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Nucleogenic neon in high ³He/⁴He lavas from the Manus back-arc basin: a new perspective on He–Ne decoupling

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Abstract

We report new neon isotope data obtained for well-characterised basaltic glasses from the Manus back-arc basin where helium studies have identified a mantle plume component (mean ${}^{3}\text{He}/{}^{4}\text{He} \sim 12 R_{A}$). In three-isotope neon space, seven of the Manus samples lie along a trajectory between air and an endmember more nucleogenic than MORB i.e., compared to typical MORB, samples have a higher ²¹Ne/²²Ne ratio for a given ²⁰Ne/²²Ne ratio. Thus the slope of the Manus Basin line is less than that of the MORB line [Sarda et al., Earth Planet. Sci. Lett. 91 (1988) 73-88]. This is the first observation of lavas with high ³He/⁴He ratios having nucleogenic neon isotope systematics, indicating a unique decoupling of He from Ne. We evaluate five possible explanations for the observed trend. We discount: (1) crustal contamination, (2) devolatisation of the subducting Solomon Sea plate and (3) addition of neon from an ancient recycled slab component - based upon mass balance considerations of the availability of nucleogenic Ne. Two possibilities remain - both of which must produce an elevated He/Ne ratio in the Manus Basin source region to account for the nucleogenic neon: (4) a previous degassing event which would leave a Ne-depleted residual reservoir, or (5) a deep mantle source heterogeneity preserving a unique signature inherited from Earth's accretion. We find that isolation times as short as 10 Ma for a previously degassed source are sufficient to grow in the nucleogenic Ne without significantly altering the plume-like ³He/⁴He ratios. Alternatively, solubility-controlled outgassing/ingassing of a magma ocean in contact with a proto-atmosphere may have produced the requisite high He/Ne ratio, although an opensystem style of equilibration is necessary. At present, insufficient evidence is available to discriminate between these alternatives. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

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Neon and helium isotopes are highly sensitive indicators of mantle plume involvement in petrogenesis. The current paradigm is that plumes tap the relatively undegassed lower mantle, which is thought to have preserved a high proportion of primitive noble gases [4,5]. In the case of helium

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and neon, these primitive gases may have had solar-like isotopic compositions [5]: however, radiogenic ⁴He and nucleogenic ²¹Ne in-growth over time will have altered the initial ratios. Potentially complicating factors, such as magma mixing, crustal assimilation, and air contamination, can further modify isotopic ratios of a magma body on its path towards the Earth's surface. In most cases, these modifications can be identified since the different sources (e.g., upper mantle, lower mantle, crust and air) have diagnostic helium and neon isotopic ratios.

In the case of helium isotope systematics, ³He is overwhelmingly primordial in origin, whereas ⁴He is the product of U and Th decay. Ocean island basalts (OIB) erupting in plume-related localities such as Hawaii (e.g. [6]) and Iceland [7] have high ³He/⁴He ratios (up to 37 R_A , where R_A is the ratio in air = 1.4×10^{-6}). In contrast, most midocean ridge basalts have ³He/⁴He ratios equal to $8 \pm 1 R_A$ [8,9]. This constancy suggests that the upper mantle source for MORB is homogeneous in composition with respect to helium. Crustal ³He/⁴He ratios are significantly lower than the air value due to the dominance of radiogenic helium.

The three isotopes of neon (²⁰Ne, ²¹Ne and ²²Ne) can be used to resolve atmospheric, primordial and nucleogenic contributions to mantle-derived magmas. ²⁰Ne and ²²Ne are predominantly primordial - trapped during accretion of Earth, whereas ²¹Ne is produced by (α, n) reactions on ¹⁸O [10]. The consequence of producing nucleogenic ²¹Ne is to increase the ²¹Ne/²²Ne ratio over time. In plots of ²⁰Ne/²²Ne versus ²¹Ne/ ²²Ne (three-isotope neon space), both MORB and OIB follow linear arrays from the atmospheric value $({}^{20}\text{Ne}/{}^{22}\text{Ne} = 9.80 \text{ and } {}^{21}\text{Ne}/{}^{22}\text{Ne} = 0.029 - 0.029 \text{ J}$ since a certain degree of atmospheric contamination is inevitable in terrestrial samples) to more nucleogenic ²¹Ne/²²Ne ratios. On such plots, OIB magmas tend to follow a trajectory with a slope steeper than MORB samples (i.e. they show lower ²¹Ne/²²Ne values for a given ²⁰Ne/²²Ne ratio) - this observation has been explained by assuming that the lowermost mantle is less degassed than the source of MORB (i.e., lower mantle contains more primordial ²²Ne) so that the isotopic

shift induced by addition of nucleogenic ²¹Ne is less pronounced. Significantly, because production of ²¹Ne is coupled to that of ⁴He (²¹Ne/ ⁴He= 4.5×10^{-8} ; [11]), an increase in ²¹Ne/²²Ne should be accompanied by a decrease in ³He/ ⁴He. Therefore, since the He–Ne isotope systematics are coupled, it is possible to predict the ³He/ ⁴He through knowledge of the Ne isotope systematics (see [5,12] for details).

Recent work on Icelandic [13-15] (and East Pacific Rise [16]) lavas has challenged the view that He and Ne isotope systematics are necessarily coupled in all mantle-derived materials. The picture that emerges from the Icelandic work is one where the Ne isotope systematics seem to show virtually no addition of nucleogenic ²¹Ne (i.e., the neon is purely solar in composition), vet ³He/⁴He ratios are significantly lower than solar values indicating addition of radiogenic helium. Thus, in three-isotope neon space the gradient of the Icelandic trajectory is much steeper than expected from the helium isotope systematics. In this work, we present another example where the He and Ne isotope systematics are apparently decoupled; however, for the Manus Basin, the decoupling is in the opposite sense. The ${}^{3}\text{He}/{}^{4}\text{He}$ ratios are consistent with mantle plume involvement, yet the neon isotopes show significant addition of nucleogenic ²¹Ne. The resulting gradient in three-isotope Ne space is significantly lower than expected. Such an observation is unique in noble gas studies of mantle plumes. In this contribution, we consider the ramifications of this result for both the development of the Manus Basin magma source region and its degassing history through time.

2. Samples and analytical procedures

Samples selected for this study have all been analysed previously for their ${}^{3}\text{He}/{}^{4}\text{He}$ ratios ([1,2]). They are fresh tholeiitic lavas (<130000 yr old), which are well-characterised for their major and trace element chemistry and radiogenic isotope systematics [17]. They are distinguished into the following seven types: MORB-1, MORB-smt, MORB-2, E-MORB, Arc basalt, back-arc basin basalt (BABB), and Extreme BABB (XBABB) (see discussion in [1,2]). Sample locations and eruption depths are given in Table 1.

Prior to analysis, a binocular microscope was used to select glass samples free of large vesicles, phenocrysts and surface alteration. In all cases, an effort was made to choose a small number of large grains to minimise the surface area to volume ratio. Prior to loading, the glasses were ultrasonically cleaned in a 50:50 acetone-methanol mixture and weighed to ± 0.05 mg.

Neon concentration and isotopic measurements (together with helium abundances) were carried out on a modified VG5400 mass spectrometer [18] using stepped heating as the extraction method. Samples were loaded onto a sample sled and pre-heated under UHV (to 150°C) for several days prior to analysis. Volatiles were extracted in four temperature steps: 600°C, 800°C, 1200°C and 1400°C, each for 30 min duration. Volatiles released during the 600°C steps were not collected since we assume that they are dominated by atmospheric contamination. Similarly, the fusion step (1400°C) was discarded as it contains residual volatiles – generally at blank levels. We report gas yields at the 800°C and 1200°C release steps (Table 1) and use the total concentrations in the following discussion. All results take into account corrections for procedural blanks and contributions to ²⁰Ne and ²²Ne due to doubly charged ⁴⁰Ar and CO₂ respectively. Best-fit regressions of +/++ ratios as a function of H₂ and CO₂ partial pressures were used to estimate the contributions of the doubly charged species (see procedure in [19]). Blanks for ²⁰Ne and ⁴He throughout these experiments were in the range of $8 \pm 1 \times 10^{-13}$ cm³STP and $2 \pm 1 \times 10^{-9}$ cm³STP, respectively which typically represented $\sim 5\%$ of sample yields.

3. Results

A total of 19 glasses were analysed as part of this study. Neon isotopic compositions together with abundances of neon and helium are reported in Table 1.

3.1. Neon isotopes and abundances

Neon concentrations for the various petrogenic rock-types vary over approximately two orders of magnitude as shown in Fig. 1. In this plot, we illustrate the deviation of the neon isotopic composition of a given sample from the air value using the $\delta^{20}Ne$ and $\delta^{21}Ne$ notation (see details in [20]). We note that the arc-type, BABB and XBABB samples all have air-like Ne isotope compositions, regardless of concentration. However, the MORB lavas (particularly MORB-1) show a trend whereby the lowest concentration samples have the highest δ^{20} Ne and δ^{21} Ne values i.e., they are the least contaminated by air Ne. The standard three-isotope neon plot (Fig. 2) provides further illustration that the majority of the Manus Basin samples lie within error of the air value.



Fig. 1. Plot of the neon isotopic enrichment (in %) of Manus Basin samples relative to air as a function of total ²⁰Ne concentration. Neon isotope ratios are plotted in the delta notation where $\delta^i Ne = \{(iNe)^{22}Ne_{sample}/iNe)^{22}Ne_{air})-1\} \times 100$ ($iNe = {^{20}Ne}$ or ${^{21}Ne}$). The dashed line represents air ($\delta^{20}Ne$ and $\delta^{21}Ne = 0$). Errors are given at the 1 σ level.

Table 1 Helium and neon abundances and isotopic ratios

Sample (type), location depth ^a , weight	³ He/ ⁴ He ^b	Т	⁴ He	²⁰ Ne	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne
	$(R/R_{\rm A})$	(°C)	$(10^{-9} \text{ cm}^3 \text{ STP/g})$	$(10^{-12} \text{ cm}^3 \text{ STP/g})$		
32-5 (MORB-1) 3°25.2′S, 149°00.9′E						
2300–2420 m, 282.68 mg	13.5 ± 0.2	800	3390 ± 30	69.0 ± 0.2	9.95 ± 0.03	0.0311 ± 0.0008
		1200	263 ± 2	23.6 ± 0.1	10.19 ± 0.06	0.034 ± 0.001
		Total	3650 ± 30	92.6 ± 0.2	10.01 ± 0.04	0.0318 ± 0.0007
33-3 (MORB-1) 3°31.7′S, 149°28.7′E						
2090–2115 m, 287.60 mg	12.7 ± 0.3	800	5260 ± 50	10.41 ± 0.07	10.8 ± 0.1	0.047 ± 0.004
		1200	669 ± 7	4.54 ± 0.06	11.0 ± 0.2	0.037 ± 0.004
		Total	5930 ± 50	14.94 ± 0.09	10.9 ± 0.1	0.044 ± 0.003
36-2 (MORB-1) 3°26.8′S, 149°57.8′E						
2155–2165 m, 308.67 mg	11.7 ± 0.2	800	8540 ± 70	77.1 ± 0.2	10.10 ± 0.02	0.0329 ± 0.0006
		1200	77.5 ± 0.7	11.21 ± 0.09	10.13 ± 0.06	0.032 ± 0.003
		Total	8620 ± 70	88.3 ± 0.2	10.10 ± 0.04	0.0329 ± 0.0006
38-3 (MORB-1) 3°19.9′S, 150°04.9′E						
2200–2225 m, 298.81 mg	12.7 ± 0.2	800	11700 ± 100	27.7 ± 0.1	10.58 ± 0.07	0.0402 ± 0.002
		1200	28.1 ± 0.2	3.06 ± 0.09	10.2 ± 0.1	0.0349 ± 0.003
		Total	11700 ± 100	30.7 ± 0.2	10.5 ± 0.1	0.0397 ± 0.001
39-1 (MORB-1) 3°17.4′S, 150°07.7′E						
2285–2370 m, 294.95 mg	12.7 ± 0.2	800	3560 ± 30	186.8 ± 0.3	9.85 ± 0.02	0.0295 ± 0.0004
		1200	95.4 ± 0.8	9.6 ± 0.1	9.97 ± 0.07	0.031 ± 0.003
		Total	3660 ± 30	196.4 ± 0.3	9.85 ± 0.03	0.0295 ± 0.0004
31-1 (E-MORB) 3°30.1′S, 149°15.5′E						
2075–2245 m, 311.10 mg	10.3 ± 0.3	800	906 ± 9	52.37 ± 0.09	9.88 ± 0.03	0.0302 ± 0.0009
		1200	63.7 ± 0.6	18.42 ± 0.06	10.16 ± 0.05	0.034 ± 0.002
		Total	970 ± 9	70.8 ± 0.1	9.95 ± 0.03	0.0312 ± 0.0008
23-2 (MORB-smt. ^c) 3°52.4′S, 149°58.0′	Έ					
1390–1865 m, 306.85 mg	15.1 ± 0.3	800	2220 ± 20	67.4 ± 0.1	9.79 ± 0.02	0.030 ± 0.001
		1200	631 ± 6	13.11 ± 0.06	10.06 ± 0.07	0.034 ± 0.002
		Total	2850 ± 20	80.5 ± 0.1	9.84 ± 0.03	0.0304 ± 0.0009
41-1 (MORB-2) 3°12.0′S, 150°12.5′E						
2375–2400 m, 311.70 mg	0.8 ± 0.8	800	3.48 ± 0.05	626.9 ± 0.7	9.85 ± 0.01	0.0291 ± 0.0001
		1200	2.05 ± 0.03	2.01 ± 0.06	10.1 ± 0.2	0.031 ± 0.007
		Total	5.53 ± 0.05	628.9 ± 0.7	9.85 ± 0.02	0.0291 ± 0.0002
42-1 (MORB-2) 3°09.6′S, 150°17.1′E						
2480–2490 m, 313.30 mg	11.4 ± 0.3	800	213 ± 2	10.18 ± 0.05	10.12 ± 0.08	0.029 ± 0.004
		1200	7.86 ± 0.08	15.84 ± 0.06	9.91 ± 0.06	0.025 ± 0.002
		Total	221 ± 2	26.02 ± 0.07	9.99 ± 0.06	0.027 ± 0.002
44-1 (MORB-2) 3°05.2′S, 150°23.7′E						
2600–2630 m, 287.05 mg	12.1 ± 0.2	800	$35/0 \pm 30$	135.2 ± 0.3	9.89 ± 0.02	0.0303 ± 0.0005
		1200	18.7 ± 0.2	24.2 ± 0.1	9.85 ± 0.06	0.029 ± 0.001
		Total	3590 ± 30	159.3 ± 0.3	9.88 ± 0.03	0.0301 ± 0.0005
45-3 (MORB-2) 3°03.9′S, 150°27.3′E	127102	000	1100 + 10	527101	0.(1.1.0.02	0.020 1.0.001
2570–2670 m, 297.68 mg	12.7 ± 0.2	800	1190 ± 10	52.7 ± 0.1	9.61 ± 0.02	0.030 ± 0.001
		1200	110.1 ± 0.9	5.34 ± 0.09	10.5 ± 0.1	0.041 ± 0.005
20 1 (DADD) 2020 4/5 140040 4/5		Total	1300 ± 10	58.0 ± 0.2	9.69 ± 0.04	0.031 ± 0.001
20-1 (BABB) 3°39.4°S, 149°40.4°E	12.0 ± 0.2	800	2010 ± 20	20.1 ± 0.1	0.05 ± 0.05	0.020 ± 0.001
2370–2440 m, 285.30 mg	12.0 ± 0.3	800 1200	2010 ± 20 5 17 ± 0.05	28.1 ± 0.1	9.95 ± 0.05	0.029 ± 0.001
		Tetal	3.17 ± 0.03 2020 ± 20	0.37 ± 0.09 28.6 ± 0.1	10.7 ± 0.4 0.06 ± 0.0	0.03 ± 0.01 0.020 + 0.001
		rotai	2020 ± 20	20.0 ± 0.1	2.20 - 0.2	0.022 ± 0.001

Sample (type), location depth ^a , weight	3 He/ 4 He ^b ($R/R_{\rm A}$)	Т (°С)	⁴ He (10 ⁻⁹ cm ³ STP/g)	20 Ne (10 ⁻¹² cm ³ STP/g)	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne
2445–2510 m, 283.40 mg	12.1 ± 0.3	800	1130 ± 10	37.1 ± 0.1	9.96 ± 0.05	0.028 ± 0.002
		1200	0.38 ± 0.02	0.90 ± 0.09	10.4 ± 0.3	0.05 ± 0.02
		Total	1140 ± 10	38.0 ± 0.1	9.97 ± 0.08	0.029 ± 0.002
40-6 (BABB) 3°14.9'S, 150°07.7'E						
2300–2310 m, 305.60 mg	11.5 ± 0.4	800	237 ± 2	40.8 ± 0.1	9.91 ± 0.03	0.026 ± 0.001
		1200	4.09 ± 0.04	0.42 ± 0.09	10.5 ± 0.4	0.01 ± 0.01
		Total	241 ± 2	41.2 ± 0.1	9.91 ± 0.06	0.026 ± 0.001
41-3 (BABB) 3°12.0′S, 150°12.5′E						
2375–2400 m, 308.10 mg	11.6 ± 0.8	800	44.1 ± 0.4	15.9 ± 0.1	9.78 ± 0.06	0.027 ± 0.002
		1200	0.87 ± 0.02	0.51 ± 0.09	10.0 ± 0.3	0.027 ± 0.02
		Total	45.0 ± 0.4	16.5 ± 0.1	9.8 ± 0.1	0.027 ± 0.002
14-5 (arc-type) 3°42.9'S, 152°10.4'E						
1755–1950 m, 318.10 mg	1.0 ± 0.2	800	1.90 ± 0.03	264.5 ± 0.3	9.76 ± 0.02	0.0289 ± 0.0004
		1200	1.76 ± 0.03	0.4 ± 0.1	10.5 ± 0.4	0.042 ± 0.009
		Total	3.67 ± 0.04	264.9 ± 0.3	9.76 ± 0.02	0.0290 ± 0.0004
17-1 (arc-type) 3°44.4'S, 151°38.8'E						
1685–1860 m, 308.50 mg	0.6 ± 0.3	800	3.31 ± 0.05	136.1 ± 0.3	9.86 ± 0.2	0.0288 ± 0.0004
		1200	2.89 ± 0.05	0.92 ± 0.05	10.7 ± 0.4	0.033 ± 0.007
		Total	6.20 ± 0.07	137.0 ± 0.2	9.87 ± 0.03	0.0288 ± 0.0004
19-12 (BABB s.r.) 3°45.1'S, 151°09.5'E						
2625–2635 m, 289.50 mg	3 ± 2	800	14.1 ± 0.2	7.03 ± 0.05	9.91 ± 0.09	0.031 ± 0.002
		1200	2.43 ± 0.05	1.04 ± 0.07	10.6 ± 0.5	0.03 ± 0.01
		Total	16.5 ± 0.2	8.07 ± 0.09	10.0 ± 0.2	0.031 ± 0.003
24-9 (XBABB) 3°32.7′S, 149°51.4′E						
2155–2185 m, 299.45 mg	8.7 ± 0.4	800	9.5 ± 0.1	405.8 ± 0.6	9.82 ± 0.01	0.0290 ± 0.0002
		1200	7.55 ± 0.07	0.5 ± 0.1	10.3 ± 0.3	0.03 ± 0.02
		Total	17.1 ± 0.1	406.3 ± 0.6	9.82 ± 0.02	0.0290 ± 0.0002

All errors are given to 1σ except for the ³He/⁴He ratios, which are reported at the 2σ level.

^a Start and finish depth of dredge.

^b From [1,2].

Table 1 (continued)

^c Sample 23-2 was dredged from a seamount.

However, we note that seven samples (five MORB-1, one E-MORB and one MORB-smt) are displaced from air, and lie on a linear array (dashed line in Fig. 2) passing through the atmospheric value. The correlation line defined by the seven samples has a gradient less than that of the MORB correlation line, i.e. samples show higher 21 Ne/ 22 Ne ratios relative to MORB [3] for a given 20 Ne/ 22 Ne ratio. This observation is in stark contrast to other high 3 He/ 4 He samples, e.g. from Iceland, Loihi, Samoa, the Kola Peninsula and Reunion [6,13–15,21–23], that consistently fall on correlation lines with slopes steeper than the MORB Ne-trend [5]. We believe that this is the

first observation of oceanic lavas having high ³He/ ⁴He ratios and such nucleogenic neon isotope ratios.

3.2. Helium isotopes and abundances

As noted previously [1,2], the 3 He/ 4 He ratios of the Manus Basin samples are generally higher than the MORB-value of $8 \pm 1 R_{A}$. The MORBtype samples range from 10.3 to 15.1 R_{A} (with the exception of one sample, 41-1, which is air-like), and the BABB values range from 11.5 to 12.1 R_{A} (apart from one sample from the Southern Rift at 3 R_{A}). One XBABB sample has a 3 He/ 4 He ratio



Fig. 2. Traditional three-isotope neon plot showing seven Manus Basin samples (filled symbols) trending towards an endmember composition more nucleogenic than typical MORB. The Manus trend (dotted line) is represented by the equation $({}^{20}\text{Ne}/{}^{22}\text{Ne}) = 70.352({}^{21}\text{Ne}/{}^{22}\text{Ne}) + 7.756$ ($r^2 = 0.994$). This line was calculated from an error-weighted regression through the seven data points and the air value. Also shown for comparison is the MORB trend [3] (${}^{20}\text{Ne}/{}^{22}\text{Ne} = 85.06({}^{21}\text{Ne}/{}^{22}\text{Ne}) + 7.47$). Errors are reported at the 1 σ level.

of 8.7 R_A and the arc-type samples have air-like ${}^{3}\text{He}/{}^{4}\text{He}$ values.

The helium abundances (Table 1) are highly variable ranging from 4 to 11700 ncm³ STP/g. In the majority of cases, most of the helium was released at the 800°C step, which we take to represent the vesicle-sited gas. The distribution of He between vesicles and glass along with its relationship to CO_2 will be considered elsewhere (Shaw et al., manuscript in preparation).

4. Discussion

A fundamental tenet of the solar neon hypothesis [5] is that the He and Ne isotope systematics are coupled. Therefore, for a given Ne isotopic composition, a corresponding ³He/⁴He ratio can be calculated (see methodology in [5]). Using the correlation line of samples in this work (Fig. 2) and assuming coupled He–Ne systematics (where ³He/²²Ne=7.7 as in [12]), we would predict the Manus Basin samples to have a ³He/⁴He ratio of 4.6 R_A . The average ³He/⁴He ratio of these seven samples, however, is 12.7 R_A suggesting

an apparent decoupling between the He and Ne isotope systematics. An alternative way of viewing this apparent decoupling is to calculate the difference in ²¹Ne/²²Ne between the Manus sample trend and the value predicted assuming the solar neon hypothesis for a ${}^{3}\text{He}/{}^{4}\text{He}$ ratio of 12.7 R_{A} (Fig. 3 shows this schematically). Using this approach, we calculate a shift in the 21 Ne/ ${}^{\overline{22}}$ Ne ratio $(\Delta^{21}\text{Ne}/^{22}\text{Ne})$ of 0.035 assuming a solar $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of 13.8. If the solar neon component in Earth originated from a trapped meteoritic component $({}^{20}\text{Ne}/{}^{22}\text{Ne} = 12.5)$ rather than a pristine solar component (20 Ne/ 22 Ne = 13.8) then the calculated value of the shift would be lower (see [24,25] for a detailed discussion of Earth's mantle neon component); however, the trajectories in three-isotope neon space would be the same. In either case, it appears that the Manus Basin samples are characterised by anomalously nucleogenic neon.

In the following discussion, we investigate five possible mechanisms by which this nucleogenic neon component could have been produced: (1) crustal contamination; (2) devolatisation of the Solomon Sea plate; (3) the addition of slabderived neon from ancient recycled oceanic litho-



Fig. 3. Neon isotope plot showing the Manus trend (dashed line), which would extrapolate to a ²¹Ne/²²Ne ratio of 0.0859 at a mantle source (solar) value of ²⁰Ne/²²Ne = 13.8 [37]. In contrast, we would anticipate an extrapolated ²¹Ne/²²Ne value of 0.0514 for a ³He/⁴He ratio of 12.7 R_A (the mean value of the seven Manus samples which define the Manus trend) assuming the solar Ne hypothesis [5]. Thus, the Manus Basin samples are more nucleogenic than expected by Δ^{21} Ne/²²Ne = 0.035.

sphere; (4) sampling from a previously degassed source, or (5) a heterogeneous deep mantle plume component preserved since Earth's accretion.

4.1. Crustal contamination

The dominant reaction producing ²¹Ne is the (α,n) reaction on ¹⁸O. This reaction is particularly important in crustal lithologies due to the relatively high abundance of α -emitters (U- and Th-decay series nuclides). There are three possible mechanisms by which crustal contamination can occur: (1) in situ post-eruptive in-growth (2) magma ageing, and (3) wallrock contamination.

A potential means of adding nucleogenic neon to the Manus Basin lavas is in situ production since the time of eruption. Using a maximum age estimate of 130 000 yr [17], a U concentration of 0.04 ppm (based on average values in Manus lavas [17]), together with the ⁴He production rate $(2.15 \times 10^{-7} \text{ cm}^3 \text{STP/ppm U/Myr [26]})$ and the ²¹Ne/⁴He production ratio (4.5×10^{-8} [11]), we calculate 5.03×10^{-17} cm³STP ²¹Ne/g can be produced. This amount is <5% of the lowest concentration of excess nucleogenic ²¹Ne in any of the 7 samples lying along the so-called Manus Ne trajectory (samples 23-2 has 0.11×10^{-14} cm^3STP/g – see below for details how this value is calculated). Therefore, due to the young age of these lavas, we can discount post-eruptive ingrowth as a possible means of altering the ²¹Ne/ ²²Ne ratio.

In the second case, radiogenic/nucleogenic gases may be added to magma prior to eruption via the process of magma ageing in crustal magma chambers [27]. We evaluate this possibility with reference to one of the seven samples lying along the so-called Manus Ne trajectory (sample 33-3 has a total amount of ${}^{21}\text{Ne} = 6.0 \times 10^{-14} \text{ cm}^3\text{STP/g}$). First, we calculate the proportion of air Ne in the sample assuming that its measured ²⁰Ne/ 22 Ne ratio (10.9) reflects mixing between air (9.8) and 'mantle+nucleogenic' Ne (²⁰Ne/ 22 Ne = 13.8). The fraction of air Ne in the sample = 0.73which means approximately 1.66×10^{-14} cm³STP/g (0.27 of the total) is comprised of mantle neon+21Ne excess. We can further resolve this non-air ²¹Ne into mantle ²¹Ne

 (0.99×10^{-14}) $cm^3 STP/g$) excess 21 Ne and $(0.67 \times 10^{-14} \text{ cm}^3 \text{STP/g})$ assuming that the mantle component has ${}^{20}\text{Ne}/{}^{22}\text{Ne} = 13.8$ and a ${}^{21}\text{Ne}/{}^{22}$ 22 Ne = 0.05146 – the predicted value from the solar hypothesis for a measured ³He/⁴He ratio of 12.7 R_A . Because of the constant ²¹Ne/⁴He production ratio of 4.5×10^{-8} , addition of this amount of ²¹Ne would simultaneously add 0.15×10^{-6} cm³STP ⁴He/g. A residence or ageing time of 17.2 Ma is required to produce this much ⁴He (and ²¹Ne) assuming a [U] of 0.04 ppm [17]. Calculated residence times vary between 21 Ma (samples 36-2 and 38-3) and 2.8 Ma (sample 23-2) for the other lavas which lie along the Manus trajectory (Fig. 2). These values are minimum estimates as magmatic degassing could have occurred simultaneously with production of nucleogenic ²¹Ne over these calculated ages. These times are far in excess of postulated crustal residence ages of magma, which are generally on the order of $< 10^5$ yr [28,29]. Moreover, such long residence times are not consistent with the relatively young age of opening for the Manus spreading centre (estimated at 0.78 Ma [30]).

The third possibility involves addition of nucleogenic neon through crustal assimilation as magma ascends to the surface. This scenario also seems unlikely given that all samples lying along the 'nucleogenic MORB trajectory' (Fig. 2) were erupted along an active spreading centre or within an extensional zone. At such localities, the opportunity for crustal interaction appears minimal and, in any case, not greater than for mid-ocean spreading centres - where there is no evidence of such nucleogenic neon additions. Furthermore, even if crustal assimilation had occurred, the oceanic crust where these lavas are sited is young (<1 Ma) and unlikely to have grown-in and stored sufficient ⁴He and ²¹Ne to modify source ³He/⁴He and ²¹Ne/²²Ne ratios.

4.2. Devolatisation of the Solomon Sea slab

Another possibility for the source of the nucleogenic neon is the down-going Solomon Sea plate. The source of the neon associated with subduction-related processes could be fluids or melts derived from either subducted sediments or the underlying oceanic crust. In the case of sediments, noble gas data obtained on a variety of sediment types of different ages [31] indicate the overwhelming dominance of air-like isotope signatures. Therefore, even with prolonged ageing, an air-like neon signature would be expected to dominate over any neon produced in situ. For subducted sediments to be considered a viable possibility, the in situ-produced Ne component would have to be retained preferentially in the sediment matrix while the air-like component would have to be lost. One potential mechanism that could effect this separation would be loss of the airlike Ne via pore fluids which are expected to be expelled during sediment burial and diagenesis. The question then becomes whether the sediment matrix could still retain the nucleogenic Ne component. Diffusion experiments on interplanetary dust particles (IDP) in deep-sea sediments [32] suggest that neon is extremely mobile in sedimentary material at relatively moderate temperatures (200-500°C). If the diffusivity of Ne in sediments is similar to that in IDPs, it suggests that trapped (in situ-produced) Ne would be completely released from sediments at relatively shallow subduction depths, and probably able to escape to the forearc region. Previous studies have emphasised the difficulty of subducting helium [33]: the above considerations suggest that this may also be the case for neon.

The subducted oceanic crust is also an improbable candidate for the nucleogenic Ne signal for the same reasons. First, any intrinsic volatiles would be expected to have been modified during seawater alteration to atmospheric-like values. Indeed, if the analogy of on-land ophiolite complexes is appropriate for oceanic crust, the zone of alteration may extend to depths of 3–5 km (e.g. [34]). Therefore, a significant fraction of the uppermost oceanic crust may have an air-like neon isotope composition. Second, we would have to invoke the scenario of preferential loss of the air-like Ne component, and the retention of the nucleogenic neon despite heating by subduction which would accentuate diffusional loss.

Perhaps the most persuasive argument against a slab origin for the nucleogenic neon in this study is that the most pronounced ²¹Ne excess signal is

observed in the MORB Type-1 lavas (i.e., the four highest ²¹Ne/²²Ne ratios along the Manus trend are found in MORB-1 lavas). Although some geochemical support for slab involvement is apparent in the Manus sample suite as a whole [17], the MORB-1 magmas are the magma-type least affected by subduction-related processes and it is these samples that possess the most nucleogenic neon.

4.3. Ancient recycled slab

The nucleogenic neon signal could result from the addition of slab-derived neon from ancient oceanic lithosphere, recycled via the deep mantle and entrained by the Manus plume en-route to the surface. A recycled altered oceanic crustal component has been suggested to explain anomalously low δ^{18} O values (5.3–5.8%) found in the same samples [2]. In this scenario, the original noble gas signature (along with other volatiles) of the recycled slab would have been lost in the so-called 'subduction barrier' [31] but over time the slab would acquire a nucleogenic signature through in situ production.

To assess the plausibility of this suggestion, we have modelled the above scenario using an estimate of the ²¹Ne concentration in 2-Ga-old subducted slab. We calculate that 1.3×10^{-12} cm³STP ²¹Ne/g would be produced in 2 Ga assuming a [U] value of 0.066 ppm for altered oceanic crust [35], a ²¹Ne/⁴He production ratio of 4.5×10^{-8} [11], a ⁴He production rate of 2.15×10^{-7} cm³STP/ppm U/Myr (which assumes a Th/U ratio of 3.3 and includes ⁴He produced from Th decay in its production rate; see [26] for details) and 100% retention of produced ²¹Ne. If the initial (mantle) plume ²¹Ne concentration was 3.35×10^{-12} cm^3 STP/g [36] with a ²¹Ne/²²Ne ratio of 0.032 [37] then a slab/plume mixing ratio of ~ 4.4 is required to produce a resultant ²¹Ne/²²Ne ratio of 0.0859. This slab/plume ratio could be as low as ~ 1 (i.e. a mixture of 50% slab and 50% plume components) if the altered oceanic crust had a U content of 0.3 ppm [38].

Both estimates of the slab/plume mixing ratio are exceptionally high. For comparison, Marcantonio et al. [39] used Os–Pb relationships of lavas



Fig. 4. Plot showing the relationship between degree of degassing and the required time since degassing (separation time) in order to produce a Δ^{21} Ne/²²Ne = 0.035 for a MORB source (solid line) and plume source (dashed line). Initial source (U/²²Ne)_i ratios from Porcelli and Wasserburg [36].

erupted in the Canary Islands to constrain the contribution of ancient recycled crust to this (HIMU-type) mantle source to between 15 and 30%. In contrast, the Manus Basin lavas have a maximum ²⁰⁶Pb/²⁰⁴Pb ratio of 18.8 [17] which is considerably lower than that found at the Canaries. Therefore, if the recycled component is traced by radiogenic lead ($^{206}Pb/^{204}Pb > 20$), the Manus Basin data imply that the proportion of ancient slab must be considerably lower (< 10%). Similarly, Macpherson et al. [2] used O-isotopes to limit the contribution of a deep hydrothermally altered oceanic crust component to < 35%. Again this estimate is substantially less than the above values (50-85%) based on our Ne isotope results. More significantly, if the proportion of ancient slab material was as large as calculated above, the present sample suite would be expected to possess exceptionally strong slab-derived trace element (e.g., La/Nb) signals - these are conspicuously absent in the Manus Basin lavas.

4.4. A previously degassed plume source

During partial melting of a mantle source, noble gases are expected to act as incompatible trace elements partitioning into the melt phase as a function of their relative partition coefficients (D). Experimental data indicate that the partition coefficients of He and Ne are similar ($D_{\text{He}} = 0.02-0.7$, $D_{\text{Ne}} = 0.03-0.6$ [40] and references therein; and $D_{\text{He}} \approx D_{\text{Ne}}$, [41]). This implies that during the formation of melt, little or no fractionation between helium and neon is expected. However, this is not the case during formation of a free vapour phase. During melt-vapour partitioning (degassing), Ne will partition preferentially into the vapour phase, as its solubility (S) in a tholeiitic melt is less than that of helium ($S_{\text{He}}/S_{\text{Ne}} \sim 2$, [42]; $S_{\text{He}}/S_{\text{Ne}} \sim 10$, [43]). The consequence of a degassing event, therefore, is to increase the He/Ne ratio in the degassed melt compared with its starting value.

Another corollary of volatile loss is that the intrinsic Ne and He concentrations in the degassed melt would be reduced relative to U and Th: therefore, as a function of time and extent of degassing, ³He/⁴He ratios would decrease and ²¹Ne/²²Ne ratios increase in the melt phase residual after degassing. Thus, the original neon isotope values could be altered more readily than those of helium due to the lower (residual) neon contents compared to helium. Such a scenario has been invoked to explain anomalously nucleogenic neon in samples from the North Chile Ridge [44] although, in that case, the ³He/⁴He ratios were less than MORB. Here, we explore whether this type of model can explain the nucleogenic neon measured in the Manus Basin samples.

The principal point to consider is whether the plume-degassing hypothesis can account for the shift in ²¹Ne/²²Ne ($\Delta = 0.035$) in reasonable time periods and/or degrees of degassing without drastically altering the helium isotope systematics. In Fig. 4, we plot degree of degassing versus the time required following degassing (the separation time) to grow-in the amount of ²¹Ne to produce the requisite $\Delta^{21} \text{Ne}/^{22} \text{Ne}$. We see that for a plume source $(U/^{22}Ne \sim 3500 \text{ [36]})$ the grow-in time would be 1 Ga if degassing of initial Ne was 100-fold, or 100 Ma if degassing was 1000-fold. For comparison, the separation times for a MORB-type source (U/²²Ne ~ 10^5 ; [36]) would be 36 Ma and 3.6 Ma for 100- and 1000-fold degassing factors respectively (because Manus Ba- $\sin^{3}\text{He}^{4}\text{He}$ ratios \gg MORB such a source is clearly inappropriate in the present case). Since ²¹Ne in-growth is accompanied by ⁴He production, the ${}^{3}\text{He}/{}^{4}\text{He}$ ratio of the source will decrease ²¹Ne/²²Ne simultaneously with increasing although to a lesser extent due to greater retention of helium after the degassing event (helium is more soluble in basaltic melt than neon). Assuming a Rayleigh 'open-system' style of degassing and using the above degassing factors ($\times 100$ and $\times 1000$) with associated grow-in times for a plume source (1 Ga and 100 Ma respectively), an initial ³He/⁴He ratio of 30 R_A (with 5.6×10⁻⁵ cm³STP ⁴He/g [36]) will decrease to 26.6 R_A or 29.5 R_A respectively for a $S_{\text{He}}/S_{\text{Ne}} = 10$ [43], and to 16.7 R_A or 24.0 R_A respectively for a S_{He} / $S_{\rm Ne} = 2$ [42]. Increased separation times and/or degassing factors will lower these ³He/⁴He ratios even further.

As illustrated above, production of nucleogenic neon (plus retention of ${}^{3}\text{He}/{}^{4}\text{He}$ ratios > MORB) by the plume-degassing mechanism depends upon two key factors: (1) the extent of degassing and (2) the separation time. There is evidence to suggest that helium loss during plume-degassing can be extensive. For example, Hilton et al. [45] estimated that the (plume) source region of Kilauea volcano (15 R_A) is degassed by a factor of 2000 compared to model-dependent values for a lower mantle source [36]. Thus, one of the key constraints of the plume-degassing hypothesis under consideration, i.e., the requirement of significant degrees of degassing, may be a feature common to the process of plume impingement with oceanic lithosphere. The second factor is the separation time, which can be relatively short provided the degassing factor is sufficiently high. For example, a conservative 100-fold decrease in plume He (corresponding to 10⁴ decrease in Ne for a Rayleigh distillation with $S_{\text{He}}/S_{\text{Ne}} = 2$) would require a separation time of only 10 Ma in order to achieve a Δ^{21} Ne/²²Ne = 0.035. From these considerations, therefore, we suggest that the degassing factor and/or separation times required to produce the nucleogenic neon in the Manus Basin lavas fall within reasonable limits.

The other major constraint is the requirement that previously degassed melt forms part of the source region. Macpherson and Hall [46] have tracked the Manus Basin plume from the middle Eocene to the present-day, and it appears to have experienced a complex history of interaction with plate boundaries in the western Pacific. Although speculative, we suggest that plume material, which was extensively but incompletely degassed (therefore able to retain high ³He/⁴He ratios), could have been retained in the mantle in or around the upwelling plume for the relatively short time periods suggested above to produce the requisite Δ^{21} Ne/²²Ne. Therefore, we conclude that plumedegassing (following melting but prior to eruption) could provide a viable means by which to effectively decouple He-Ne systematics with the superimposition of a nucleogenic neon signal onto a plume-like ³He/⁴He ratio.

4.5. Lower mantle heterogeneity

Finally, we consider the possibility that the mantle source supplying the Manus Basin has distinct He-Ne systematics compared to other plume sources. Recent work [13-15] on the Icelandic plume has identified cases of apparently decoupled He-Ne isotope systematics: solar-like Ne isotope compositions are accompanied by 3 He/ 4 He ratios ranging from 12–29 $R_{\rm A}$ – significantly lower than estimated solar ³He/⁴He ratios (145 R_A [15]). This observation has been attributed to either a high concentration of solar Ne in the source, which has overwhelmed nucleogenic ²¹Ne, or extremely low (U+Th) source contents. The Manus Basin glasses uniquely reveal a decoupling in the opposite sense: the ³He/⁴He ratios preserve evidence of a plume contribution whereas the ²¹Ne/²²Ne systematics are more evolved, consistent with a source having lower solar Ne contents and/or high U and Th.

A simple test of whether the Manus Basin lavas sample a distinct source is to consider their ${}^{3}\text{He}/{}^{22}\text{Ne}$ ratio in comparison to other high- ${}^{3}\text{He}/{}^{4}\text{He}$ lavas. Following the approach of Moreira et al. [15], the ${}^{3}\text{He}/{}^{22}\text{Ne}$ ratio of the Manus Basin source can be calculated from the following equation:

$$\{({}^{4}\text{He}/{}^{3}\text{He})_{Manus} - ({}^{4}\text{He}/{}^{3}\text{He})_{solar}\} =$$

$$({}^{4}\text{He}/{}^{21}\text{Ne})^{*}\{({}^{21}\text{Ne}/{}^{22}\text{Ne})_{\text{Manus}} -$$

 $({}^{21}\text{Ne}/{}^{22}\text{Ne})_{\text{solar}} \times ({}^{22}\text{Ne}/{}^{3}\text{He})$

 $(^{21}\text{Ne}/^{4}\text{He})^{*} = 4.5 \times 10^{-8}$ [11]. ⁴He/ where ${}^{3}\text{He}_{\text{solar}} = 5000$ [15], ${}^{21}\text{Ne}/{}^{22}\text{Ne}_{\text{solar}} = 0.032$ [37], ${}^{3}\text{He}/{}^{4}\text{He}_{\text{Manus}} = 12.7 R_{\text{A}}$ (this work) and ${}^{21}\text{Ne}/{}^{4}$ 22 Ne_{Manus} = 0.0859 (this work). In this way, we determine the ³He/²²Ne ratio of the Manus Basin source to be 23.4. This value is much higher than estimates of 4 for Iceland, 9 for Loihi Seamount and 11 for MORB-mantle (see [15]). It implies that the plume source sampled by Manus Basin lavas is highly fractionated in its He-Ne elemental abundance compared to the Icelandic and Loihi plume sources. Clearly, the fact that its He/Ne ratio is higher than that of other mantle sources (including MORB mantle) is why its ²¹Ne/²²Ne ratio is more susceptible to change than its ³He/ ⁴He ratio.

One possible process that can fractionate He from Ne is diffusion: however, this mechanism would require a helium and/or neon concentration gradient between the Manus Basin source region and ambient mantle. We suggest that for a significant portion of the history of the Manus plume, assuming a lower mantle derivation, it would have a helium concentration greater than that of ambient (upper) mantle [36]. In this case, a transient lowering of the ³He/²²Ne ratio in the plume would occur by diffusive exchange with lower concentration ambient mantle due to the higher diffusivity of He [47]. Degassing of the plume would lead to lower He concentrations and thus eventually to a He-Ne fractionation in the opposite sense, i.e. a transient increase in the plume ³He/²²Ne ratio. However, if the estimated separation time (~ 10 Ma) is correct, He would diffuse a maximum of only 1.8 m (for a diffusion coefficient $(D_{\rm He}) \sim 10^{-4}$ cm²/s [26]) and thus a negligible volume of degassed plume would be affected. We conclude, therefore, that diffusion is not a viable mechanism by which to change the He/Ne ratio of the Manus magma source.

If diffusion fails to account for the high He/Ne ratios, the alternative is that the high He/Ne (Manus plume) ratio is inherently representative of its source region in the lowermost mantle. If

correct, then this feature would have to have been (1) a primary feature of the Earth accretion process, (2) resistant to >4 Ga of convective mantle mixing, and (3) sampled uniquely by the Manus Basin plume. Should these conditions prevail, there are important implications for modelling the means by which Earth acquired its noble gases. Two models - both variants of the magma ocean hypothesis (e.g., [48]) have been suggested as capable of fractionating mantle He/Ne ratios. In the first case, noble gases acquired during accretion would have degassed from the magma ocean as a function of their relative solubilities. The greater solubility of He in silicate melts would result in preferential degassing of Ne. This hypothesis is supported by the fact that mantle-derived materials have an average ${}^{3}\text{He}/{}^{22}\text{Ne}$ (7.7) which is twice that of the solar ratio (3.8)[49], consistent with equilibrium degassing models assuming $S_{\text{He}}/S_{\text{Ne}} = 2$ [42]. In the Manus Basin case, the high He/Ne ratio $({}^{3}\text{He}/{}^{22}\text{Ne} = 23.4)$ of the source could be achieved only by both extensive and open-system style degassing. For example, to increase the ³He/²²Ne ratio from the solar value to 23.4 in the (residual) magma ocean by Rayleigh fractionation would involve loss of 84% of the original helium (assuming $S_{\text{He}}/S_{\text{Ne}} = 2$ [42]).

The second possibility is that the Earth acquired its noble gases by ingassing of solar noble gases from a massive proto-atmosphere [50]. In this scenario, once the Earth had reached sufficient size it would become capable of capturing rare gases gravitationally from the solar nebula, thus forming a massive atmosphere of solar gases. The accretional energy along with a heat-trapping atmosphere would provide the heat necessary to induce melting of the growing Earth [51]. The solar rare gases from the atmosphere would have dissolved into a molten silicate Earth as a function of their solubilities [52].

Consequently, due to the greater solubility of He, ingassing would lead to an enhanced He/Ne ratio in the Earth's mantle. Again, we would have to invoke an open-system style of ingassing to produce such a high Manus Basin ³He/²²Ne ratio. Honda and McDougall [49] discuss the relative merits of the two alternative models: at present, however, there are insufficient grounds for favour-

ing one model over the other. Therefore, at this stage, we suggest that the magma ocean hypothesis (either variant) could also be the potential cause of the high ³He/²²Ne ratios found in the Manus Basin. Further studies on the Ne isotope systematics of plume-related materials may reveal if the presence of nucleogenic neon is a wide-spread phenomenon giving greater credence to this heterogeneous mantle hypothesis.

5. Summary

This study of the Manus Basin shows new evidence for a mantle source in which the helium and neon isotope systematics are decoupled. Significantly, the gradient of the Manus slope in three-isotope neon space is lower than that of the MORB line, a unique observation for high ${}^{3}\text{He}/{}^{4}\text{He}$ lavas. A mechanism is required to enrich the source in nucleogenic neon, without obscuring the plume signature manifested in the ${}^{3}\text{He}/{}^{4}\text{He}$ ratios (>MORB).

We have assessed a number of possible scenarios to explain these observations. Post-eruptive contamination (by in-growth of ²¹Ne) cannot produce sufficient nucleogenic neon to affect measured ²¹Ne/²²Ne values. Crustal contamination immediately prior to eruption was dismissed also due to the fact that the crust through which magma erupts is young and therefore lacking in a discernible nucleogenic neon signature. Likewise, magma residence times would have to be unrealistically large if the nucleogenic neon signal were to be grown-in through magma ageing. Although a slab component is present in some Manus Basin lavas, it is unlikely that slab-related noble gases would survive the subduction process to erupt at the Manus spreading centre. Addition of ancient recycled crust to a plume-like source was also evaluated but the proportion of ancient crust required would be too large to account for the observed geochemical traits of the Manus Basin lavas.

We conclude, therefore, that the source supplying the Manus Basin must have a highly fractionated He/Ne ratio (at least twice that of the MORB source) to account for the nucleogenic neon. We present two scenarios to explain this feature. Either the Manus Basin mantle has undergone extensive degassing followed by a period of in-growth of ²¹Ne, or the fractionated He–Ne systematics were inherited during the accretion process. In the first case, plume material may have undergone degassing through interaction with the oceanic lithosphere and become re-entrained in a region of plume upwelling.

Alternatively, He and Ne may be heterogeneously distributed in the source region of mantle plumes, presumably as a result of the accretion process. Additionally, this region of anomalous mantle must have survived ~4 Ga of mantle convection processes acting to homogenise the source. Gas partitioning between a magma ocean and proto-atmosphere has the potential to produce the requisite ${}^{3}\text{He}/{}^{22}\text{Ne}$ ratio but would involve an open-system degassing/ingassing mechanism. At present, there are insufficient grounds for distinguishing between the two possibilities.

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