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New constraints on the HIMU mantle from neon and helium isotopic compositions of basalts from the Cook–Austral Islands

Rita Parai a,*, Sujoy Mukhopadhyay a, John C. Lassiter b

- ^a Department of Earth and Planetary Sciences, Harvard University, 20 Oxford Street, Cambridge, MA 02138, USA
- ^b Department of Geological Sciences, University of Texas at Austin, 1 University Station C1100, Austin, TX 78712, USA

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ABSTRACT

High ${}^4\text{He}/{}^3\text{He}$ ratios of 100 000 to 160 000 found at HIMU ocean islands ("high μ ," where μ is the U/Pb ratio) are usually attributed to the presence of recycled oceanic crust in the HIMU mantle source. However, significantly higher ⁴He/³He ratios are expected in recycled crust after residence in the mantle for periods greater than 1 Ga. In order to better understand the helium isotopic signatures in HIMU basalts, we have measured helium and neon isotopic compositions in a suite of geochemically well-characterized basalts from the Cook-Austral Islands. We observe 4He/3He ratios ranging from 56000 to 141000, suggesting the involvement of mantle reservoirs both more and less radiogenic than the mantle source for mid-ocean ridge basalts (MORBs). In addition, we find that the neon isotopic compositions of HIMU lavas extend from the MORB range to compositions less nucleogenic than MORBs. The Cook-Austral HIMU He-Ne isotopic compositions, along with Sr, Nd, Pb, and Os isotopic compositions, indicate that in addition to recycled crust, a relatively undegassed mantle end-member (e.g., FOZO) is involved in the genesis of these basalts. The association of relatively undegassed mantle material with recycled crust provides an explanation for the close geographical association between HIMU lavas and EM (enriched mantle)-type lavas from this island chain: EM-type signatures represent a higher mixing proportion of relatively undegassed mantle material. Mixing between recycled material and relatively undegassed mantle material may be a natural result of entrainment processes and convective stirring in deep mantle.

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1. Introduction

Ocean island volcanism is attributed to the upwelling of mantle plumes from a boundary layer deep within the mantle. Consequently, ocean island basalts (OIBs) serve as chemical probes of the Earth's interior. Highly variable chemical and isotopic compositions observed in OIBs, relative to mid-ocean ridge basalts (MORBs), indicate the existence of distinct chemical reservoirs within the mantle (Zindler and Hart, 1986a; Hart et al., 1992; Farley et al., 1992; Graham, 2002; Porcelli and Ballentine, 2002). Helium and neon are powerful tracers of these mantle sources, and can potentially constrain the origin of chemical heterogeneities in the mantle.

Basalts derived from mantle melting exhibit a large range of helium isotopic compositions. MORBs that are removed from the influence of hotspots exhibit relatively homogeneous ⁴He/³He ratios, ranging from 80 000 to 100 000 (Kurz et al., 1982; Graham et al., 1992b; Graham et al., 1996). The MORB helium isotopic composition reflects a balance between radiogenic production of ⁴He and volatile depletion by degassing and melt extraction from the convective mantle along mid-

ocean ridges. OIBs exhibit greater variation in helium isotopic ratios than MORBs, with $^4\text{He}/^3\text{He}$ ratios ranging from ~15 000 to 200 000 (Kurz et al., 1983; Farley et al., 1992; Hilton et al., 1999; Kurz and Geist, 1999; Graham 2002; Stuart et al., 2003). Low $^4\text{He}/^3\text{He}$ ratios (<80 000) measured at ocean islands such as Hawaii, Iceland, Galapagos, and Samoa are conventionally attributed to the sampling of a relatively undegassed mantle source with low time-integrated (U+Th)/ ^3He (Kurz et al., 1983; Farley et al., 1992; Hilton et al., 1999; Kurz and Geist, 1999; Moreira et al., 1999; Stuart et al., 2003). On the other hand, basalts from HIMU ocean islands ("high μ ," where μ is U/Pb) exhibit $^4\text{He}/^3\text{He}$ ratios higher than the canonical MORB range. For example, HIMU lavas from St. Helena, the Canary Islands, Tubuai, and Mangaia exhibit $^4\text{He}/^3\text{He}$ ratios that vary from 100 000 to 160 000 (Vance et al., 1989; Graham et al., 1992a; Hanyu and Kaneoka, 1997; Barfod et al., 1999; Hanyu et al., 1999; Hilton et al., 2000).

Radiogenic Pb and Os isotopic compositions observed in HIMU basalts suggest that significant quantities of recycled oceanic crust are present in the HIMU source (Hofmann and White, 1982; Zindler et al., 1982; Palacz and Saunders, 1986; Hauri and Hart, 1993). The observed HIMU ⁴He/³He signature also indicates a component that is more radiogenic than the MORB mantle, and therefore appears to be consistent with the hypothesis that the HIMU signature derives from recycled oceanic crust. However, if HIMU lavas are largely derived from

^{*} Corresponding author. Tel.: +1 617 384 9335; fax: +1 617 495 8839. E-mail address: parai@fas.harvard.edu (R. Parai).

recycled oceanic crust, then it is surprising that HIMU 4 He/ 3 He ratios are as *low* as 100 000 to 160 000 (Hanyu et al., 1999; Moreira et al., 1999; Hilton et al., 2000). Recycled oceanic crust should be devoid of 3 He, but should have significant quantities of 4 He produced by radioactive decay of U and Th. As a result, 4 He/ 3 He ratios $> 10^7$ are expected in recycled crust after residence in the mantle for periods greater than 1 Ga.

To explain this apparent discrepancy, Hanyu and Kaneoka (1998) developed an open-system evolution model of helium in recycled materials, in which helium in the subducted oceanic crust diffusively equilibrates with the MORB source (depleted MORB mantle, DMM). However, the model requires high He diffusivities of $\sim 10^{-4} \, {\rm cm}^2/{\rm s}$, which correspond to the diffusivity of He in a basaltic melt and may therefore be unrealistic. Consequently, alternate hypotheses have been proposed to account for the helium isotopic signature of the HIMU component.

Based on He and Ne isotopic measurements in ultramafic xenoliths, basaltic lavas, and carbon dioxide gases from the Cameroon line of volcanoes, Barfod et al. (1999) propose that the radiogenic signatures in HIMU lavas result from closed-system aging of Loihi-like plume-derived melts for 108 years. Hilton et al. (2000) suggest two scenarios, both of which incorporate the involvement of a low ⁴He/³He mantle source to explain HIMU helium and lead signatures measured in basalts from the Canary Islands. In the first scenario, a high ³Hemantle plume, variably depleted in its He content, mixes with recycled crust characterized by radiogenic He and Pb isotopic compositions. The second scenario is their preferred explanation: a composite plume, composed of recycled crust and a low ⁴He/³He component, mixes with DMM to generate ⁴He/³He ratios slightly lower than MORBs. Subsequent addition of radiogenic He from melts frozen in the lithosphere produces ⁴He/³He ratios that are more radiogenic than MORBs and decouples helium from lead isotopic compositions.

Unlike models calling upon the involvement of a low ${}^4\text{He}/{}^3\text{He}$ component in the HIMU source, Moreira and Kurz (2001) argue that helium isotopes in HIMU lavas are dominated by a contribution from recycled oceanic lithosphere. They suggest that ${}^4\text{He}/{}^3\text{He}$ ratios in recycled oceanic lithosphere would evolve to values only slightly more radiogenic than the MORB mantle on timescales of 1 Ga (Moreira and Kurz, 2001).

To evaluate these fundamentally different hypotheses for the generation of HIMU He isotopic signatures, we selected a set of geochemically well-characterized HIMU lavas from the Cook–Austral Islands for helium and neon isotopic analysis. Volcanoes in the Cook–Austral chain erupt both HIMU and EM - type (Enriched Mantle) lavas, and extensive trace element and isotopic data from these islands already exist (Palacz and Saunders, 1986; Nakamura and Tatsumoto, 1988; Hauri and Hart, 1993; Schiano et al., 2001; Lassiter et al., 2003). We demonstrate that the HIMU and EM-type basalts erupted in the Cook–Austral Islands can be generated by the admixture of recycled material and a relatively undegassed mantle component with a depleted mantle component.

2. Samples and analytical methods

Our samples are sub-aerial basaltic lava flows from the islands of Raivavae, Mangaia, Rurutu, Tubuai, and Rapa. The samples were previously characterized for trace elements and radiogenic isotopes (Hauri and Hart 1993; Lassiter et al., 2003). Basalt samples with visible olivine phenocrysts were selected for analysis. Samples were coarsely crushed, and the magnetic groundmass was removed using a hand-held Nd-B magnet. Olivine phenocrysts 500 µm to 2 mm in size were hand-picked and checked with a binocular microscope to ensure that they were free of adhering matrix. In order to remove any surficial alteration, phenocrysts were leached in 1% oxalic acid for 1–1.5 h in a hot water bath (70–80 °C), and then ultrasonically cleaned in distilled water and acetone.

Approximately 170 to 650 mg of olivine were picked for helium isotopic analyses. The olivine grains were loaded into a piston crusher (Fig. 1). The grains were crushed under vacuum using a hydraulic ram

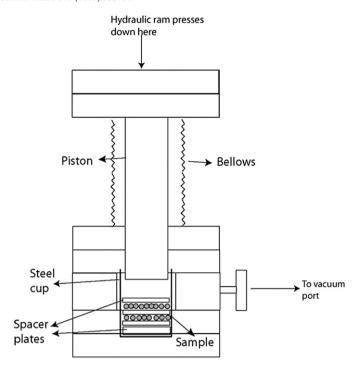


Fig. 1. Schematic of the crusher used in the Harvard laboratory. The olivine grains were sandwiched between two steel plates and crushed. For large samples (>1 g), multiple sandwich layers were used.

to release magmatic gases trapped in fluid and melt inclusions. The total time associated with each crushing step was ≤4 min. The evolved gases were purified by sequential exposure to hot and cold SAES getters, and noble gases were trapped on a cryogenic cold-finger at 12 K. Helium was liberated by heating the cold-finger to 32 K, and was inlet to a Nu Noblesse noble gas mass spectrometer. 4He was measured on a Faraday cup and ³He was measured using an ETP discrete dynode multiplier operating in pulse counting mode. The multiplier used for measuring ³He is located on axis and equipped with an energy filter to reduce scattered ions. Typical crusher blank values were $\sim 1-2 \times 10^{-11}$ cm³ ⁴He STP, or 0.1–1% of the typical measured signal. Helium isotopic ratios and concentrations were determined by normalizing to a standard that was prepared at Harvard (HH3; ${}^{4}\text{He}/{}^{3}\text{He} = 81\,800$). The 1σ variability of standards with helium concentrations similar to that of the samples was 0.7-4% for the ${}^{4}\text{He}/{}^{3}\text{He}$ ratio and $\leq 1\%$ for ${}^{4}\text{He}$ abundance.

Based on sample availability and relatively high helium concentrations, four of the samples were selected for neon isotopic analysis. Aliquots of 1 to 3 g of olivine were hand-picked for the neon measurements, and treated with 1% oxalic acid to remove any surface alteration. After samples were loaded, the crushers were baked out at 125–130 °C for 24–36 h. The crushers were then pumped for an additional 24–36 h to ensure low neon backgrounds. The samples were step-crushed under vacuum. The gases evolved from each step were purified as before and Ne was trapped on the cryogenic cold-finger at 32 K. Ne was released by warming the cold-finger to 74 K and inlet was inlet to mass spectrometer.

The three isotopes of neon were measured in the Nu Noblesse mass spectrometer in multi-collection mode using three discrete dynode multipliers operating in pulse counting mode. ^{21}Ne was measured on the axial multiplier, while ^{20}Ne and ^{22}Ne were measured on the low and high mass multipliers, respectively. Blank values were $<3.5\times10^{-14}~\text{cm}^3$ STP for ^{22}Ne , and blanks were atmospheric in composition. An automated liquid nitrogen trap located close to the ion source was used to keep the Ar and CO₂ backgrounds in the mass spectrometer low. The Ne measurements were made at 200 μA trap current, and to reduce the double charge species of ^{40}Ar and $^{44}\text{CO}_2$, the electron acceleration

voltage in the ion source was kept at 60 V. Ne measurements were corrected for isobaric interferences from doubly-charged Ar and doubly-charged CO₂. The $^{40}\text{Ar}^{+}$ / $^{40}\text{Ar}^{+}$ ratio was 0.065±0.002 and the CO₂*/CO₂ ratio was 0.004±0.001. Variations in these ratios were not observed as a function of Ar, CO₂, or H₂ partial pressure in the mass spectrometer. HF and H₂O are resolved from ^{20}Ne and so no corrections for these species are required. Sample neon isotopic compositions were determined by normalization to an Air standard. The 1σ variation on Air standards similar in size to the samples was 0.2–0.7% for $^{20}\text{Ne}/^{22}\text{Ne}$ ratios and 0.5–1.5% for $^{21}\text{Ne}/^{22}\text{Ne}$ ratios.

3. Results

3.1. Helium isotope systematics

Twenty-three samples were analyzed for helium isotopic composition. The results are summarized in Table 1. We find no strong correlations between He concentration and He isotopic composition (Fig. 2a). Furthermore, we find good correlations between helium and radiogenic isotopic tracers such as Os and Sr, which indicate that the measured He isotopic compositions are not significantly affected by post-eruptive radiogenic or cosmogenic He. Samples from the islands of Raivavae, Mangaia, Tubuai, and old Rurutu exhibit 4 He 3 He ratios that range from 93 200 to 137 000 (3 He 4 He is 5.25 to 7.72 R_A ; Fig. 2b). These values extend from the MORB range to more radiogenic values, and are consistent with previous studies of Cook–Austral volcanism (Hanyu et al., 1999). Samples from the islands of Rapa and young Rurutu have 4 He 3 He ratios ranging from 56 900 to 85 700 (3 He 4 He ranges from 8.40 to 12.6 R_A), which extend from the MORB range to less radiogenic values (Fig. 2b).

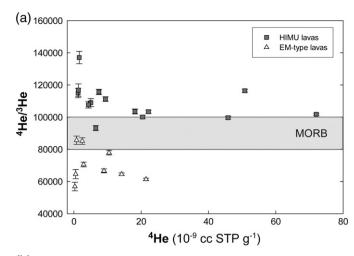
3.2. Relationships between He and other radiogenic tracers

The high ⁴He/³He ratios measured in basalts from the islands of Raivavae, Mangaia, and Tubuai are associated with low ⁸⁷Sr/⁸⁶Sr, high ¹⁴³Nd/¹⁴⁴Nd, and very radiogenic Os and Pb isotopic compositions (Fig. 3). We consider samples with ²⁰⁶Pb/²⁰⁴Pb > 20 to be typical HIMU

Table 1He isotopic composition of Cook–Austral basalts

Location	Sample name	Mass	$(10^{-9} \text{ cm}^3 \text{ STP g}^{-1})$	⁴ He/ ³ He	±1σ	³ He/ ⁴ He
		(g) [⁴ He]				(R_{A})
Raivavae	RVV 302	0.52	20.41	100 000	1000	7.19
	RVV 310	0.64	1.24	115000	3000	6.26
	RVV 316	0.36	1.56	137000	4000	5.25
	RVV 318	0.48	22.14	103 000	1000	6.96
	RVV 340	0.20	5.03	109000	3000	6.60
	RVV 344	0.55	7.43	116000	2000	6.22
	RVV 346	0.19	18.14	104000	2000	6.95
	RVV 360a	0.28	1.32	117000	4000	6.16
	RVV 370	0.32	45.81	99600	900	7.22
Mangaia	MGA B26	0.59	9.39	111000	1000	6.47
	MGA B47	0.43	50.80	116000	900	6.18
Tubuai	TBA B3	0.37	4.30	108 000	2000	6.68
	TBA B22	0.65	72.04	102000	600	7.07
Rurutu (old)	RRT B21	0.34	6.49	93200	1600	7.72
Rapa	RPA 02	0.32	0.51	64600	2800	11.1
	RPA 14	0.27	21.42	61300	700	11.7
	RPA 31	0.58	0.79	85600	2600	8.40
	RPA 71	0.35	2.47	85100	2000	8.45
	RPA 367	0.45	2.89	70500	1500	10.2
	RPA 473	0.46	14.18	64600	800	11.1
	RPA 488	0.17	10.43	77900	1400	9.24
	RPA 502	0.23	8.93	66600	1200	10.8
Rurutu	RRT B30	0.48	0.29	56900	2700	12.7
(young)						

Additional information and sample locations for the Raivavae and Rapa samples can be found in Lassiter et al. (2003); information for the Mangaia, Tubuai, and Rurutu samples can be found in Hauri and Hart (1993). $R_{\rm A}$ is the $^{3}{\rm He}/^{4}{\rm He}$ ratio normalized to the atmospheric ratio of 1.39×10^{-6} .



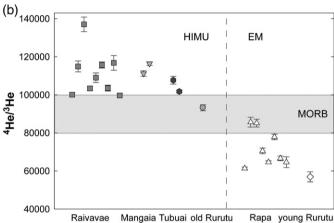


Fig. 2. (a) Helium concentration versus ${}^4\text{He}/{}^3\text{He}$ ratios. There are no significant correlations, indicating that post-eruptive radiogenic and cosmogenic He do not affect the measured isotopic ratios. Error bars on ${}^4\text{He}/{}^3\text{He}$ ratios are 1σ values. (b) Helium isotopic composition in Cook–Austral samples. Samples from HIMU-type ocean islands are characterized by ${}^4\text{He}/{}^3\text{He}$ ratios more radiogenic than MORBs, while samples from EM-type islands exhibit ${}^4\text{He}/{}^3\text{He}$ ratios less radiogenic than MORBs. Error bars on ${}^4\text{He}/{}^3\text{He}$ ratios are 1σ values.

lavas. The association of radiogenic helium isotopes with well-defined Os, Sr, and Nd isotopic compositions indicates that the high ${}^4\mathrm{He}/{}^3\mathrm{He}$ ratios reflect the mantle source composition of the lavas and thus do not arise from contamination with oceanic lithosphere or from prolonged storage of magmas in shallow crustal chambers (Zindler and Hart, 1986b; Hilton et al., 1995). The low ${}^4\mathrm{He}/{}^3\mathrm{He}$ ratios sampled at Rapa and in young Rurutu lavas are also associated with well-defined Os, Sr, and Nd isotopic signatures, which are enriched compared to both the HIMU lavas and MORBs. Thus, there is a robust association between the He isotopic composition and the Os, Pb, Sr, and Nd isotopic compositions of the Cook–Austral basalts (Fig. 3).

3.3. Neon isotopic systematics

Fig. 4 presents neon isotopic data from an Icelandic basalt glass (DICE; Trieloff et al., 2000) as well as a basalt glass from the Hotu Matua seamount chain that has a $^4\text{He}/^3\text{He}$ ratio in the MORB range (84 300; Table 2). Our neon isotopic data from DICE are identical to Trieloff et al.'s (2000) measurements, although our error bars are smaller, while the Hotu Matua sample is indistinguishable from popping rock (2 Π D43; Moreira et al., 1998). These data demonstrate our ability to very precisely measure neon isotopic compositions in mantle-derived basalts. Our high-precision measurements from the Cook–Austral chain detected mantle-derived neon in basalts from Raivavae,

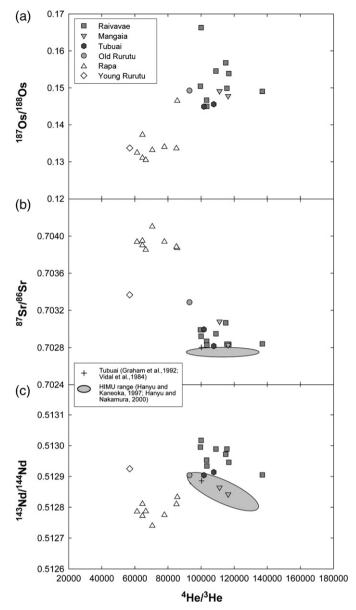


Fig. 3. Relationships between He isotopic composition and (a) Os, (b) Sr, and (c) Nd isotopic composition. The initial ¹⁸⁷Os/¹⁸⁸Os ratios are shown in panel (a), as posteruptive in-growth of ¹⁸⁷Os in some of these samples can be significant. In each plot, two distinct isotopic groups are evident: the HIMU group (high ⁴He/³He ratios) and the EM-type group (low ⁴He/³He ratios). The radiogenic isotopic data are from Hauri and Hart (1993) and Lassiter et al. (2003). The elliptical shaded region defines the range in He, Sr, and Nd isotopic compositions in HIMU lavas from Mangaia, Tubuai, and old Rurutu (Hanyu and Kaneoka, 1997; Hanyu and Nakamura, 2000). A previously studied sample from Tubuai is also shown (He, Sr, and Nd; Vidal et al., 1984; Graham et al., 1992a). Lithophile isotopic data and analytical procedures can be found in Hauri and Hart (1993) and Lassiter et al. (2003); new lithophile data for Raivavae and Rapa samples can be found in the supplement.

Mangaia, Tubuai, and Rapa (Table 2; Fig. 5). The highest $^{20}\text{Ne}/^{22}\text{Ne}$ ratio measured in the HIMU lavas is 10.94, while the highest value for the EM-type lavas from Rapa is 10.50 (Fig. 5a). The highest $^{21}\text{Ne}/^{22}\text{Ne}$ ratios are 0.0401 and 0.0301 for HIMU and EM-type lavas, respectively (Fig. 5a). With the exception of the Mangaia sample, enrichments in $^{20}\text{Ne}/^{22}\text{Ne}$ and $^{21}\text{Ne}/^{22}\text{Ne}$ ratios with respect to atmospheric values are modest. However, our high-precision measurements clearly demonstrate non-atmospheric compositions, and extrapolation of $^{21}\text{Ne}/^{22}\text{Ne}$ ratios to solar $^{20}\text{Ne}/^{22}\text{Ne}$ values (to correct for atmospheric contamination: $^{21}\text{Ne}/^{22}\text{Ne}_{\text{E}}$) results in relatively small uncertainties. The $^{21}\text{Ne}/^{22}\text{Ne}_{\text{E}}$ ratios range from $0.036^{+0.01}_{-0.03}$ to 0.066 ± 0.004 (Table 2). The EM-

type lava from Rapa is significantly less nucleogenic than both HIMU lavas and MORBs, while the HIMU lavas extend from the MORB range to compositions less nucleogenic than MORBs. Hence, while HIMU lavas have more radiogenic He isotopic compositions than MORBs, the Ne isotopic composition is clearly not more nucleogenic than MORBs (Fig. 5).

Barfod et al. (1999) report the only other non-atmospheric neon measurements in HIMU lavas from the Cameroon line of volcanoes (Fig. 5b). However, the Pb, Sr, and Nd isotopic compositions of the Cameroon line of volcanoes are distinct from typical HIMU localities, such as St. Helena or Mangaia (e.g., Halliday et al., 1990). Therefore, it was uncertain how representative the Ne isotopic data from the Cameroon line of volcanoes were of the typical HIMU mantle source. Our high-precision Ne isotopic data are consistent with the Ne isotopic compositions from the Cameroon line of volcanoes, and indicate that the HIMU basalts may generally be characterized by Ne isotopic compositions that extend from the MORB range to compositions less nucleogenic than MORBs.

4. Discussion

There are three notable features of the Cook–Austral data presented above: (1) ⁴He/³He ratios both lower and higher than canonical MORB values of 80 000–100 000 are measured in lavas erupted in close geographical proximity; (2) clear co-variations in He and Os, Pb, Sr, and Nd isotopic compositions are present; and (3) ²¹Ne/ ²²Ne ratios in EM-type lavas are less nucleogenic than MORB neon, while HIMU lavas extend from MORB range to less nucleogenic compositions. In the following sections, we will evaluate the hypotheses previously proposed for the origin of the HIMU noble gas signature, and discuss the origin of the HIMU and EM-type components sampled by volcanoes in the Cook–Austral Islands. We will propose a simple mixing model to explain the noble gas compositions measured in both the HIMU and EM-type lavas simultaneously.

4.1. Origin of the HIMU lavas

The range of ⁴He/³He ratios that we measured in the HIMU lavas (Table 1) is consistent with ⁴He/³He ratios previously reported from the Cook–Austral Islands (Hanyu et al., 1999) and ⁴He/³He ratios at other

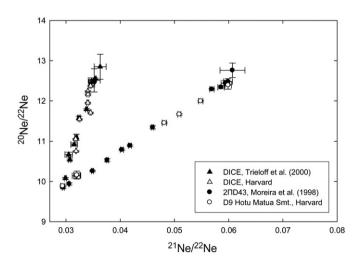


Fig. 4. Neon three isotope plot showing our reanalysis of DICE glass and a basaltic glass (D9) from the Hotu Matua seamount chain off the East Pacific Rise, which has a 4 He/ 3 He ratio of 84 300±600 (8.56±0.06 R_A). Our analysis of DICE is indistinguishable from previous measurements by Trieloff et al. (2000), and the neon isotopic composition of D9 is indistinguishable from that of popping rock (Moreira et al., 1998). These measurements demonstrate our ability to measure high-precision neon isotopic measurements in mantle-derived rocks.

Table 2Neon isotopic composition of Cook–Austral basalts

Location	Sample name	Mass (g)	Step [²² Ne]	$(10^{-12} \text{ cm}^3 \text{ STP g}^{-1})$	²⁰ Ne/ ²² Ne	±1σ	²¹ Ne/ ²² Ne	±1σ	21 Ne/ 22 Ne _E	±1σ
Raivavae	RVV 318	3.00	1	0.53	10.23	0.03	0.0322	0.0003	0.0585	0.0068
Mangaia	MGA B47	1.82	1	0.27	10.94	0.05	0.0401	0.0006	0.0660	0.0036
iviangala	WIGH D47	1.02	2	0.19	10.79	0.06	0.0373	0.0006	0.0000	0.0050
Tubuai Rapa	TBA B22	2.06	1	5.11	10.17	0.009	0.0309	0.0001	0.0501	0.0036
	15/15/2	2.00	2	0.20	10.06	0.05	0.0309	0.0006	0.0301	0.0050
	RPA 502	1.04	1	1.33	10.00	0.03	0.0297	0.0003		
кара	1071 302	1.01	2	0.49	9.94	0.04	0.0290	0.0003	0.036	0.011
			3	0.23	10.50	0.05	0.0301	0.0004	0.030	0.011
"Standard" s	samples									
DICE, Midfel	ll, Iceland	3.46	1	14.1	10.76	0.01	0.0319	0.0001		
			2	13.9	12.24	0.01	0.0340	0.0001		
			3	15.2	11.05	0.01	0.0318	0.0001		
			4	5.49	11.54	0.01	0.0325	0.0001	0.0381	0.0003
			5	2.53	12.37	0.01	0.0345	0.0001		
			6	1.79	11.71	0.01	0.0345	0.0001		
			7	2.32	11.95	0.01	0.0340	0.0002		
			8	2.55	12.16	0.01	0.0341	0.0002		
D9, Hotu Ma	atua Smt. 15.29°S,	0.94	1	9.96	11.45	0.02	0.0481	0.0004		
116.03°W w	ater depth		2	25.80	11.67	0.02	0.0509	0.0003	0.0759	0.0008
3330 m (~3	00 km west		3	11.51	11.99	0.02	0.0548	0.0004		
of EPR)			4	23.76	10.14	0.07	0.0319	0.0008		
		0.61	1	1608.34	9.89	0.02	0.0294	0.0004		
			2	0.92	12.39	0.10	0.0599	0.0006	0.0763	0.0032
			3	0.70	10.16	0.10	0.0320	0.0006		

Additional information on the DICE sample can be found in Harrison et al. (1999). Sample D9 has 4 He/ 3 He=84 300±600 (3 He/ 4 He=8.53±0.06 R_A).

HIMU localities such as Cape Verde, St. Helena, and Canary Islands (Graham et al., 1992a; Barfod et al., 1999; Hilton et al., 2000; Moreira and Kurz, 2001; Christensen et al., 2001). Our coupled helium and neon isotopic measurements in the Cook-Austral lavas allow us to evaluate the hypothesis that diffusive equilibration between recycled crust and DMM generates the ⁴He/³He ratios observed in HIMU lavas (Hanyu and Kaneoka, 1998). Recycled crust would be extensively outgassed of neon (e.g., Hiyagon, 1994; Moreira et al., 2003) and would have high timeintegrated (U+Th)/²²Ne ratios. Nucleogenic ²¹Ne would be produced over time in recycled crust through $^{18}O(\alpha n)^{21}Ne$ and $^{25}Mg(n,\alpha)^{21}Ne$ reactions. Hence, radiogenic ⁴He/³He ratios would be associated with nucleogenic ²¹Ne/²²Ne ratios. Noble gas diffusivities in mantle materials are not well-constrained. However, the atomic volume of Ne is ~40% larger than He (Zhang and Xu, 1995) and Ne diffusivities should be lower than He diffusivities. Barfod et al. (1999) demonstrated that the generation of ${}^{4}\text{He}/{}^{3}\text{He}$ ratios of ~120000 (${}^{3}\text{He}/{}^{4}\text{He}$ of 6 R_{A}) in HIMU lavas through diffusive equilibration between recycled crust and DMM would result in neon isotopic compositions in HIMU lavas that are more nucleogenic than DMM. Even if we assume that the diffusivities of He and Ne are the same in the mantle, diffusive equilibration between DMM and recycled crust cannot explain why HIMU He isotopic compositions are more radiogenic than MORBs, while HIMU Ne isotopic compositions are similar to or less nucleogenic than MORBs. As a result, the Ne isotopic compositions from the Cook-Austral Islands and the Cameroon line of volcanoes (Barfod et al., 1999) rule out diffusive equilibration between recycled crust and DMM as the dominant process responsible for generating the observed ⁴He/³He ratios and ²¹Ne/²²Ne ratios in HIMU lavas. The Ne isotopic data further indicate that the radiogenic ⁴He/³He ratios in HIMU lavas from the Cook-Austral Islands do not arise from sampling of recycled oceanic lithosphere (Moreira and Kurz, 2001). Oceanic lithosphere would have ⁴He/³He ratios that are more radiogenic than MORBs (Moreira and Kurz, 2001) paired with ²¹Ne/²²Ne ratios that are more nucleogenic than MORBs.

We consistently observe good co-variations between helium isotopic composition and radiogenic isotopic composition in the Cook–Austral chain (Fig. 3), suggesting that helium is not decoupled from other isotopic tracers by the late addition of radiogenic ⁴He from melts frozen in the lithosphere (e.g., Hilton et al., 2000). We also

observe a correlation between Ne isotopic compositions and Pb isotopic compositions (Fig. 6). Although the data set is small (n=4), the Mangaia sample with the highest 206 Pb/ 204 Pb has the highest 21 Ne/ 22 Ne $_{\rm E}$ ratio, and Raivavae samples with less radiogenic Pb have lower 21 Ne/ 22 Ne $_{\rm E}$ ratios. Based on the observed co-variations, we suggest that a relatively undegassed mantle component is involved in the genesis of HIMU basalts. We will evaluate this hypothesis quantitatively in Section 4.3.

4.2. Origin of the EM lavas

The low ⁴He/³He ratios sampled in Rapa and young Rurutu lavas are associated with well-defined Os, Pb, Sr, and Nd isotopic signatures (Fig. 3). The compositions observed in these lavas indicate the involvement of a mantle component that is more enriched than both HIMU and DMM. We note that the compositions in the young Rurutu and Rapa samples are, however, quite distinct from the isotopically enriched lavas at Samoa (EM II), or at the Ko'olau volcano (EM I) in Hawaii.

The nature of the enriched mantle (EM) component in Cook–Austral basalts has been debated in the literature. HIMU and EM-type volcanism are often geographically associated, which may indicate a genetic link between these two components (Hart et al., 1986; Chauvel et al., 1992). Hemond et al. (1994) and Hauri et al. (1996) suggest that the EM signature may result from addition of recycled sediments to a HIMU source, which would explain the close association between the two components. However, sediments have high Pb concentrations and very high Pb/Os ratios, so the addition of sediments to a HIMU source would lead to very radiogenic Os isotopic compositions in the EM-type lavas from Rapa, which are not observed (Fig. 3; Lassiter et al., 2003).

Models of recycled or rafted continental lithospheric as the source of the EM-type signature have also been suggested (Milner and leRoex, 1996; Schiano et al., 2001). However, mixing between recycled continental lithosphere and recycled crust should generate extremely hyperbolic mixing arrays in Os–Pb and Os–Nd space (Lassiter et al., 2003). Lassiter et al. (2003) suggest that the EM component represents subduction-modified sub-arc mantle. In this scenario, slab-derived melts or fluids enriched in incompatible elements impart enriched

9

0.03

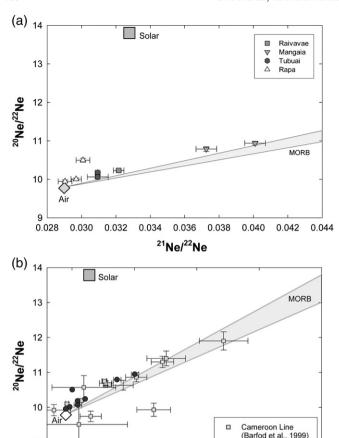


Fig. 5. Neon three isotope diagram showing (a) Cook–Austral data and (b) Cook–Austral data and Cameroon data (from Barfod et al., 1999). Shaded region represents the MORB range from Sarda et al. (2000) (21 Ne/ 22 Ne $_{\rm E}$ =0.075±0.005). Solar values are 21 Ne/ 22 Ne=0.0328 and 20 Ne/ 22 Ne=13.8 (Benkert et al., 1993) and atmospheric values are 21 Ne/ 22 Ne=0.029 and 20 Ne/ 22 Ne=9.8. Note that the Cook–Austral data are less nucleogenic than MORBs. Error bars shown are 1 σ .

0.05

²¹Ne/²²Ne

0.04

Cook-Austral Islands

0.07

0.06

isotope and trace element signatures to the mantle wedge. Viscous coupling between sub-arc mantle and subducted slab could then account for the association between HIMU and EM signatures.

Our noble gas data have bearing on all three of these hypotheses. The lowest measured ⁴He/³He ratio from a Rapa sample is 61 300 (³He/ 4 He of 11.7 $R_{\rm A}$), while the young Rurutu sample has a ratio of 56 900 $(^{3}\text{He})^{4}\text{He of } 12.6~R_{A})$. These $^{4}\text{He}/^{3}\text{He ratios are relatively unradiogenic}$ and are significantly lower than canonical MORB ratios. In addition, the low ⁴He/³He ratios from Rapa are associated with a ²¹Ne/²²Ne_E ratio that is significantly less nucleogenic than MORBs (Fig. 7, Table 2). Relatively unradiogenic ⁴He/³He ratios of 57000 to 62000 and ²¹Ne/ ²²Ne_E of 0.036 do not originate from subducted sediments. Nor do these noble gas signatures arise from continental lithospheric mantle, which is characterized by ⁴He/³He ratios more radiogenic than MORBs (Gautheron and Moreira, 2002). Consequently, there is no evidence for the involvement of recycled continental lithosphere in the source of the enriched Rapa lavas. Likewise, sub-arc mantle that has been enriched isotopically and in trace elements by slab-derived melts or fluids will not have low ⁴He/³He ratios or low ²¹Ne/²²Ne_E ratios. He and Ne are not subducted back into the mantle and hence, slab derived melts and fluids should be characterized by radiogenic He and nucleogenic Ne isotopic compositions. Based on the geochemical signature of the EM-type lavas (Figs. 3-6), we hypothesize that the relatively undegassed material present in HIMU lavas occurs in greater proportion in the source of the EM-type lavas. This hypothesis is also consistent with measurements of 4 He/ 3 He ratios at the Macdonald seamount (45 000 to 73 200; Moreira and Allegre, 2004), which suggest the involvement of a relatively undegassed mantle component in EM lavas from the Cook–Austral Islands.

4.3. Mixing models for generating HIMU and EM-type lavas simultaneously

The Ne isotopic data require that both the HIMU and EM lavas sample a mantle component that is relatively undegassed, characterized by low ⁴He/³He ratios and low ²¹Ne/²²Ne ratios. Fig. 7 shows the Cook-Austral and Cameroon data (Barfod et al., 1999) in ⁴He/³He-²¹Ne/²²Ne_E space. A simple two component mixture involving a HIMU component and a relatively undegassed mantle component can generate the observed trends. The HIMU component is modeled with ${}^{4}\text{He}/{}^{3}\text{He}$ of 7×10^{7} (${}^{3}\text{He}/{}^{4}\text{He}$ of 0.01 R_{A}), which corresponds to ⁴He/³He ratios expected in recycled crust after 1 Ga residence in the mantle. Alternatively, the HIMU component can be modeled with an endmember composition of 160 000 (3 He/ 4 He of 4.5 R_A ; Fig. 7), which corresponds to the highest ⁴He/³He ratios measured in olivine crystals at St. Helena (Graham et al., 1992a). For the relatively undegassed mantle, we use the lowest measured OIB ⁴He/³He ratio of 14 400 (³He/ ⁴He of 50 R_A ; Stuart et al., 2003) and the solar ²¹Ne/²²Ne ratio of 0.0328 (Benkert et al., 1993; Dixon et al., 2000). We have abbreviated the relatively undegassed mantle as RUM for the purposes of the discussion below, but hope that the acronym does not become ingrained in the literature. The He-Ne data are consistent with mixing between the HIMU component and the RUM component, with the EM-type lava from Rapa having a higher proportion of RUM than the HIMU lavas. Within the uncertainty of the measurements, mixing between a HIMU component and depleted mantle may account for some of the data points. However, in such a scenario, a third component that is less nucleogenic than DMM would still be required to explain all of the data.

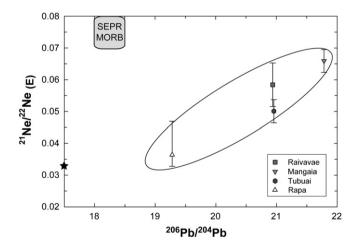


Fig. 6. Correlation between $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{21}\text{Ne}/^{22}\text{Ne}_\text{E}$ isotopic compositions in the Cook–Austral basalts. Symbols as in Fig. 3 (filled symbols are HIMU lavas and open symbol is an EM-type lava). $^{21}\text{Ne}/^{22}\text{Ne}_\text{E}$ values were determined by extrapolating measured $^{21}\text{Ne}/^{22}\text{Ne}$ ratios to a mantle $^{20}\text{Ne}/^{22}\text{Ne}$ of 13.8 using an error-weighted fit. Note that extrapolating to $^{20}\text{Ne}/^{22}\text{Ne}$ = 12.5 does not affect our conclusions. Only sample steps with $^{20}\text{Ne}/^{22}\text{Ne}$ ratios ≥ 10 and distinct from atmospheric ratios within 1 σ were used for the extrapolation. In spite of the limited data set, there appears to be a positive correlation: the most radiogenic $^{206}\text{Pb}/^{204}\text{Pb}$ values are associated with the highest $^{21}\text{Ne}/^{22}\text{Ne}_\text{E}$. The correlation suggests that the HIMU Ne isotopic composition could result largely from mixing of a HIMU component and a relatively undegassed mantle component. The star represents the solar $^{21}\text{Ne}/^{22}\text{Ne}$ ratio. The SEPR MORB field is drawn based on Pb data from Mahoney et al. (1994), while the Ne isotopic range is based on the inferred MORB end-member in the SEPR (Kurz et al., 2005), our measurements in the Hotu Matua seamount chain off the SEPR (Table 2), and the MORB range of Sarda et al. (2000). Error bars on $^{21}\text{Ne}/^{22}\text{Ne}_\text{F}$ are 1 σ .

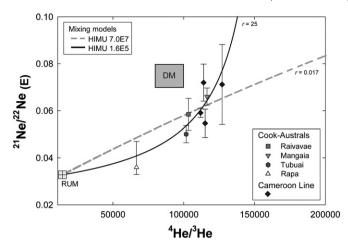


Fig. 7. Helium-neon mixing systematics. The Cook–Austral data are from this study, and data from the Cameroon line of volcanoes are from Barfod et al. (1999). Sample steps with 20 Ne/ 22 Ne ratios ≥ 10 and distinct from atmospheric ratios within 10 were selected for error-weighted extrapolation of measured 21 Ne/ 22 Ne to mantle 20 Ne/ 22 Ne ratios of 13.8. Error bars are 10 . The dashed line represents two-component mixing between a relatively undegassed mantle (RUM) end-member with 4 He/ 3 He ratio of 7 ×10 7 . The solid line represent mixing between RUM and a HIMU end-member with 4 He/ 3 He ratio of 7 ×10 7 . The solid line represent mixing between RUM and a HiMU end-member with 4 He/ 3 He of 160 000. The 21 Ne/ 22 Ne ratio of the HIMU end-member is taken to 0.4 (Barfod et al., 1999) and the RUM end-member has a solar 21 Ne/ 22 Ne ratio. The curvature of the lines, 7 , is defined as (22 Ne/ 3 He)_{RUM}/ 22 Ne/ 21 Ne/ 11 He/ 11 He/ 11 He/HMI- 11 He/HMI- 11 He/ 11

While a simple two component mixture between a HIMU component and RUM is consistent with the major trends in He and lithophile isotopic composition of the Cook–Austral basalts (see supplement), previous studies based on Sr, Nd, Pb and Os isotopic data have identified at least three components in the Cook–Austral islands: a HIMU component, a depleted mantle component (DM), and an EM component (e.g., Nakamura and Tatsumoto, 1988; Hemond et al., 1994; Lassiter et al., 2003). Based on He and Ne isotopic measurements, we have argued that the EM component is relatively undegassed mantle. We now investigate whether a single mixing scenario involving a relatively undegassed mantle component, a HIMU component and a depleted mantle component can account for the compositions of both HIMU and EM-type lavas simultaneously.

4.3.1. Compositions of the mantle components

We use compositions previously identified for the HIMU component, the low ⁴He/³He mantle component, and the depleted mantle component. The HIMU component is modeled with ${}^{4}\text{He}/{}^{3}\text{He}$ of 7×10^{7} , ²⁰⁶Pb/²⁰⁴Pb of 23, ¹⁸⁷Os/¹⁸⁸Os ratio of 10, ⁸⁷Sr/⁸⁶Sr of 0.7026, and 143 Nd/ 144 Nd of 0.51295, consistent with previous estimates of recycled crustal compositions (Chauvel et al., 1992; Niedermann et al., 1997; Barfod et al., 1999; Becker, 2000). Additional three-component mixing models are presented in the supplement using a HIMU end-member composition of 160 000 for the ⁴He/³He ratio. The RUM component is taken to have ⁴He/³He of 14400 (Stuart et al., 2003) and lithophile isotopic compositions similar to FOZO-A: ⁸⁷Sr/⁸⁶Sr of 0.7047, ¹⁴³Nd/ ¹⁴⁴Nd of 0.5127, ²⁰⁶Pb/²⁰⁴Pb of 18.5, and ¹⁸⁷Os/¹⁸⁸Os of 0.129. FOZO-A is the carrier of the low ⁴He/³He signature in Southern Hemisphere OIBs, and is slightly enriched in composition compared to Bulk Silicate Earth (Jackson et al., 2007). Our mixing calculations would, however, be equally consistent if we selected PHEM (Farley et al., 1992) as the starting composition. The depleted mantle (DM) component that is present in the source of the Cook-Austral basalts is not normal asthenospheric mantle (Lassiter et al., 2003). Instead, based on Pb and Os isotopic compositions, Lassiter et al. (2003) argued that the DM component may derive from low-degree melts of relatively enriched materials present in the ambient asthenospheric mantle (e.g., pyroxenite veins). We have taken the ${}^{4}\text{He}/{}^{3}\text{He}$ of the DM component to be 90 000 (${}^{3}\text{He}/{}^{4}\text{He}$ of 8 R_{A}), and use ²⁰⁶Pb/²⁰⁴Pb of 18.5, ¹⁸⁷Os/¹⁸⁸Os ratio of 0.160, ⁸⁷Sr/⁸⁶Sr of 0.7028, and 143Nd/144Nd of 0.5133 (See Figs. 3-6 in Lassiter et al., 2003).

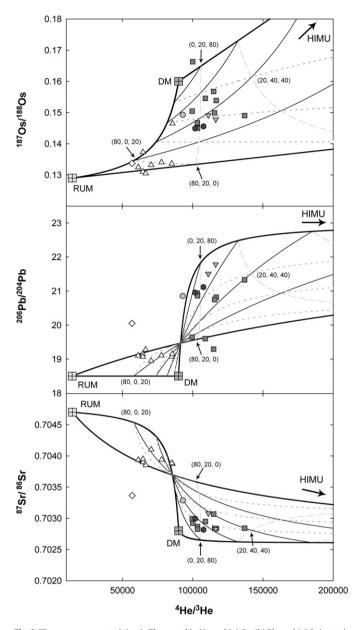


Fig. 8. Three-component mixing is illustrated in He and (a) Os, (b) Pb, and (c) Sr isotopic space. Symbols as in Fig. 3. The isotopic compositions of the components are shown in the figures and listed in Section 4.3.1. The relatively undegassed mantle (RUM) component has an isotopic composition similar to FOZO-A. For simplicity, the concentrations in the RUM component are taken to be similar to primitive mantle (21 ppm Sr, 1.34 ppm Nd, 0.185 ppm Pb and 3.4 ppb Os; McDonough and Frey, 1989; Hilton et al., 2000). However, randomly varying these concentrations by 30-40% does not affect the mixing surface significantly. Concentrations in the HIMU component are based on estimates of recycled crust: 94.3 ppm Sr, 9.32 ppm Nd, 0.23 ppm Pb and 6 ppt Os (Chauvel et al., 1992; Becker, 2000). The depleted mantle (DM) component is taken to have 8 ppm Sr, 0.6 ppm Nd, 0.02 ppm Pb and 3 ppb Os (Salters and Stracke, 2004; Workman and Hart, 2005). We first determine relative ¹⁸⁸Os/³He ratios in the three components that best fit the data set. The relative helium concentrations in the components were then propagated though to He-Sr and He-Pb isotopic spaces to generate an internally consistent set of mixing surfaces (see Section 4.3.2). The contours represent a constant proportion of component X in the mixture (solid black line X=DM: dashed grey line X=HIMU; dotted line X=RUM). Intersections between some contours have been labeled with ordered triples (%RUM, %HIMU, %DM) to give a sense of relative proportions. In all isotopic spaces considered, the Cook-Austral data are consistent with three-component mixing between the HIMU component, the relatively undegassed mantle component, and the depleted mantle component. We note that a component with geochemical characteristics more primitive than either DM or HIMU is clearly required (also see Figs. 6 and 7).

4.3.2. He, Os, Pb, and Sr mixing systematics

Determining the absolute helium abundance in either the mantle source or in the parental magma is problematic due to magmatic degassing. As a result, we first determined the relative $(^{188}Os/^{3}He)$ ratios in the three components required to generate a mixing surface that fits the He–Os data $[(^{188}Os/^{3}He)_{RUM}:(^{188}Os/^{3}He)_{HIMU}:(^{188}Os/^{3}He)_{HIMU}:(^{188}Os/^{3}He)_{DM}=1: 0.35: 0.16]$. Using the X/Os ratios in the components (where X is Sr or Pb concentrations), the $(X/^{3}He)$ ratios were then computed.

The Cook-Austral ⁴He/³He ratios are plotted against ¹⁸⁷Os/¹⁸⁸Os, ²⁰⁶Pb/²⁰⁴Pb, and ⁸⁷Sr/⁸⁶Sr ratios in Fig. 8a-c, along with mixing surfaces generated by three-component mixing of RUM, HIMU, and DM components. Mixing systematics in He-Nd space are also consistent with the data, but have been omitted for clarity. The results from the three-component mixing calculations indicate that the combined He, Os, Pb, Sr, and Nd isotopic compositions in the Cook-Austral basalts can be attributed to a mixture of relatively undegassed mantle component, a recycled crustal component, and a depleted mantle component. In all isotopic spaces, HIMU lavas have a higher proportion of recycled crust than EM-type lavas, while the EM lavas have a higher proportion of relatively undegassed mantle material. We note that the implied ³He concentration in the undegassed mantle component is lower than in the implied ³He concentration in the depleted mantle component, the so-called Heparadox. The resolution of the He-paradox may be related to a higher CO₂ content in the undegassed mantle component. A higher CO₂ content for the EM lavas at Rapa, compared to the HIMU lavas from Raivavae, is consistent with melt inclusions studies (Lassiter et al., 2002). Higher CO₂ content would lead to deeper degassing and consequently, more He loss. Furthermore, higher CO₂ contents in the parental magma would dilute the He partial pressure, which would decrease He solubility and concurrently lead to greater loss of He from the magma (Gonnermann and Mukhopadhyay, 2007). However, this indicates that the magmas degas prior to mixing, presumably in shallow crustal magma chambers. Such a scenario is supported by the significantly wider range in Pb isotopic compositions seen in olivinehosted melt inclusions at Mangaia, compared to whole rock samples from Mangaia (Saal et al., 1998).

Our suggestion that a relatively undegassed mantle reservoir is involved in the genesis of the Cook-Austral HIMU lavas is consistent with observations from other HIMU localities, such as the Cameroon line of volcanoes, Canary Islands, and Cape Verde (Barfod et al., 1999; Hilton et al., 2000; Christensen et al., 2001; Moreira and Allegre, 2004). We suggest that the observed ⁴He/³He ratios do not require either closed system aging of melts derived from the low ⁴He/³He plume source for $\sim 10^8$ years (Barfod et al.; 1999), or the addition of radiogenic ⁴He from melts frozen in the lithosphere to the relatively undegassed, low ⁴He/³He mantle component (Hilton et al.; 2000). Rather, the helium isotopic signatures are generated by mixing of relatively undegassed mantle with recycled crust and a depleted mantle component. Accordingly, OIBs with ⁴He/³He ratios higher than MORBs (HIMU OIBs) and OIBs with ⁴He/³He ratios lower than MORBs (e.g., Hawaii, Samoa) share a common source characterized by low ⁴He/³He ratios and low ²¹Ne/²²Ne ratios.

The ²¹Ne/²²Ne ratios in HIMU basalts provide additional insight into slab recycling processes. If slabs were recycled back to the surface from the upper mantle, the He and Ne isotopic signatures would be constrained to lie between the MORB range and recycled crust (Fig. 7). On the other hand, lower mantle material can have ²¹Ne/²²Ne ratios that are less nucleogenic than MORBs (e.g., Moreira and Allegre, 1998; Kurz et al., 2005) and the occurrence of ²¹Ne/²²Ne ratios in HIMU basalts that are less nucleogenic than MORBs may suggest slab subduction into the lower mantle. Subduction of slabs into the lower mantle does not invalidate the hypothesis that the lower mantle is relatively undegassed compared to the depleted upper mantle. Gonnermann and Mukhopadhyay (in review) recently suggested

that recycling and mixing of slabs with the ambient lower mantle actually helps to preserve primordial gas concentrations in the lower mantle. Subducted slabs are severely depleted in primordial noble gases and hence, mixing of slabs with the lower mantle dilutes noble gas concentrations in this mixed mantle assemblage. Because mantle degassing rates depend not only on magma production rates but also on the concentration of the noble gases in the mantle source, there has been a dramatic decrease in the degassing efficiency of the noble gases over time. As a result, high ³He concentrations (>40 times MORB source) and low ⁴He/³He ratios (down to 14000) can be preserved in the lower mantle, even if the mass flux across the 670 km boundary is on order one lower mantle mass over 4.5 Ga (Gonnermann and Mukhopadhyay, in review). Therefore, convective isolation of the lower mantle is not required for any length of time, and the RUM material can originate from the lower mantle. Slab subduction into the lower mantle would result in mixing between RUM and recycled material through convective stirring, which would presumably produce a range of compositions. Thus, the close geographical association between EM-type lavas and HIMU lavas could result from mixing and entrainment processes in the lower mantle, with the EM-type lavas have a higher proportion of the undegassed material than the HIMU lavas.

5. Conclusions

We have measured He and Ne isotopic compositions in basalts from the Cook-Austral Islands. Samples that were previously classified as HIMU lavas based on Os, Sr, Nd, and Pb isotopic character exhibit ⁴He/³He ratios more radiogenic than MORBs, while EM-type lavas exhibit ⁴He/³He ratios less radiogenic than MORBs. The Ne isotopic composition of the EM-type lava is less nucleogenic than MORBs, while HIMU lavas extend from the MORB range to compositions less nucleogenic than MORBs. The Ne isotopic compositions of Cook-Austral HIMU lavas are consistent with compositions reported from the Cameroon line of volcanoes (Barfod et al., 1999), and indicate that HIMU basalts may generally be characterized by Ne that is similar to or less nucleogenic than MORBs. Therefore, the Ne isotope systematics in HIMU lavas from the Cook-Austral Islands require the involvement of a relatively undegassed mantle component. A simple mixing scenario involving a HIMU component, a relatively undegassed mantle component, and a depleted mantle component is consistent with the noble gas and Os, Pb, Sr, and Nd isotopic compositions of Cook-Austral lavas. We suggest that the close geographical association between HIMU and EM-type lavas reflects variable mixing proportions of relatively undegassed mantle material and recycled oceanic crust with depleted mantle. The association between recycled crust and relatively undegassed mantle may be a natural consequence of mixing and entrainment processes resulting from slab subduction into a relatively undegassed lower mantle. We suggest that the Ne isotopic composition of HIMU basalts may be a useful indicator of subduction of oceanic crust into the lower mantle.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.epsl.2008.10.014.

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