples (e.g., Hanyu *et al.*, 2001; Hopp and Trieloff, 2005). There seems a systematic variation also exist in $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of mantle plumes that has barely been noticeable from the nominal isotope ratios with variable atmospheric overprints.

Comparison of $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ corrected for air contamination: Implications to the plume $^{40}\text{Ar}/^{36}\text{Ar}$ ratios

Hereafter, we will discuss the variation in the extrapolated $^{40}\text{Ar}/^{36}\text{Ar}$ ratios found in samples from different mantle plumes in conjunction with their $^3\text{He}/^4\text{He}$ ratios. As shown in Fig. 4, there is an apparent co-variation found between helium isotope ratio and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios corrected for the atmospheric argon with compositions of the popping rock and the Loihi glass at the both ends. This trend could be explained by (1) a presence of distinct mantle sources with different time-integrated $^{40}\text{K}/^{36}\text{Ar}$ and $(\text{U} + \text{Th})/^3\text{He}$ ratios or (2) a binary mixing between a MORB component and a plume component. Note that in this diagram, a binary mixing curve becomes a line if both endmembers have the same $^3\text{He}/^{36}\text{Ar}$ ratios.

The first scenario could explain grouping of data in Fig. 4 in which $^3\text{He}/^4\text{He}$ and air-corrected $^{40}\text{Ar}/^{36}\text{Ar}$ ratios show some restricted ranges for each locality. The presence of discrete plume sources was suggested from He–Ne systematics of plume-derived samples (Hanyu *et al.*, 2001). However, this scenario fails to account for the variation existing among the samples from the single locality, because it is difficult to envisage the significantly different time-integrated $^{40}\text{K}/^{36}\text{Ar}$ and $(\text{U} + \text{Th})/^3\text{He}$ ratios and heterogeneous $^3\text{He}/^{36}\text{Ar}$ ratios developed within a single mantle source. Thus, the mixing scenario seems to be more plausible for explaining the local co-variation between $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios. Samples from Hawaiian volcanoes are known to show a similar mixing trend with increasing contribution of MORB type He towards the older members of the volcanic chain (Kaneoka

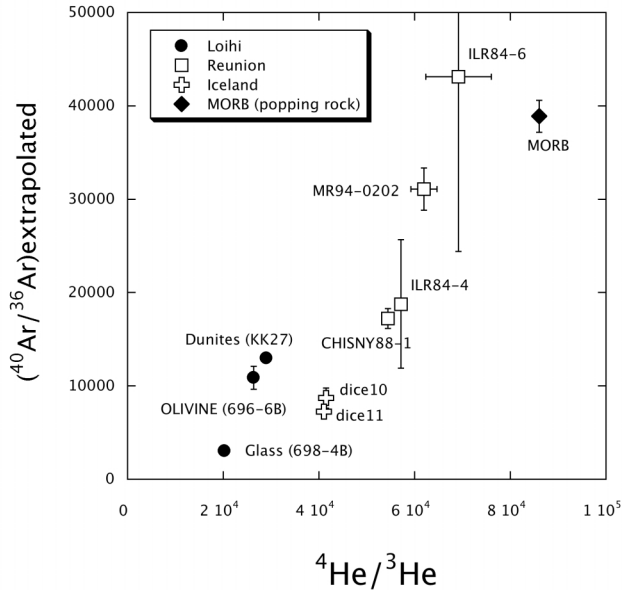


Fig. 4. Comparison between helium isotope ratio ($^4\text{He}/^3\text{He}$) and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios corrected for atmospheric contamination based on the stepcrushing results and an assumption of $^3\text{He}/^{36}\text{Ar} = 1$ for the mantle. For Loihi Dunites in which the extrapolation was not possible without correcting elemental fractionation between helium and argon, we assumed $^4\text{He}/^{40}\text{Ar}^* = 2$. A simple linear regression of all data points in this diagram passes through $^{40}\text{Ar}/^{36}\text{Ar} \sim 1000$ at $^3\text{He}/^4\text{He} = 50$ Ra ($^4\text{He}/^3\text{He} = 1.4 \times 10^5$).

et al., 2002). The present result demonstrates that the magma appeared to have been influenced by noble gases from the asthenospheric component even for the samples from the present hotspot center (Loihi). Indeed, by its definition, the extrapolated $^{40}\text{Ar}/^{36}\text{Ar}$ ratios warrant the absence of atmospheric influence, but this does not exclude the possibility that the magma had been a mixture of components with different $^{40}\text{Ar}/^{36}\text{Ar}$ ratios.

As we noted, the extrapolated $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of Loihi basaltic glass appeared to be about 3000. This value is somewhat lower compared with the value of about 8000 estimated based on the analysis of Loihi dunites. With the first clear identification of non-atmospheric xenon isotope signatures in these Loihi dunites, their isotope ratios are regarded as fairly robust representatives for the mantle source for Loihi (Harrison *et al.*, 2003; Torieloff and Kunz, 2005). Note that we made our estimate on the sample with the highest $^3\text{He}/^4\text{He}$ ratio of 35 Ra so far reported for Loihi samples, while $^3\text{He}/^4\text{He}$ ratio of Loihi dunites are about 24 Ra. As shown in Fig. 4, this difference in $^3\text{He}/^4\text{He}$ ratios is consistent with an increased contribution of the MORB-type component to the noble gases trapped in those dunites.

We also emphasize that $^{40}\text{Ar}/^{36}\text{Ar}$ ratios estimated above and observed $^4\text{He}/^3\text{He}$ ratios should be regarded as an upper limit for the source of Loihi magma because we cannot rule out the possible contribution from the MORB-type component even in the most primitive sample (698-4B). There are in fact some reports of much higher $^3\text{He}/^4\text{He}$ ratios from the plume-related materials. For example, the $^3\text{He}/^4\text{He}$ ratios in Baffine Island picrites are reported to be as high as 50 Ra (Stuart *et al.*, 2003). If such ratio is projected onto the apparent mantle array defined between $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ (Fig. 4), then the corresponding $^{40}\text{Ar}/^{36}\text{Ar}$ ratio appears to be about 1000. A mantle reservoir with even higher $^3\text{He}/^4\text{He}$ ratio is predicted to exist based on the observed significantly higher $^3\text{He}/^4\text{He}$ ratio of >70 Ra in the Archean komatiites (Seta *et al.*, 2001; Matsumoto *et al.*, 2002b), suggesting that the mantle plume might be derived from a reservoir with $^{40}\text{Ar}/^{36}\text{Ar}$ ratio smaller than 1000. Previously proposed $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of the mantle source for high $^3\text{He}/^4\text{He}$ OIBs range from 5000 to 8000, which are significantly higher than our estimate presented above. Note that low (and near atmospheric) $^{40}\text{Ar}/^{36}\text{Ar}$ ratios were indeed reported in early noble gas studies on OIBs including those from Hawaii (e.g., Allègre *et al.*, 1987). These have subsequently been demonstrated to be the result of atmospheric contamination (Patterson *et al.*, 1990). We have to emphasize that our estimate is based on OIB-MORB datasets corrected for the air contamination by utilizing stepwise crushing gas extraction, so we believe we made a robust case for a high $^3\text{He}/^4\text{He}$ mantle source with a significantly less radiogenic argon isotope composition. Although we have to admit our scheme still contains an important assumption (i.e., a uniform $^3\text{He}/^{36}\text{Ar}$ ratio in the mantle) that needs to be justified, we can draw some important implications about mantle noble gas systematics even within the framework of this assumption.

Implications of low $^{40}\text{Ar}/^{36}\text{Ar}$ ratio in high $^3\text{He}/^4\text{He}$ source

We have shown above that a plume source should have a $^{40}\text{Ar}/^{36}\text{Ar}$ ratio being significantly less radiogenic than has usually been anticipated. In this respect, there is also a report of $^{21}\text{Ne}/^{22}\text{Ne}$ ratios not modified by nucleogenic ^{21}Ne (produced by a nuclear reaction of oxygen with alpha-particles emitted by local U and Th decay) in some Icelandic glass samples (Dixon *et al.*, 2000). Such a source should either be undegassed of the primordial noble gases or isolated from radioactive parents over the most of earth's history. To accommodate with a view that the whole mantle has been well processed over 4.5 Ga since accretion, the preservation of primordial noble gases into the core would be an attractive alternative (Davis, 1990; Macpherson *et al.*, 1998; Tolstikhin and Marty, 1998; Porcelli and Halliday, 2001; Lee and Steinle-Neumann, 2006). But this scenario also requires that some radioac-

tive parents (such as U, Th, K, Pu and I) need not to be partitioned into the core to significant extents, while sufficiently large amount of noble gases were to be partitioned into the core. However, experimentally determined partition coefficients of noble gases between the metal and silicates are very small (Matsuda *et al.*, 1993), so the suggestion that the core is a source of a primordial (isotopically less radiogenic) noble gas component remains highly speculative.

Another hypothesis that can potentially explain the low $^{40}\text{Ar}/^{36}\text{Ar}$ ratio in highly processed mantle would be the recycling of atmospheric noble gases into the mantle by subduction. So far, an existence of the recycled atmospheric noble gas component is recognized in samples from the subcontinental and subarc lithospheric mantle (Matsumoto *et al.*, 2001, 2005; Yamamoto *et al.*, 2004), in basalts from Mid-Atlantic Ridge (Sarda *et al.*, 1999), and in some continental well gases (Holland and Ballentine, 2006). These samples are from relatively shallow part of the mantle, and there has been no clear evidence directly supports the recycled atmospheric noble gases in the source of mantle plumes. We showed that the plume source is expected to have high $^3\text{He}/^4\text{He}$ ratio ($>50 Ra$) and low $^{40}\text{Ar}/^{36}\text{Ar}$ ratio (<1000). Such a combination is expected for the mantle domain that affected by the recycled component having significant air-Ar without associated helium (Matsumoto *et al.*, 2001; 2005). The radiogenic isotope geochemistry of OIB materials suggest that they are derived from mantle domains variably affected by subducted crustal materials and their sediment cover (e.g., Hofmann, 1997). In this respect, we note that the recycling of atmospheric noble gases can accommodate well with other geochemical observations. This recycling hypothesis has significant implications, as this requires extensive revision as to how the noble gas reservoirs had been evolved. However, as a feasibility of noble gas recycling into the deep mantle region has not been well explored yet, further clarification is required to further understand the origin of the less radiogenic noble gas component in the mantle source region of the high $^3\text{He}/^4\text{He}$ plumes.

CONCLUDING REMARKS

As discussed above, we demonstrated that $^{40}\text{Ar}/^{36}\text{Ar}$ ratios can also equally be useful like helium and neon isotopes if we can systematically treat the effect of air-contamination based on the analyses by multiple stepcrushing. Practically, $^{40}\text{Ar}/^{36}\text{Ar}$ ratios are relatively easy to analyze compared with $^{21}\text{Ne}/^{22}\text{Ne}$ ratios, and information obtained by argon isotopes could be useful to infer the behavior of less abundant heavier noble gases (Kr and Xe). In this respect, we have to note that non-atmospheric xenon signature found in some plume-related

samples, such as in Loihi dunite (Trieloff *et al.*, 2000) and in Samoan xenoliths (Poreda and Farley, 1992) might also be derived from the asthenospheric mantle component. It was unfortunate that the present samples did not yield sufficient amounts of neon and xenon for precise isotope analyses. If there were appropriate samples with abundant neon and xenon with extremely high $^3\text{He}/^4\text{He}$ ratios, multiple stepcrushing on such samples would provide further constraints on the heavy noble gas compositions in the primitive reservoirs.

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REFERENCES

- Allègre, C. J., Staudacher, T. and Sarda, P. (1987) Rare gas systematics: formation of the atmosphere, evolution and structure of the Earth's mantle. *Earth Planet. Sci. Lett.* **81**, 127–150.
- Ballentine, C. J. and Barfod, D. N. (2000) The origin of air-like noble gases in MORB and OIB. *Earth Planet. Sci. Lett.* **180**, 39–48.
- Burnard, P., Graham, D. and Turner, G. (1997) Vesicle-specific noble gas analyses of “popping rock”: Implications for primordial noble gases in earth. *Science* **276**, 568–571.
- Davies, G. F. (1990) Mantle plumes, mantle stirring and hotspot chemistry. *Earth Planet. Sci. Lett.* **99**, 94–109.
- Dixon, E. T., Honda, M., McDougall, I., Campbell, I. H. and Sigurdsson, I. (2000) Preservation of near-solar neon isotopic ratios in Icelandic basalts. *Earth Planet. Sci. Lett.* **180**, 309–324.
- Garcia, M. O., Foss, D. P., West, H. B. and Mahoney, J. J. (1995) Geochemical and isotopic evolution of Loihi volcano, Hawaii. *J. Petrol.* **36**, 1647–1674.
- Hanyu, T. and Kaneoka, I. (1997) The uniform and low $^3\text{He}/^4\text{He}$ ratios of HIMU basalts as evidence for their origin as recycled materials. *Nature* **390**, 273–276.
- Hanyu, T., Dunai, T. J., Davies, G. R., Kaneoka, I., Nohda, S. and Uto, K. (2001) Noble gas study of the Reunion hotspot: evidence for distinct less-degassed mantle sources. *Earth Planet. Sci. Lett.* **193**, 83–98.
- Harrison, D., Burnard, P. G., Trieloff, M. and Turner, G. (2003) Resolving atmospheric contaminants in mantle noble gas analyses (vol. 4, art no. 1023, 2003). *Geochem. Geophys. Geosy.* **4**.
- Hiyagon, H., Ozima, M., Marty, B., Zashu, S. and Sakai, H. (1992) Noble-gases in submarine glasses from midoceanic ridges and Loihi seamount—Constraints on the early history of the Earth. *Geochim. Cosmochim. Acta* **56**, 1301–1316.

- Hofmann, A. W. (1997) Mantle geochemistry: the message from oceanic volcanism. *Nature* **385**, 219–229.
- Holland, G. and Ballentine, C. J. (2006) Seawater subduction controls the heavy noble gas composition of the mantle. *Nature* **441**, 186–191.
- Honda, M. and Patterson, D. B. (1999) Systematic elemental fractionation of mantle-derived helium, neon, and argon in mid-oceanic ridge glasses. *Geochim. Cosmochim. Acta* **63**, 2863–2874.
- Honda, M., McDougall, I., Patterson, D. B., Dougeris, A. and Clague, D. A. (1991) Possible solar noble-gas component in Hawaiian basalts. *Nature* **349**, 149–151.
- Hopp, J. and Trieloff, M. (2005) Refining the noble gas record of the Reunion mantle plume source: Implications on mantle geochemistry. *Earth Planet. Sci. Lett.* **240**, 573–588.
- Javoy, M. and Pineau, F. (1991) The volatiles record of a “popping” rock from the Mid-Atlantic Ridge at 14°N—Chemical and isotopic composition of gas trapped in the vesicles. *Earth Planet. Sci. Lett.* **107**, 598–611.
- Kamijo, K., Hashizume, K. and Matsuda, J. (1998) Noble gas constraints on the evolution of the atmosphere-mantle system. *Geochim. Cosmochim. Acta* **62**, 2311–2321.
- Kaneoka, I., Hanyu, T., Yamamoto, J. and Miura, Y. N. (2002) Noble gas systematics of the Hawaiian Volcanoes based on the analysis of Loihi, Kilauea and Koolau submarine rocks. *Hawaiian Volcanoes: Deep Underwater Perspectives, Geophysical Monograph 128* (Takahashi, E. *et al.*, eds.), American Geophysical Union.
- Lee, K. K. M. and Steinle-Neumann, G. (2006) High-pressure alloying of iron and xenon: “Missing” Xe in the Earth’s core? *J. Geophys. Res.-Sol. Ea.* **111**, B02202.
- Macpherson, C. G., Hilton, D. R., Sinton, J. M., Poreda, R. J. and Craig, H. (1998) High $^3\text{He}/^4\text{He}$ ratios in the Manus backarc basin: Implications for mantle mixing and the origin of plumes in the western Pacific Ocean. *Geology* **26**, 1007–1010.
- Marty, B. and Humbert, F. (1997) Nitrogen and argon isotopes in oceanic basalts. *Earth Planet. Sci. Lett.* **152**, 101–112.
- Matsuda, J., Sudo, M., Ozima, M., Ito, K., Ohtaka, O. and Ito, E. (1993) Noble-gas partitioning between metal and silicate under high-pressures. *Science* **259**, 788–790.
- Matsuda, J., Matsumoto, T., Sumino, H., Nagao, K., Yammaoto, J., Miura, Y., Kaneoka, I., Takahata, N. and Sano, Y. (2002) The $^3\text{He}/^4\text{He}$ ratio of the new internal He Standard of Japan (HESJ). *Geochem. J.* **36**, 191–195.
- Matsumoto, T. (2006) The noble art of recycling. *Nature* **441**, 169–170.
- Matsumoto, T., Chen, Y. L. and Matsuda, J. (2001) Concomitant occurrence of primordial and recycled noble gases in the Earth’s mantle. *Earth Planet. Sci. Lett.* **185**, 35–47.
- Matsumoto, T., Pinti, D. L., Matsuda, J. and Umino, S. (2002a) Recycled noble gas and nitrogen in the subcontinental lithospheric mantle: Implications from N–He–Ar in fluid inclusions of SE Australian xenoliths. *Geochem. J.* **36**, 209–217.
- Matsumoto, T., Seta, A., Matsuda, J., Takebe, M., Chen, Y. L. and Arai, S. (2002b) Helium in the Archean komatiites revisited: significantly high $^3\text{He}/^4\text{He}$ ratios revealed by fractional crushing gas extraction. *Earth Planet. Sci. Lett.* **196**, 213–225.
- Matsumoto, T., Morishita, T., Matsuda, J., Fujioka, M., Takebe, K., Yamamoto, K. and Arai, S. (2005) Noble gases in the Finero Phlogopite-Peridotite, Western Italian Alps. *Earth Planet. Sci. Lett.* **238**, 130–145.
- Moreira, M., Kunz, J. and Allègre, C. (1998) Rare gas systematics in popping rock: Isotopic and elemental compositions in the upper mantle. *Science* **279**, 1178–1181.
- Ozima, M. and Zahnle, K. (1993) Mantle degassing and atmospheric evolution—Noble-gas view. *Geochem. J.* **27**, 185–200.
- Patterson, D. B., Honda, M. and McDougall, I. (1990) Atmospheric contamination: a possible source for heavy noble gases in basalts from Loihi seamount, Hawaii. *Geophys. Res. Lett.* **17**, 705–708.
- Pepin, R. O. (1998) Isotopic evidence for a solar argon component in the Earth’s mantle. *Nature* **394**, 664–667.
- Porcelli, D. and Halliday, A. N. (2001) The core as a possible source of mantle helium. *Earth Planet. Sci. Lett.* **192**, 45–56.
- Porcelli, D. and Wasserburg, G. J. (1995) Mass-transfer of helium, neon, argon, and xenon through a steady-state upper-mantle. *Geochim. Cosmochim. Acta* **59**, 4921–4937.
- Poreda, R. J. and Farley, K. A. (1992) Rare gases in Samoan xenoliths. *Earth Planet. Sci. Lett.* **113**, 129–144.
- Sarda, P. and Guillot, B. (2005) Breaking of Henry’s law for noble gas and CO₂ solubility in silicate melt under pressure. *Nature*, **436**, 95–98.
- Sarda, P., Moreira, M. and Staudacher, T. (1999) Argon-lead isotopic correlation in Mid-Atlantic Ridge Basalts. *Science* **283**, 666–668.
- Sasaki, S. (1999) Presence of a primary solar-type atmosphere around the earth: evidence of dissolved noble gas. *Planet. Space Sci.* **47**, 1423–1431.
- Seta, A., Matsumoto, T. and Matsuda, J. (2001) Concurrent evolution of $^3\text{He}/^4\text{He}$ ratio in the Earth’s mantle reservoirs for the first 2 Ga. *Earth Planet. Sci. Lett.* **188**, 211–219.
- Stuart, F. M., Lass-Evans, S., Fitton, J. G. and Ellam, R. M. (2003) High $^3\text{He}/^4\text{He}$ ratios in picritic basalts from Baffin Island and the role of a mixed reservoir in mantle plumes. *Nature* **424**, 57–59.
- Takebe, M. (2001) Rare earth element composition of deep sea sediments from central to western Pacific: Discussion about main factors controlling REE composition of marine sediments by factor analysis. *J. Geol. Soc. Jpn.* **107**, 301–315.
- Takebe, M. and Yamamoto, K. (2003) Geochemical fractionation between porcellanite and host sediment. *J. Geol.*, **111**, 301–312.
- Tolstikhin, I. and Hofmann, A. W. (2005) Early crust on top of the Earth’s core. *Phys. Earth Planet. Int.* **148**, 109–130.
- Tolstikhin, I. N. and Marty, B. (1998) The evolution of terrestrial volatiles: a view from helium, neon, argon and nitrogen isotope modelling. *Chem. Geol.* **147**, 27–52.
- Trieloff, M. and Kunz, J. (2005) Isotope systematics of noble gases in the Earth’s mantle: possible sources of primordial isotopes and implications for mantle structure. *Phys. Earth Planet. Int.* **148**, 13–38.
- Trieloff, M., Kunz, J., Clague, D. A., Harrison, D. and Allègre,

- C. J. (2000) The nature of pristine noble gases in mantle plumes. *Science* **288**, 1036–1038.
- Trieloff, M., Falter, M. and Jessberger, E. K. (2003) The distribution of mantle and atmospheric argon in oceanic basalt glasses. *Geochim. Cosmochim. Acta* **67**, 1229–1245.
- Valbracht, P. J., Staudacher, T., Malahoff, A. and Allègre, C. J. (1997) Noble gas systematics of deep rift zone glasses from Loihi Seamount, Hawaii. *Earth Planet. Sci. Lett.* **150**, 399–411.
- Wada, N. and Matsuda, J. (1998) A noble gas study of cubic diamonds from Zaire: Constraints on their mantle source. *Geochim. Cosmochim. Acta* **62**, 2335–2345.
- Yamamoto, J. and Burnard, P. G. (2005) Solubility controlled noble gas fractionation during magmatic degassing: implications for noble gas compositions of primary melts of OIB and MORB. *Geochim. Cosmochim. Acta* **69**, 727–734.
- Yamamoto, J., Kaneoka, I., Nakai, S., Kagi, H., Prikhod'ko, V. S. and Arai, S. (2004) Evidence for subduction-related components in the subcontinental mantle from low $^3\text{He}/^4\text{He}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratio in mantle xenoliths from Far Eastern Russia. *Chem. Geol.* **207**, 237–259.

APPENDIX

Table A1 lists the major elemental composition of the pillow lava samples that had been determined by XRF at the Nagoya University. These data are obtained following the techniques described in Takebe (2001) and Takebe and Yamamoto (2003). Brief description of the employed method is also found in Matsumoto *et al.* (2002b).

Table A1. Major elemental compositions of the pillow basalts used for this study

	SiO ₂	TiO ₂	Al ₂ O ₃	FeO	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	Total
696-6B	46.98	2.08	10.6	10.98	0.17	14.9	9.67	2.15	0.41	0.20	98.14
698-4B	47.87	2.25	11.72	11.17	0.15	9.16	12.83	2.23	0.49	0.20	98.08