

2013

Heterogeneities from the first 100 million years recorded in deep mantle noble gases from the Northern Lau Back-arc Basin

Maria K. Peto

Sujoy Mukhopadhyay

See next page for additional authors

Follow this and additional works at: <https://digitalcommons.uri.edu/gsofacpubs>

**The University of Rhode Island Faculty have made this article openly available.
Please let us know how Open Access to this research benefits you.**

This is a pre-publication author manuscript of the final, published article.

Terms of Use

This article is made available under the terms and conditions applicable towards Open Access Policy Articles, as set forth in our [Terms of Use](#).

Citation/Publisher Attribution

Heterogeneities from the first 100 million years recorded in deep mantle noble gases from the Northern Lau Back-arc Basin. Petö, M., S. Mukhopadhyay, and K. Kelley, *Earth and Planetary Science Letters* 369-370, 13-23, doi:10.1016/j.epsl.2013.02.012, 2013.
Available at: <http://dx.doi.org/10.1016/j.epsl.2013.02.012>

This Article is brought to you for free and open access by the Graduate School of Oceanography at DigitalCommons@URI. It has been accepted for inclusion in Graduate School of Oceanography Faculty Publications by an authorized administrator of DigitalCommons@URI. For more information, please contact digitalcommons@etal.uri.edu.

Authors

Maria K. Peto, Sujoy Mukhopadhyay, and Katherine A. Kelley

1 **Heterogeneities from the first 100 million years recorded in deep mantle noble**
2 **gases from the Northern Lau Back-arc Basin**

3

4 **Maria K. Petó^{1,*}, Sujoy Mukhopadhyay¹, Katherine A. Kelley²**

5 1. Dept. of Earth and Planetary Sciences, Harvard University, Cambridge, MA

6 2. Graduate School of Oceanography, University of Rhode Island, Narragansett, RI

7 * Corresponding author (email address: mpeto@fas.harvard.edu)

8

9

10 Abstract

11 Heavy noble gases (Ne, Ar, Xe) can record long-lasting heterogeneities in the mantle
12 because of the production of isotopes from extant (^{238}U , ^{40}K) and extinct (^{129}I and ^{244}Pu)
13 radionuclides. However, the presence of ubiquitous atmospheric contamination, particularly for
14 ocean island basalts (OIBs) that sample the Earth's deep mantle, have largely hampered precise
15 characterization of the mantle source compositions. Here we present new high-precision noble
16 gas data from gas-rich basalts erupted along the Rochambeau Rift in the northwestern corner of
17 the Lau Basin. The strong influence of a deep mantle plume in the Rochambeau source is
18 apparent from low $^4\text{He}/^3\text{He}$ ratios down to 25,600 ($^3\text{He}/^4\text{He}$ of 28.1 R_A). We find that the
19 Rochambeau source is characterized by low ratios of radiogenic to non-radiogenic nuclides of
20 Ne, Ar, and Xe (i.e., low $^{21}\text{Ne}/^{22}\text{Ne}$, $^{40}\text{Ar}/^{36}\text{Ar}$, and $^{129}\text{Xe}/^{130}\text{Xe}$) compared to the mantle source
21 of mid-ocean ridge basalts (MORBs). Additionally, we observe differences in elemental
22 abundance patterns between the Rochambeau source and the mantle source of MORBs as
23 characterized by the gas-rich popping rock from the Mid-Atlantic Ridge. However, the $^3\text{He}/^{22}\text{Ne}$
24 ratio of the Rochambeau plume source is significantly higher than the Iceland and Galapagos
25 plume sources, while the $^3\text{He}/^{36}\text{Ar}$ and $^3\text{He}/^{130}\text{Xe}$ ratios appear to be similar. The difference in
26 $^3\text{He}/^{22}\text{Ne}$ between Rochambeau and the Galapagos and Iceland plume sources could reflect long
27 lasting accretional heterogeneities in the deep mantle or some characteristic of the back-arc
28 mantle source.

29 High-precision xenon isotopic measurements indicate that the lower $^{129}\text{Xe}/^{130}\text{Xe}$ ratios in
30 the Rochambeau source cannot be explained solely by mixing atmospheric xenon with MORB-
31 type xenon; nor can fission-produced Xe be added to MORB Xe to produce the compositions
32 seen in the Rochambeau basalts. Deconvolution of fissiogenic xenon isotopes demonstrate a

33 higher proportion of Pu-derived fission Xe in the Rochambeau source compared to the MORB
34 source. Therefore, both I/Xe and Pu/Xe ratios are different between OIB and MORB mantle
35 sources. Our observations require heterogeneous volatile accretion and a lower degree of
36 processing for the mantle plume source compared to the MORB source. Since differences in
37 $^{129}\text{Xe}/^{130}\text{Xe}$ ratios have to be produced while ^{129}I is still alive, OIB and MORB sources were
38 degassed at different rates for the first 100 Ma of Solar System history, and subsequent to this
39 period, the two reservoirs have not been homogenized. In combination with recent results from
40 the Iceland plume, our observations require the preservation of less-degassed, early-formed
41 heterogeneities in the Earth's deep mantle throughout Earth's history.

42

43 **Introduction**

44 The noble gas compositions of mantle-derived basalts provide information on the
45 degassing history, style of mantle convection, and volatile exchange between the deep Earth and
46 exosphere. Compared to mid-ocean ridge basalts (MORBs), ocean island basalts (OIBs) from
47 Iceland, Hawaii, Galapagos, Réunion and Samoa are characterized by lower ratios of radiogenic
48 to primordial isotopes such as $^4\text{He}/^3\text{He}$, $^{21}\text{Ne}/^{22}\text{Ne}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ (e.g., Hanyu et al., 2001; Honda
49 et al., 1993; Mukhopadhyay 2012; Poreda and Farley, 1992; Raquin and Moreira, 2009; Trieloff
50 et al., 2000; Trieloff et al., 2002; Yokochi and Marty, 2004). Likewise, lower ratios of radiogenic
51 to non-radiogenic Xe isotopes ($^{129}\text{Xe}/^{130}\text{Xe}$) are found in Hawaii, Samoa, Iceland and Reunion
52 (e.g., Mukhopadhyay 2012; Poreda and Farley, 1992; Trieloff et al., 2000; Trieloff et al., 2002).
53 These noble gas signatures in OIBs are commonly attributed to sampling parts of Earth's mantle
54 that are significantly less degassed than the MORB source (e.g., Allegre et al., 1987; Graham,
55 2002; Gonnermann and Mukhopadhyay, 2009; Kurz et al., 1982; Kurz et al., 2009; Porcelli and

56 Wasserburg, 1995; Staudacher and Allegre, 1982). Shallow-level atmospheric contamination,
57 however, often makes it difficult to decipher whether the lower measured Ar and Xe isotopic
58 ratios in OIBs are indeed reflective of the mantle source composition. Additionally, the low
59 $^{40}\text{Ar}/^{36}\text{Ar}$ and $^{129}\text{Xe}/^{130}\text{Xe}$ ratios in OIBs may arise from recycled atmospheric Ar and Xe and not
60 from a less degassed reservoir (Holland and Ballentine, 2006; Kendrick et al., 2011; Trieloff and
61 Kunz, 2005).

62 If the low $^{129}\text{Xe}/^{130}\text{Xe}$ ratios in OIBs are indeed from a less degassed reservoir, then the
63 OIB and MORB reservoirs must be partially isolated from each other since 4.45 Ga as ^{129}I ,
64 which produces ^{129}Xe , became extinct 100 million years after the start of the Solar System. Such
65 long-term separation would invalidate many models put forth to explain the chemical and
66 dynamical evolution of the mantle. On the other hand, if the differences in $^{129}\text{Xe}/^{130}\text{Xe}$ ratios in
67 OIBs are from recycling of atmospheric Xe, long-term separation of the two sources is not
68 required and extensive mixing between the sources is allowed. Hence, addressing the origin of
69 the low $^{40}\text{Ar}/^{36}\text{Ar}$ and $^{129}\text{Xe}/^{130}\text{Xe}$ ratios observed in OIBs compared to MORBs is of
70 fundamental importance in understanding whether compositional heterogeneities dating back to
71 Earth's accretion are still preserved. The preservation of old heterogeneities in the deep mantle
72 can in turn provide important constraints on long-term mixing rates and mass flow in the mantle.

73 Recently, Mukhopadhyay (2012) and Tucker et al. (in press) demonstrated that the lower
74 $^{40}\text{Ar}/^{36}\text{Ar}$ and $^{129}\text{Xe}/^{130}\text{Xe}$ in the Iceland plume compared to depleted MORBs (Moreira et al.,
75 1998; Tucker et al., in press) cannot be generated solely through recycling of atmospheric noble
76 gases. To investigate whether the composition of the Iceland plume is representative of other
77 mantle plumes, we present combined He-Ne-Ar-Xe measurements in gas-rich basaltic glasses
78 from the Rochambeau Rift in the northern Lau back-arc basin with $^4\text{He}/^3\text{He}$ ratios as low as

79 25,600 (28.1 R_A , where R_A is the $^3\text{He}/^4\text{He}$ ratio normalized to the atmospheric ratio of 1.39×10^{-6}).
80 6).

81 The Rochambeau Rift is located in the northwestern flank of the Lau back-arc basin,
82 behind the Tonga arc, in the western Pacific (Fig. 1). Shear-wave splitting analyses suggest a fast
83 direction of anisotropy that is oriented north to south in the Lau back-arc spreading center (Smith
84 et al., 2001). If this anisotropy is interpreted in terms of mantle flow, it would suggest southward
85 flow of Pacific mantle (Smith et al., 2001). While slab rollback could induce the southward
86 mantle flow in the Lau back-arc, a consequence of such flow would be introduction of Samoan
87 plume material into the northern Lau back-arc region (Smith et al., 2001) through the tear in the
88 Tonga slab beneath the Vitiiaz lineament (Millen and Hamburger, 1998).

89 The flow of Samoan plume material into the northern Lau basin is consistent with
90 observations of low $^4\text{He}/^3\text{He}$ ratios along the Rochambeau Rift that slowly increase to MORB-
91 like values southwards (Poreda and Craig, 1992; Lupton et al., 2009; Turner and Hawkesworth,
92 1998). For example, $^4\text{He}/^3\text{He}$ ratios along the Rochambeau Rift are as low as 32,700- 25,600 (22-
93 $28.1 R_A$; Hahm et al., 2012; Poreda and Craig, 1992; Lupton et al., 2009). These values are
94 similar to the lowest $^4\text{He}/^3\text{He}$ ratio of 21,000 at the nearby Samoan plume ($34.2 R_A$, Jackson et
95 al., 2007; Farley et al., 1992). $^{40}\text{Ar}/^{36}\text{Ar}$ and $^{129}\text{Xe}/^{130}\text{Xe}$ ratios of up to $11,988 \pm 156$ and $7.04 \pm$
96 0.1 , respectively, have been measured in Samoan mantle xenoliths (Poreda and Farley, 1992).
97 Consequently, if Samoan plume material influences the He isotopic composition of basalts along
98 the Rochambeau Rift, non-atmospheric $^{40}\text{Ar}/^{36}\text{Ar}$ and $^{129}\text{Xe}/^{130}\text{Xe}$ ratios should be expected in
99 these basalts. Thus, basaltic glasses from the Rochambeau Rift could be ideal for characterizing
100 the heavy noble gas composition of a low $^4\text{He}/^3\text{He}$ mantle plume. In this study, we use our
101 combined He-Ne-Ar-Xe measurements from basaltic glass samples with low $^4\text{He}/^3\text{He}$ ratios from

102 the Rochambeau Rift to constrain the mantle source Ne, Ar and Xe isotopic composition. We use
103 the source composition from Rochambeau to investigate whether the lower $^{40}\text{Ar}/^{36}\text{Ar}$ and
104 $^{129}\text{Xe}/^{130}\text{Xe}$ ratios measured in plumes can be assigned to recycled atmospheric noble gases.
105 Additionally, we utilize our Xe isotopic measurements to constrain the age of heterogeneities
106 sampled by deep mantle plumes and test whether models of the dynamical and chemical
107 evolution of the mantle are consistent with our new observations.

108

109 **2. Analytical Methods**

110 We analyzed four basaltic glass samples from the Rochambeau Rift: NLD 13, NLD 14,
111 NLD 20 and NLD 27 (Fig. 1). $^4\text{He}/^3\text{He}$ ratios were previously measured by Lupton et al (2009)
112 and range between 25,600 and 46,700 ($15.4 R_A$ to $28.1 R_A$; Table 1). Reported He concentrations
113 range from 3 to $23 \times 10^{-6} \text{ cm}^3 \text{ STP g}^{-1}$. Glass chunks were carefully selected to avoid
114 phenocrysts. In order to remove surface alteration, glasses were leached in 2% nitric acid for 10
115 to 20 minutes, and then ultrasonically cleaned in distilled water and acetone. Single pieces of
116 basaltic glass (3.2 to 6.8 grams) were baked under vacuum for 24 hours at 100°C and were
117 pumped for an additional 6 to 12 days. Samples were crushed under vacuum using a hydraulic
118 ram to release magmatic gases trapped in vesicles. The released gases were purified by
119 sequential exposure to hot and cold SAES getters and a small split of the gas was let into a
120 quadrupole mass spectrometer to determine the Ar abundance and an approximate $^{40}\text{Ar}/^{36}\text{Ar}$
121 ratio. The noble gases were then trapped on a cryogenic cold-finger. He was separated from Ne
122 at 32 K and let into the Nu Noblesse mass spectrometer. The measurements were carried out at
123 $250 \mu\text{A}$ trap current and an electron accelerating voltage of 60 eV. The three Ne isotopes were
124 simultaneously detected on three discrete dynode multipliers operating in pulse counting mode.

125 Large ^{20}Ne beams ($>100,000$ cps) were measured on a Faraday cup. An automated liquid
126 nitrogen trap was used to keep the Ar and CO_2 backgrounds low and we corrected for isobaric
127 interferences from doubly-charged Ar and CO_2 . The $^{40}\text{Ar}^{++}/^{40}\text{Ar}^+$ and $\text{CO}_2^{++}/\text{CO}_2^+$ ratios were
128 0.031 ± 0.003 and 0.0045 ± 0.0005 , respectively, and the $^{40}\text{Ar}^{++}$ and CO_2^{++} corrections were all
129 below 1%. For Ar, depending on the abundance measured by the quadrupole mass spectrometer,
130 a fraction of the gas was let into the mass spectrometer. Isotopes were measured simultaneously
131 using the Faraday for ^{40}Ar and the axial and low mass multipliers for ^{38}Ar and ^{36}Ar , respectively.
132 Xe was measured using the three discrete dynode multipliers in a combination of multicollection
133 and peak jumping mode. Additional analytical details are described in Mukhopadhyay (2012).

134 Measured blanks of ^4He , ^{22}Ne , ^{36}Ar and ^{130}Xe were all below 1.5×10^{-11} , 1.06×10^{-13} , 8.1
135 $\times 10^{-13}$, and 1.3×10^{-16} cm^3 STP, respectively, but typically were a factor of 2 lower. Blanks had
136 isotopic ratios that were statistically undistinguishable from atmospheric values. Since the
137 bubbles trapped in the glass themselves have a post-eruptive air contaminant, no blank
138 corrections were applied to the sample isotopic ratios. Mass discrimination for He was corrected
139 using the HH3 standard with a $^4\text{He}/^3\text{He}$ ratio of 81,700 ($8.81 R_A$; Gayer et al., 2008) and Ne, Ar,
140 and Xe were corrected using air as a standard. Mass discrimination was monitored using sample-
141 standard bracketing with additional standards run overnight. The reproducibility of the standards
142 was used to determine the reported 1σ uncertainty.

143

144 3. Results

145 Multiple step crushes, between 13 and 45 steps, were performed for each sample. The
146 He-Ne-Ar-Xe abundance and isotopic data are summarized in Supplemental Tables 1 and 2.

147

148 *3.1. Measured Ne, Ar and Xe isotopic ratios*

149 We measured $^{20}\text{Ne}/^{22}\text{Ne}$ ratios of up to 12.22 ± 0.03 (1σ) and $^{21}\text{Ne}/^{22}\text{Ne}$ ratios up to
150 0.0430 ± 0.0002 (1σ) in the NLD 27 sample with a $^4\text{He}/^3\text{He}$ ratio of 46,700 (15.4 R_A). Measured
151 $^{40}\text{Ar}/^{36}\text{Ar}$ ratios vary up to 9269 ± 93 (1σ) in the same sample, which is close to measured
152 $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of up to $11,988 \pm 156$ at the PPT seamount off Samoa (Poreda and Farley, 1992).
153 The maximum measured $^{40}\text{Ar}/^{36}\text{Ar}$ value in NLD 13, which has a $^4\text{He}/^3\text{He}$ ratio of 25,600 (28.1
154 R_A) is 4828 ± 48 . We find measured $^{129}\text{Xe}/^{130}\text{Xe}$ excesses with respect to the atmospheric
155 composition in all 4 of the Rochambeau samples. The highest measured $^{129}\text{Xe}/^{130}\text{Xe}$ is $6.93 \pm$
156 0.03 (1σ) from NLD 13 and represents the largest excess yet recorded in a basalt with a $^4\text{He}/^3\text{He}$
157 ratio as low as 25,600 (Supplemental Table 2).

158

159 *3.2. Ne, Ar and Xe isotopic composition of the Rochambeau Rift mantle source*

160 Shallow-level air contamination affects all Ne, Ar, and Xe isotopic measurements in
161 mantle-derived basalts (e.g., Sarda et al., 1985; Honda et al., 1993; Farley and Neroda, 1998).
162 Accurately interpreting differences in noble gas compositions of mantle sources requires
163 correcting for the shallow level atmospheric contaminant. We correct for the atmospheric
164 contaminant through least-squares fitting of well-defined arrays in $^{21}\text{Ne}/^{22}\text{Ne}$ - $^{20}\text{Ne}/^{22}\text{Ne}$,
165 $^{20}\text{Ne}/^{22}\text{Ne}$ - $^{40}\text{Ar}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ - $^{129}\text{Xe}/^{130}\text{Xe}$ space. The fits are then extrapolated to the mantle
166 $^{20}\text{Ne}/^{22}\text{Ne}$ ratio. Correction for air contamination is a least-squares linear extrapolation for
167 $^{21}\text{Ne}/^{22}\text{Ne}$ (Figs. 2 and 3). For mantle $^{40}\text{Ar}/^{36}\text{Ar}$ and $^{129}\text{Xe}/^{130}\text{Xe}$ the corrections are least-squares
168 hyperbolic extrapolations (Figs. 4 and 5).

169 While recent work based on continental well gas and the gas-rich popping rock from the
170 north mid-Atlantic Ridge suggests that the MORB source has a $^{20}\text{Ne}/^{22}\text{Ne}$ of 12.5 (Ballentine et
171 al., 2005; Holland and Ballentine, 2006; Raquin et al., 2008), the Iceland (Mukhopadhyay, 2012)
172 and Kola plume (Yokochi and Marty, 2004) have a solar $^{20}\text{Ne}/^{22}\text{Ne}$ ratio. Therefore, the mantle
173 $^{20}\text{Ne}/^{22}\text{Ne}$ and $^{21}\text{Ne}/^{22}\text{Ne}$ of the Rochambeau Rift samples were determined by projecting the best
174 fit line through the step crushes to the OIB-MORB mixing line (Fig. 2). We denote the
175 extrapolated mantle source $^{21}\text{Ne}/^{22}\text{Ne}$ ratio as $^{21}\text{Ne}/^{22}\text{Ne}_E$. The mantle source $^{20}\text{Ne}/^{22}\text{Ne}$
176 determined from the intersection of the OIB-MORB mixing line with the best fit line through the
177 sample data is used to extrapolate the hyperbolic fits in Ar and Xe isotopic space to the
178 corresponding mantle source value (Figs. 4 and 5). We note that extrapolating Ar and Xe
179 isotopic ratios to a mantle $^{20}\text{Ne}/^{22}\text{Ne}$ of 12.5 does not affect our overall conclusions.

180 The x and y error weighted linear least square fits (Mukhopadhyay, 2012; Tucker et al., in
181 press) through the Ne data yield mantle $^{21}\text{Ne}/^{22}\text{Ne}_E$ values ranging between 0.042 and 0.048 (Fig.
182 2). Thus, all of the Rochambeau samples are less nucleogenic than the N. Mid-Atlantic Ridge
183 popping rock ($^{21}\text{Ne}/^{22}\text{Ne}_E = 0.06$; Moreira et al., 1998) and the depleted MORBs from the
184 Equatorial Atlantic (0.062-0.065; Tucker et al., in press).

185 To determine the mantle source Ar and Xe isotopic ratios corrected for shallow-level
186 atmospheric contamination, we only use sample NLD 27, for which a relatively large number of
187 steps yield a well-defined hyperbola (Figs. 4 and 5; Supplemental Tables 1 and 2). The
188 extrapolated mantle $^{40}\text{Ar}/^{36}\text{Ar}$ ratio ($^{40}\text{Ar}/^{36}\text{Ar}_E$) is $16,763 \pm 1,144$. The $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of the
189 Rochambeau source is significantly lower than the estimated source values of $27,000 \pm 4000$ for
190 popping rock (Raquin et al., 2008), $41,050 \pm 2670$ for the Bravo Dome well gas (Holland and
191 Ballentine, 2006) and $41,500 \pm 9000$ for the depleted equatorial Atlantic MORBs (Tucker et al.,

192 In press). The $^{40}\text{Ar}/^{36}\text{Ar}_E$ for the Rochambeau sample is higher than the Iceland plume source
193 $^{40}\text{Ar}/^{36}\text{Ar}_E$ of $10,732 \pm 3080$ (Mukhopadhyay, 2012). While we do not have sufficient number of
194 step crushes for NLD 13 and NLD 14 to independently constrain the mantle source value for
195 these two samples, the step crush data do in general fall on the hyperbolic best fit line for NLD
196 27 (Fig 4). Hence, all of the samples may have similar mantle source $^{40}\text{Ar}/^{36}\text{Ar}$ values.

197 The hyperbolic fit for NLD 27 in Ar-Xe space yields a mantle source value of $6.92 \pm$
198 0.07 , similar to the maximum measured values at the Rochambeau Rift and in the Samoan
199 xenoliths (Fig. 5). The mantle source value for NLD 27 is significantly lower than source values
200 of 7.6 for popping rock (Moreira et al., 1998), 7.9 ± 0.14 for Bravo Dome (Holland and
201 Ballentine, 2006) and 7.77 ± 0.06 for depleted equatorial Atlantic MORBs (Tucker et al., In
202 press). The composition of NLD 27, however, overlaps with the Iceland composition of $6.98 \pm$
203 0.07 (Mukhopadhyay, 2012). NLD 13 appears to have higher $^{129}\text{Xe}/^{130}\text{Xe}$ ratios for a given
204 $^{40}\text{Ar}/^{36}\text{Ar}$ ratio and thus, may have a higher mantle source value than NLD 27 although
205 additional data will be required to verify this claim. In any case, our observations confirm that
206 the measured $^{129}\text{Xe}/^{130}\text{Xe}$ ratios are not a result of shallow-level (post-eruptive) air contamination
207 but are a characteristic of the plume source.

208

209 **4. Relationships between elemental ratios and isotopic ratios**

210 *4.1. Helium-Neon in the Rochambeau Rift source*

211 The $^4\text{He}/^3\text{He}$ and $^{21}\text{Ne}/^{22}\text{Ne}_E$ isotopic compositions of the four Rochambeau samples
212 show the influence of a mantle plume, and the He-Ne isotopic ratios can be explained by mixing
213 between a less degassed mantle source (e.g., FOZO) and a depleted MORB source (Figs. 2 and

214 3). As seen in Figure 3, for a given $^4\text{He}/^3\text{He}$ ratio, the Rochambeau samples have a higher
215 $^{21}\text{Ne}/^{22}\text{Ne}_E$ compared to Iceland and Galapagos (Dixon et al., 2000; Moreira et al., 2001;
216 Mukhopadhyay, 2012; Tieloff et al., 2000; Raquin and Moreira 2009; Kurz et al., 2009), but
217 overlap with the range of compositions seen at Hawaii and Samoa (Honda et al., 1993; Valbracht
218 et al., 1996; Jackson et al., 2009; Poreda and Farley, 1992). The higher $^{21}\text{Ne}/^{22}\text{Ne}$ at a given
219 $^4\text{He}/^3\text{He}$ ratio reflects a higher time-integrated $^3\text{He}/^{22}\text{Ne}$ in the mantle source of the Rochambeau
220 basalts compared to plume sources at Iceland and Galapagos. A high $^3\text{He}/^{22}\text{Ne}$ in the Lau back-
221 arc source has been previously noted (Hahm et al., 2012; Honda et al., 1993; Lupton et al.,
222 2012), and there are at least two possible explanations for the high $^3\text{He}/^{22}\text{Ne}$ in the Rochambeau
223 Rift samples compared to Iceland and Galapagos:

224 i) The plume material that flows into the Rochambeau Rift has a higher $^3\text{He}/^{22}\text{Ne}$ ratio than
225 the Iceland and Galapagos plumes and mixes with the depleted back-arc mantle. Such an
226 explanation, however, requires that $^3\text{He}/^{22}\text{Ne}$ differences exist between plumes. Since mantle
227 $^3\text{He}/^{22}\text{Ne}$ ratios are difficult to perturb, such variations may point to preservation of
228 heterogeneities from the first few hundred million years of Earth's history within the deep mantle
229 (Honda et al., 1993; Kurz et al. 2009; Mukhopadhyay 2012; Yokochi and Marty 2005).

230 ii) The apparently high $^3\text{He}/^{22}\text{Ne}$ ratio results from mixing between a plume source with low
231 $^3\text{He}/^{22}\text{Ne}$ (~ 3) and a back-arc mantle with $^3\text{He}/^{22}\text{Ne}$ that is elevated with respect to the MORB
232 source. We note that high $^3\text{He}/^{22}\text{Ne}$ ratios have also been observed in the Manus back-arc basin
233 (Shaw et al., 2001). The reason why these back-arc basins may have high $^3\text{He}/^{22}\text{Ne}$, however, is
234 uncertain.

235

236 *4.2. Two mantle sources inferred from Helium-Argon relationships*

237 The most gas rich sample in our study, NLD 27, has a $^4\text{He}/^{40}\text{Ar}^*$ ratio of 3.3 (where ‘*’
238 indicates radiogenic), which is within the range of 1.6-4.2 expected for the mantle production
239 ratio. Hence, NLD 27 preserves relatively unfractionated mantle noble gas elemental ratios and
240 we focus on this sample for the rest of the manuscript. The step crushes from NLD 27
241 demonstrate excellent correlation between $^3\text{He}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ ratios (Fig. 6a), where ^3He and
242 ^{36}Ar are primordial. The $^3\text{He}/^{36}\text{Ar}$ ratio of 1.33 for the NLD 27 source is higher than both the
243 popping rock and Bravo Dome well gas sources, which have $^3\text{He}/^{36}\text{Ar}$ ratios of 0.4 and 0.3,
244 respectively (Holland and Ballentine, 2006; Moreira et al., 1998). The $^3\text{He}/^{36}\text{Ar}$ ratio of the
245 mantle can decrease over time because of preferential recycling of atmospheric Ar (Fig 6a).
246 Thus, the higher $^3\text{He}/^{36}\text{Ar}$ in Iceland and Rochambeau plumes relative to MORBs cannot be
247 related to recycling of atmospheric noble gases into a MORB-like mantle source. Likewise, since
248 mixing in the $^3\text{He}/^{36}\text{Ar}$ - $^{40}\text{Ar}/^{36}\text{Ar}$ space is linear, adding recycled atmospheric Ar to MORBs
249 does not explain the low $^{40}\text{Ar}/^{36}\text{Ar}$ ratios of the plume sources (Fig. 6a). Consequently, a less
250 degassed source is required to explain the lower $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of the plume source.

251 The Iceland, Galapagos, and Rochambeau samples define very similar slopes in
252 $^3\text{He}/^{36}\text{Ar}$ - $^{40}\text{Ar}/^{36}\text{Ar}$ space even though the Rochambeau source has a higher $^3\text{He}/^{22}\text{Ne}$. While the
253 Ne-Ar measurements from Galapagos do not yet constrain the mantle $^{40}\text{Ar}/^{36}\text{Ar}$ ratio in the
254 Galapagos plume (Raquin and Moreira, 2009), the Iceland source has lower $^{40}\text{Ar}/^{36}\text{Ar}$ and
255 $^3\text{He}/^{36}\text{Ar}$ ratios than the Rochambeau source. If the measured values at Galapagos are reflective
256 of a low $^{40}\text{Ar}/^{36}\text{Ar}$ ratio in the mantle source, then both the Galapagos and Iceland source
257 compositions could be related to the Rochambeau plume source through a greater proportion of
258 recycled Ar. Hence, the He-Ar results suggest both recycling of atmospheric Ar and the
259 existence of a less degassed reservoir in the plume source.

260

261 *4.3. Ancient MORB-OIB separation inferred from Helium-Xenon relationships*

262 Similar to the $^3\text{He}/^{36}\text{Ar}$ and $^{40}\text{Ar}/^{36}\text{Ar}$ correlation, the $^{129}\text{Xe}/^{130}\text{Xe}$ ratios from the
263 individual step crushes on NLD 27 show an excellent correlation with the $^3\text{He}/^{130}\text{Xe}$ ratios (Fig.
264 6b). We note that ^3He and ^{130}Xe are primordial, while ^{129}Xe is produced from decay of extinct
265 ^{129}I . The $^{129}\text{Xe}/^{130}\text{Xe}$ ratio, therefore, stopped evolving 100 Myr after the start of the Solar
266 System.

267 The data in Figure 6b demonstrate that, compared to the MORB source, the Rochambeau
268 and Iceland mantle sources evolved with different I/Xe ratios. The step crushes from the NLD 27
269 sample displays a slope that is quite distinct from the gas-rich MORB 2PID43 (popping rock),
270 but is similar to the correlation defined by Iceland. Since mixing in $^3\text{He}/^{130}\text{Xe}$ - $^{129}\text{Xe}/^{130}\text{Xe}$ space
271 is linear, adding subducted atmospheric Xe to the MORB-source will move the source
272 composition towards air along a straight line (Fig. 6b). Hence, adding subducted atmospheric Xe
273 to the MORB source clearly cannot produce the Rochambeau and Iceland mantle source
274 compositions (Fig. 6b). Similarly, the Rochambeau and Iceland source cannot supply Xe to the
275 MORB source unless radiogenic ^{129}Xe is added to the plume Xe isotopic composition. However,
276 ^{129}I became extinct at ~ 4.45 Ga. As a result, the difference in MORB and plume $^{129}\text{Xe}/^{130}\text{Xe}$
277 ratios must have been set up early and the last major equilibration between the two reservoirs
278 must have predated 4.45 Ga as otherwise the differences in $^{129}\text{Xe}/^{130}\text{Xe}$ would not have been
279 preserved in the present-day mantle. We conclude that plumes *cannot* supply Xe and all of the
280 primordial volatiles to the MORB source.

281

282 **5. Preservation of long-term heterogeneities in the mantle inferred from xenon isotopes**

283 The Xe isotopic compositions of mantle-derived rocks provide information about early
284 degassing and mantle differentiation. In addition to ^{129}Xe that was produced from decay of
285 extinct ^{129}I , ^{136}Xe was produced by spontaneous fission of now extinct ^{244}Pu (half-life = 80 Myr).
286 However, ^{136}Xe is also generated by spontaneous fission of extant ^{238}U . Thus, the I-Xe and Pu-
287 Xe systems are sensitive to the first ~100 Ma and 500 Ma of Earth history, respectively, while
288 the U-Xe system evolves throughout Earth history.

289 The error-weighted mean xenon isotope composition ($^{129}\text{Xe}/^{136}\text{Xe}$ vs. $^{130}\text{Xe}/^{136}\text{Xe}$) of
290 NLD 27, of all four of the Rochambeau samples, Iceland (Mukhopadhyay, 2012), MORBs from
291 the Southwest Indian Ridge (Parai et al., In revision) and MORBs from the Equatorial Atlantic
292 (Tucker et al., In press) are shown in Figure 7. Our observations demonstrate that MORBs and
293 plumes have small but distinct differences in $^{129}\text{Xe}/^{136}\text{Xe}$ ratios. Because all of the plotted
294 samples were analyzed using the same procedure in the same laboratory, the differences between
295 these groups of basalts cannot be related to inter-laboratory artifacts. We note that the data
296 plotted in Figure 7 have not been corrected for post-eruptive air contamination, so the mantle
297 source compositions will lie further from the atmospheric composition along the straight line
298 joining the measured and atmospheric compositions. However, correcting for shallow-level air
299 contamination is not required to demonstrate that the Rochambeau (and Iceland) source cannot
300 be related to the MORB source by addition of atmospheric xenon. Thus, while recycling of
301 atmospheric Xe may occur to the deep Earth (Holland and Ballentine, 2006; Kendrick et al.,
302 2011; Mukhopadhyay, 2012; Tucker et al., In press), we emphasize that recycling by itself
303 cannot explain the $^{129}\text{Xe}/^{136}\text{Xe}$ isotopic difference between MORBs and plumes. Likewise,

304 mixing MORB Xe with fissiogenic ^{136}Xe in recycled slabs will lead to a decrease in the
305 $^{129}\text{Xe}/^{136}\text{Xe}$ ratio. Hence, plume Xe cannot be a mixture of MORB and fissiogenic Xe.

306 The $^{129}\text{Xe}/^{136}\text{Xe}$ ratio is a measure of the time integrated $^{129}\text{I}/(^{244}\text{Pu}+^{238}\text{U})$ ratio and the
307 differences in the Xe isotopic composition between the different basalt groups (Fig. 7) can be
308 explained by mantle processing and mixing of less degassed and more degassed mantle sources.
309 A mantle reservoir that undergoes degassing after I and Pu are extinct will have low
310 concentrations of primordial ^{130}Xe , radiogenic ^{129}Xe and fissiogenic ^{136}Xe produced by extinct
311 ^{244}Pu . Addition of ^{136}Xe from ^{238}U fission to such a degassed source would decrease both the
312 $^{129}\text{Xe}/^{136}\text{Xe}$ and the $^{130}\text{Xe}/^{136}\text{Xe}$ ratios of the reservoir (Fig. 7b). Hence, we conclude that the
313 MORB sources are more degassed than the plume sources, a conclusion that is based only on the
314 Xe isotopic ratios and independent of the absolute concentration of noble gases or the
315 partitioning of noble gases with respect to their radiogenic parents.

316

317 *5.1. Pu-U-I systematics in the Rochambeau Rift source*

318 The ^{244}Pu - and ^{238}U -produced fission isotopes of Xe ($^{131,132,134,136}\text{Xe}$) provide information
319 about mantle processing rates, particularly during the Hadean (e.g., Allegre et al., 1987; Coltice
320 et al., 2009; Kunz et al., 1998; Ozima et al., 1985; Yokochi and Marty, 2005). ^{244}Pu and ^{238}U
321 produce the four fission isotopes in different proportions and fission Xe yields from Pu are
322 significantly larger than from U. A reservoir that has remained completely closed to volatile loss
323 over Earth's history will have $^{136}\text{Xe}_{\text{Pu}}*/^{136}\text{Xe}_{\text{U}}*$ of ~ 27 , where '*' refers to fissiogenic Xe
324 (Tolstikhin et al., 2006; Tolstikhin and O'Nions, 1996). ^{244}Pu became extinct at ~ 4 Ga and
325 reservoirs that underwent extensive degassing over the past 4 Ga would have lost a significant
326 fraction of the Pu-produced fission Xe and, thus, have a large proportion of ^{238}U -derived fission

327 Xe; i.e., $^{136}\text{Xe}_{\text{Pu}^*}/^{136}\text{Xe}_{\text{U}^*}$ in degassed reservoirs will be $\ll 27$. Consequently, deconvolving
328 ^{244}Pu - from ^{238}U -produced fission Xe using the measured isotopic ratios provides a direct
329 constraint on the degree of outgassing of a mantle reservoir.

330 To deconvolve ^{238}U - from ^{244}Pu -derived fissiogenic Xe, five Xe isotopes were used
331 ($^{130,131,132,134,136}\text{Xe}$). A sufficient number of step-crushes are available for NLD 27 and
332 deconvolution of Pu- from U-derived Xe was performed for only this sample. In mantle-derived
333 basalts, different vesicles have different proportions of mantle Xe and post-eruptive atmospheric
334 Xe contamination. To determine the mantle source composition for the fission isotopes, the
335 $^{129}\text{Xe}/^{132}\text{Xe}$ ratios in the individual steps were regressed against the $^{40}\text{Ar}/^{36}\text{Ar}$ ratio using a least
336 squares hyperbolic fit, which yielded a mantle $^{129}\text{Xe}/^{132}\text{Xe}$ ratio of 1.038 at a $^{40}\text{Ar}/^{36}\text{Ar}$ of 16,763
337 (Supplemental Figure 1). Next, the $^{130,131,134,136}\text{Xe}/^{132}\text{Xe}$ ratios in the individual crushing steps
338 were regressed against the $^{129}\text{Xe}/^{132}\text{Xe}$ ratio. From the slopes and uncertainties in the slopes, the
339 mantle $^{130,131,134,136}\text{Xe}/^{132}\text{Xe}$ ratios, along with their uncertainties, were calculated for a mantle
340 $^{129}\text{Xe}/^{132}\text{Xe}$ ratio of 1.038 for the Rochambeau source (Supplemental Table 3). To investigate
341 whether inclusion of some of the less precise measurements affect the fission deconvolution, the
342 above analyses were redone using a filtered data set; only data points with $^{132}\text{Xe}/^{136}\text{Xe}$ distinct
343 from the atmospheric composition at the 2σ level and with a relative error of $<1\%$ were selected.
344 Such filtering only eliminates 4 data points and does not affect the deconvolution.

345 Following determination of the mantle source composition, the least-squares solution to
346 the system $Ax = b$ was found with the following additional constraints: $\sum x_i = 1$ and $0 \leq x_i \leq 1$
347 (also see Caffee et al., 1999; Mukhopadhyay, 2012). Here, A defines the composition of the end-
348 members, x the fraction of each component, and b the sample composition. End-member and
349 mantle source compositions (A and b , respectively) were normalized to the standard deviations in

350 the mantle source isotope ratios to assign equal weight to each isotope ratio. To compute the
351 uncertainties, a Monte Carlo technique was used whereby the estimated sample composition was
352 varied at random within the 1σ uncertainty and the least squares fit recomputed using the new
353 values. For all simulations, it was verified that convergence to a minimum was achieved.

354 For the initial Xe isotopic composition of the mantle, we investigated chondritic (AVCC)
355 and solar Xe. We selected AVCC and solar Xe based on i) recent observations of AVCC Kr in
356 the mantle (Holland et al., 2009), ii) $^{128}\text{Xe}/^{130}\text{Xe}$ excess with respect to air in continental well
357 gases (Caffee et al., 1999; Holland et al., 2009) and iii) lower extent of Xe mass fractionation in
358 the Archean atmosphere compared to the present day atmosphere (Pujol et al., 2011). The initial
359 mantle compositions along with the isotopic compositions of Pu- and U-produced fission Xe are
360 listed in Supplemental Table 3.

361 Depending on whether the initial mantle Xe is solar or chondritic, the fraction of ^{136}Xe
362 derived from ^{244}Pu fission is 0.87 ± 0.11 or 0.85 ± 0.14 , respectively (Table 1). The Pu-derived
363 ^{136}Xe fractions are similar to those from the Iceland plume (Mukhopadhyay, 2012; Table 1), but
364 higher than values of 0.30-0.60 inferred for the Kola plume (Yokochi and Marty, 2012). The
365 values for the Kola plume were inferred based on elemental correlations between ^4He - ^{21}Ne -
366 ^{136}Xe , and the lower values at Kola may arise in part because elemental ratios can be fractionated
367 through a combination of solubility and diffusivity controlled degassing (Gonnermann and
368 Mukhopadhyay, 2007; Paonita and Martelli, 2007; Yokochi and Mary, 2005). The Iceland and
369 Rochambeau plume values are higher than values of 0.25 to 0.29 for the depleted MORB source
370 (Tucker et al., in press; Table 1). Since the Iceland, Rochambeau and depleted MORB data were
371 obtained in the same laboratory using the same techniques, the higher proportion of ^{244}Pu -
372 derived ^{136}Xe in plumes is a robust result. A higher fraction ^{244}Pu -derived ^{136}Xe is a clear

373 indicator of a less degassed source, and hence, we conclude that the Rochambeau and Iceland
374 plume sources must sample a less degassed mantle than the MORB source.

375 The combined I-Pu-Xe system can constrain the closure time for volatile loss of a mantle
376 reservoir through the $^{129}\text{Xe}^*/^{136}\text{Xe}_{\text{Pu}}^*$ ratio, where $^{129}\text{Xe}^*$ is the decay product of ^{129}I decay and
377 $^{136}\text{Xe}_{\text{Pu}}^*$ is ^{136}Xe produced from ^{244}Pu fission (Allegre et al., 1987; Azbel and Tolstikhin, 1993;
378 Coltice et al., 2009; Kunz et al., 1998; Ozima et al., 1985; Pepin and Porcelli, 2006; Staudacher
379 and Allegre, 1982; Yokochi and Marty, 2005). Since ^{129}I has a shorter half-life than ^{244}Pu , higher
380 $^{129}\text{Xe}^*/^{136}\text{Xe}_{\text{Pu}}^*$ ratios are indicative of earlier closure to volatile loss. We find that for the
381 Rochambeau source, the $^{129}\text{Xe}^*/^{136}\text{Xe}_{\text{Pu}}^*$ ratio is $2.9^{+0.6}_{-0.4}$ to $3.2^{+1.2}_{-0.4}$ (Table 2). While detailed
382 modeling of the fission and radiogenic Xe isotopes is beyond the scope of this paper, we note
383 that the $^{129}\text{Xe}^*/^{136}\text{Xe}_{\text{Pu}}^*$ values at Rochambeau are comparable to those from Iceland but
384 significantly lower than the MORB source (Table 1). Interpreting these values as closure ages for
385 a mantle with an initially homogenous I/Pu ratio, the higher $^{129}\text{Xe}^*/^{136}\text{Xe}_{\text{Pu}}^*$ ratio in the depleted
386 MORB source would imply that the shallow upper mantle became closed to volatile loss prior to
387 the deep mantle reservoir supplying noble gases to the mantle plumes. Such a conclusion appears
388 paradoxical. Rather, a simpler explanation is that the lower $^{129}\text{Xe}^*/^{136}\text{Xe}_{\text{Pu}}^*$ in the Rochambeau
389 and Iceland source reflects a lower initial I/Pu ratio for the plume source compared to the MORB
390 source. This difference would suggest that the initial phase of Earth's accretion was volatile poor
391 compared to the later stages of accretion because Pu is a refractory element while I is a volatile
392 element (e.g., Mukhopadhyay, 2012). Since this difference in I/Pu ratio is still preserved in the
393 present day Xe isotopic ratio of the mantle, we argue that the whole mantle was never
394 completely homogenized.

395

396 **6. Implications for the age of mantle heterogeneities and mantle evolution**

397 Our fundamental observation from Iceland and Rochambeau is that plumes have lower
398 $^{129}\text{Xe}/^{130}\text{Xe}$ ratio than MORBs and that the difference cannot be related solely through recycling
399 of atmospheric noble gases. This observation requires that the reservoirs supplying noble gases
400 to plumes and MORBs were processed and outgassed to different extents within the first 100
401 million years of Earth's history and that subsequently these reservoirs have not been
402 homogenized. Models that seek to explain the geochemical evolution of the mantle must satisfy
403 this fundamental constraint from the $^{129}\text{Xe}/^{130}\text{Xe}$ ratio.

404 We now discuss the constraints Xe isotopes place on mantle reservoirs and in particular
405 evaluate whether our observations are consistent with two classes of models put forth to explain
406 the geochemical evolution of the mantle: i) the steady-state mantle models (e.g., Porcelli and
407 Wasserburg, 1995; Tolstikhin and O'Nions, 1996; Tolstikhin et al., 2006) and ii) models that
408 generate reservoirs over Earth's history with primitive He isotopic signatures (e.g., Davies, 2010;
409 Lee et al., 2010).

410

411 *6.1. Steady-state mantle models*

412 The differences in noble gas compositions between MORBs and OIBs are often
413 interpreted in terms of steady-state mantle models that require primordial ^3He , ^{22}Ne , ^{36}Ar and
414 ^{130}Xe and radiogenic ^{129}Xe in the volatile-depleted MORB source to be derived from a more
415 primitive volatile-rich plume source (Kellogg and Wasserburg 1990; Porcelli and Wasserburg,
416 1995; Tolstikhin and O'Nions, 1996). Mixtures of the plume-derived noble gases, radiogenic
417 noble gases produced in the MORB source, and subducted atmospheric Ar and Xe into the

418 MORB source leads to the more radiogenic noble gas isotopic compositions observed in
419 MORBs. While originally the plume source was assumed to be the whole lower mantle, the basic
420 framework could still be viable if instead of the whole lower mantle, the plume source was much
421 smaller, such as D'' (e.g., Tolstikhin et al., 2006).

422 If the primordial gases in the MORB source are derived from the plume source, elemental
423 abundance ratios are expected to be the same in the two sources. However, the Iceland plume has
424 different elemental abundances than MORBs (Mukhopadhyay, 2012). Our Rochambeau data
425 also show differences from MORBs. For example, the $^3\text{He}/^{36}\text{Ar}$ is 1.3 in Rochambeau Rift
426 source vs. 0.3 in the Bravo Dome well gas source (Holland and Ballentine, 2006). More
427 importantly, in the steady-state models, $^{129}\text{Xe}/^{130}\text{Xe}$ ratio in the plume source is higher than in
428 the MORB source, a prediction that is clearly refuted by our observations of lower $^{129}\text{Xe}/^{130}\text{Xe}$ in
429 the plume source. As noted earlier, the lower $^{129}\text{Xe}/^{130}\text{Xe}$ ratio in the plume source cannot arise
430 solely from recycling. Thus, we suggest that all of the primordial gases and the radiogenic ^{129}Xe
431 in the MORB source cannot be derived from the plume source. Therefore, the two reservoir
432 steady-state mantle models are not consistent with the observations and need to be re-evaluated.

433

434 *6.2. Generation of a 'primordial-looking' reservoir*

435 In contrast to many models that assign the low $^4\text{He}/^3\text{He}$ ratios observed in many OIBs to
436 a primordial reservoir, Lee et al. (2010) suggested that a 'primordial-looking' reservoir could
437 have been produced during the first billion years of Earth's history through a process termed
438 upside-down differentiation. Lee et al. (2010) suggest that a hotter mantle during the Hadean and
439 earliest Archean leads to partial melting at depths between 660 and 410 km, producing Fe-rich
440 melts. At these depths, melts are denser than the surrounding mantle and sink to the core-mantle

441 boundary, possibly forming the two large low shear wave velocity provinces at the base of the
442 mantle. Since the melts never degas, they are volatile-rich, and because partial melting transfers
443 the incompatible elements to the melts without fractionation, the melts have primordial time
444 integrated $U/{}^3\text{He}$, ${}^{87}\text{Sr}/{}^{86}\text{Sr}$, ${}^{143}\text{Nd}/{}^{144}\text{Nd}$, ${}^{206}\text{Pb}/{}^{204}\text{Pb}$ today, i.e., a ‘primordial-looking’ reservoir
445 (Lee et al., 2010). As the noble gases are incompatible (e.g., Heber et al., 2007), the model
446 predicts that the primordial noble gas elemental ratios (e.g., ${}^3\text{He}/{}^{22}\text{Ne}$, ${}^3\text{He}/{}^{36}\text{Ar}$) of the melts
447 would be the same as the solid convecting mantle. Importantly, differences in ${}^{129}\text{Xe}/{}^{130}\text{Xe}$
448 between the MORB source and the low ${}^4\text{He}/{}^3\text{He}$ reservoir are not expected, since the process of
449 generating the ‘primordial’ reservoir occurs over for 1 Gyr, well past the 100 Ma lifetime of ${}^{129}\text{I}$.

450 MORBs and low ${}^4\text{He}/{}^3\text{He}$ plumes, however, have different elemental ratios (Fig. 6; also
451 see Mukhopadhyay, 2012). Furthermore, the ${}^{20}\text{Ne}/{}^{22}\text{Ne}$ ratio of the plume and MORB sources is
452 different (Mukhopadhyay, 2012; Yokochi and Marty, 2004). Since Ne is not subducted back to
453 the mantle in significant quantities, the difference in ${}^{20}\text{Ne}/{}^{22}\text{Ne}$ is related to Earth’s accretion
454 (Ballentine et al., 2005; Mukhopadhyay, 2012) and cannot result from melting processes.
455 Furthermore, the ${}^{129}\text{Xe}/{}^{130}\text{Xe}$ data contradict the hypothesis that the primitive looking noble gas
456 reservoir could be generated by melt segregation to the CMB over timescales of 1 Ga. If the
457 noble gases are from a reservoir that was produced after 4.45 Ga, plumes and MORBs would
458 have the same ${}^{129}\text{Xe}/{}^{130}\text{Xe}$ ratio, or have ${}^{129}\text{Xe}/{}^{130}\text{Xe}$ ratios that can be related to each other
459 through addition of subducted air. Thus, we rule out the upside-down differentiation as the main
460 mechanism for producing a reservoir with primitive noble gas signatures. We stress that we do
461 not argue against the generation of Fe-rich melts during the Hadean and early Archean (Lee et
462 al., 2010), but argue that such a process by itself cannot generate the noble gas signature seen in
463 mantle plumes.

464 Davies (2010) suggested a somewhat similar hypothesis to Lee et al. (2010) to explain
465 the primitive noble gas signatures of OIBs with two important distinctions: the process of
466 generating the primitive-looking noble gas reservoir occurs throughout Earth's history and the
467 process occurs under mid-ocean ridges in the shallow upper mantle when undegassed melts react
468 with the peridotites to produce pyroxenites. The pyroxenites are heavier than the peridotites and
469 are assumed to sink to the D'' region and are sequestered there for long periods of time. Because
470 the geochemical consequences for the noble gases are the same as the upside-down
471 differentiation model (Lee et al. 2010), the same arguments presented above allow us to rule out
472 Davies' (2010) hypothesis as the primary mechanism for generating the primitive-looking OIB
473 reservoir.

474 Several studies have suggested that the primitive-looking $^4\text{He}/^3\text{He}$ ratios in OIBs are
475 signatures of depleted residues of mantle melting because U is more incompatible than He (e.g.,
476 Coltice and Ricard, 1999; Parman et al., 2005). In such scenarios, separation of the MORB and
477 low $^4\text{He}/^3\text{He}$ reservoirs is not required over Earth's history. Rather, because the residues have
478 very low U/ ^3He ratios, the $^4\text{He}/^3\text{He}$ ratio of the convecting mantle gets frozen in the residues. For
479 residues generated at 2-3 Ga, the convecting mantle $^4\text{He}/^3\text{He}$ could have the same values as
480 observed in many OIBs. Our results from Iceland and Rochambeau suggest that if low $^4\text{He}/^3\text{He}$
481 ratios in OIBs are indeed due to sampling of depleted residues of mantle melting, then the
482 $^{129}\text{Xe}/^{130}\text{Xe}$ ratios require the depleted residues to be generated prior to 4.45 Ga. In other words,
483 the low $^4\text{He}/^3\text{He}$ reservoir has essentially behaved as a closed system over Earth's history.

484

485 *6.3 The nature of the large low shear wave velocity provinces (LLSVPs)*

486 Several recent studies have suggested that plumes might originate from the LLSVPs at
487 the base of the mantle (e.g., Burke, 2011; Dziewonski et al., 2010; Torsvik et al., 2010). Both
488 primitive (Deschamps et al., 2011; Jackson and Carlson, 2011; Mukhopadhyay 2012) and
489 recycled material (Hutko et al., 2006; Tackley, 2011; Tan and Gurnis, 2005) have been invoked
490 for LLSVPs. If plumes are indeed drawing material from LLSVPs, then based on the Iceland and
491 Rochambeau Xe data we can conclusively say that these features must have been produced prior
492 to 4.45 Ga (Figs. 6 and 7). Therefore, LLSVPs are long lasting structures in the deep mantle and
493 are essentially as old as the age of the Earth.

494 Our observation that the Rochambeau and Iceland plume sources have high proportions
495 of Pu-derived fission Xe as well as recycled atmospheric Xe requires that plumes sample both
496 primitive and recycled material. We note that the DICE 10 sample from Iceland has amongst the
497 most primitive $^{21}\text{Ne}/^{22}\text{Ne}$ ratio, yet ~90% of its Xe is from a recycled source (Table 1). Hence, if
498 all of the plume material is derived from LLSVPs then these features must also be composed of
499 both recycled and primitive lithologies. Alternatively, deep mantle flow could channel subducted
500 slabs towards the margins of the LLSVPs, where they get entrained by the rising plumes. In this
501 regard, we urge caution in using the measured lithophile isotopic compositions in low $^4\text{He}/^3\text{He}$
502 ratio plume basalts as a direct measure of the composition of primitive mantle (Jackson et al.,
503 2010; Jackson and Carlson, 2011).

504

505 **7. Conclusions**

506 We measured He, Ne, Ar, and Xe abundances and isotopic compositions of four plume
507 influenced basalts with low $^4\text{He}/^3\text{He}$ ratios from the Rochambeau Rift in the northern Lau back-
508 arc basin. We documented that sample NLD 13 with a $^4\text{He}/^3\text{He}$ ratio of 25,600 (28.1 R_A) has a

509 $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of at least 4828 and $^{129}\text{Xe}/^{130}\text{Xe}$ ratio of at least 6.93 ± 0.03 . For NLD 27, which
510 had a sufficient number of step crushes, we infer mantle source $^{40}\text{Ar}/^{36}\text{Ar}$ ratio of $16,763 \pm 1,144$
511 and $^{129}\text{Xe}/^{130}\text{Xe}$ ratio of 6.92 ± 0.07 . These values are consistent with the mantle plume at the
512 Rochambeau Rift to be from Samoa (also see Poreda and Farley, 1992).

513 The new results from the Lau basin confirm the Xe isotopic findings from Iceland that
514 the plume reservoir has a low $^{129}\text{Xe}/^{130}\text{Xe}$ that cannot result solely from adding subducted
515 atmospheric Xe to MORB Xe. Rather, the plume source has a lower I/Pu ratio compared to the
516 MORB source. Given the short half-life of ^{129}I , the result suggests that the plume source was
517 more volatile-poor compared to the MORB source and the two reservoirs were separated from
518 each other within the first 100 million years of Earth's history. Subsequent to this period, the two
519 reservoirs could not have been homogenized as otherwise the difference in $^{129}\text{Xe}/^{130}\text{Xe}$ would
520 not be preserved in the present-day mantle. Models that seek to explain the dynamical and
521 chemical evolution of the mantle must be compatible with these results. For example, if plumes
522 are indeed derived from LLSVPs, then the Xe data require LLSVPs to have existed since 4.45
523 Ga.

524

525 **Acknowledgements:** This work was supported by NSF grant EAR 0911363.

526

527 **References**

- 528 Allegre C., Hofmann A., ONions K., 1996. The Argon constraints on mantle structure. *Geophys.*
529 *Res. Lett.* 23, 3555–3557.
- 530 Allegre C., Staudacher T., Sarda P., 1987. Rare Gas Systematics: Formation of the Atmosphere,
531 Evolution and Structure of the Earth's Mantle. *Earth Planet. Sci. Lett.* 81, 127–150.
- 532 Azbel, I. Y., Tolstikhin, I., 1993. Accretion and early degassing of the Earth: Constraints from

533 Pu-U-Xe isotopic systematics. *Meteoritics* 28, 609–621.

534 Ballentine, C. J., Holland, G., 2008. What CO₂ well gases tell us about the origin of noble gases
535 in the mantle and their relationship to the atmosphere. *Philosophical Transactions of the*
536 *Royal Society A: Mathematical, Physical and Engineering Sciences* 366, 4183–4203.

537 Ballentine C. J., et al., 2005. Neon isotopes constrain convection and volatile origin in the Earth's
538 mantle. *Nature* 433, 33-38.

539 Caffee M. W., et al., 1999. Primordial Noble Gases from Earth's Mantle: Identification of a
540 Primitive Volatile Component. *Science* 285, 2115–2118.

541 Coltice N., Ricard Y., 1999. Geochemical observations and one layer mantle convection. *Earth*
542 *Planet. Sci. Lett.* 174, 125-137.

543 Coltice N., Marty B., Yokochi R., 2009. Xenon isotope constraints on the thermal evolution of
544 the early Earth. *Chem. Geol.*, 266, 4-9.

545 Davies, G. F., 2010. Noble gases in the dynamic mantle. *Geochemistry Geophysics Geosystems*
546 11, Q03005, DOI: 3010.01029/02009GC002801.

547 Deschamps F., Kaminski E., Tackley P. J., 2011. A deep mantle origin for the primitive
548 signature of ocean island basalt, *Nature Geoscience* 4, 879-882.

549 Dixon E. T., Honda M., McDougall I.H., 2000. Preservation of near-solar neon isotopic ratios in
550 Icelandic basalts. *Earth Planet. Sci. Lett.* 180, 309–324.

551 Farley K. A. and Neroda E., 1998. Noble gases in the Earth's mantle. *Ann. Rev. Earth Planet.*
552 *Sci.* 26, 189-218.

553 Farley K. A., Natland J. H., Craig H., 1992. Binary mixing of enriched and undegassed
554 (primitive?) mantle components (He, Sr, Nd, Pb) in Samoan lavas. *Earth Planet. Sci. Lett.*
555 111, 183-199.

556 Gayer E., Mukhopadhyay S., Meade B. J., 2008. Spatial variability of erosion rates inferred from
557 the frequency distribution of cosmogenic ³He in olivines from Hawaiian river sediments.
558 *Earth Planet. Sci. Lett.* 266, 303-315.

559 Gonnermann H. M., Mukhopadhyay S., 2009. Preserving noble gases in a convecting mantle.
560 *Nature* 459, 560–563.

561 Gonnermann H. M., Mukhopadhyay, S., 2007. Non-equilibrium degassing and a primordial
562 source for helium in ocean–island volcanism. *Nature* 449, 1037–1040.

563 Graham D. W., et al., 2001. Upper mantle dynamics revealed by helium isotope variations along

564 the Southeast Indian Ridge. *Nature* 409, 701-703.

565 Graham D. W., 2002. Noble gas isotope geochemistry of mid-ocean ridge and ocean island
566 basalts: characterization of mantle source reservoirs. In Porcelli D., Ballentine C. J.,
567 Wieler R. (Eds). *Noble gases in geochemistry and cosmochemistry: Reviews in*
568 *Mineralogy and Geochemistry*, vol 47, 247-318.

569 Hahm D., et al., 2012. An overview of the volatile systematics of the Lau basin- Resolving the
570 effects of source variation, magmatic degassing and crustal contamination. *Geochim.*
571 *Cosmochim. Acta* 85, 88-113.

572 Hanyu T., et al., 2001. Noble gas study of the Reunion hotspot: evidence for distinct less-
573 degassed mantle sources. *Earth Planet. Sci. Lett*, 193, 83-98.

574 Heber V. S., et al., 2007. Crystal-melt partitioning of noble gases (helium, neon, argon, krypton,
575 and xenon) for olivine and clinopyroxene. *Geochim. Cosmochim. Acta* 71, 1041-1061.

576 Hilton D. L., et al., 1993. Helium and argon isotope systematics from the central Lau Basin and
577 Valu Fa Ridge: evidence of crust/mantle interactions in a back-arc basin. *Geochim.*
578 *Cosmochim. Acta* 57, 2819-2841.

579 Holland G., Ballentine C. J., 2006. Seawater subduction controls the heavy noble gas
580 composition of the mantle. *Nature* 441, 186–191.

581 Holland G., Cassidy M., Ballentine C. J., 2009. Meteorite Kr in Earth's Mantle Suggests a Late
582 Accretionary Source for the Atmosphere. *Science* 326, 1522–1525.

583 Honda M., et al., 1993. Noble gases in submarine pillow basalt glasses from the Lau Basin:
584 detection of a solar component in backarc basin basalts. *Earth Planet. Sci. Lett.* 120, 135–
585 148.

586 Honda M., et al., 1993. Noble gases in submarine pillow basalts from Loihi and Kilauea, Hawaii:
587 A solar component in the Earth. *Geochim. Cosmochim. Acta* 57, 858-874.

588 Hutko A., et al., 2006, Seismic detection of folded, subducted lithosphere at the core-mantle
589 boundary. *Nature* 441, 333–336.

590 Jackson M. G., Kurz M. D., Hart S. R., Helium and neon isotopes in phenocrysts from Samoan
591 lavas: Evidence for heterogeneity in the terrestrial high $^3\text{He}/^4\text{He}$ mantle. *Earth Planet. Sci.*
592 *Lett.* 287, 519-528.

593 Jackson M. G., et al., 2007. New Samoan lavas from Ofu Island reveal a hemispherical
594 heterogeneous high $^3\text{He}/^4\text{He}$ mantle. *Earth Planet. Sci. Lett.* 264, 360-374.

595 Kellogg L. H., Wasserburg G. J., 1990. The role of plumes in helium fluxes. *Earth Planet. Sci.*
596 *Lett.* 99, 276-289.

597 Kendrick M.A., et al., 2011. High abundances of noble gas and chlorine delivered to the mantle
598 by serpentinite subduction. *Nature Geoscience* 4, 807–812.

599 Kunz J., Staudacher T., Allègre C.J., 1998. Plutonium-fission xenon found in Earth's mantle.
600 *Science* 280, 877–880.

601 Kurz M. D., Jenkins W., Hart S., 1982. Helium Isotopic Systematics of Oceanic Islands and
602 Mantle Heterogeneity. *Nature* 297, 43–47.

603 Kurz M. D., et al., 2009. Primitive neon from the center of the Galapagos hotspot. *Earth Planet.*
604 *Sci. Lett.* 286, 23-34.

605 Lee C. T. A., et al., 2010. Upside-down differentiation and generation of a 'primordial' lower
606 mantle. *Nature* 463, 930-933.

607 Leya I., Wieler R., 1999. Nucleogenic production of Ne isotopes in Earth's crust and upper
608 mantle induced by alpha particles from the decay of U and Th. *J. Geophys. Res.* 104,
609 15439-15450.

610 Lupton J. E., et al., 2012. Mantle hotspot neon in basalts from the Northwest Lau back-arc basin.
611 *Geophys. Res. Lett.* 39, L08308, doi:10.1029/2012GL051201.

612 Lupton J. E. et al., 2009. Helium isotope variations in seafloor basalts from the northwest Lau
613 back-arc basin: Mapping the influence of the Samoan plume. *Geophys. Res. Lett.* 36,
614 L1731, doi:10.1029/2009GL039468.

615 Mahoney J. J., et al., 1993. Isotope and trace element characteristics of a super-fast spreading
616 ridge: East Pacific Rise, 13-238S. *Earth Planet. Sci. Lett.* 121, 173- 193.

617 Marty B., 1989. Neon and Xenon Isotopes in MORB: implications for the earth-atmosphere
618 evolution. *Earth Planet. Sci. Lett.* 94, 45–56.

619 Millen D. W., Hamburger M. W., 1998. Seismological evidence for tearing of the Pacific plate at
620 the northern termination of the Tonga subduction zone. *Geology* 26, 659-662.

621 Moreira M., Kunz J., Allegre C., 1998. Rare gas systematics in popping rock: Isotopic and
622 elemental compositions in the upper mantle. *Science* 279, 1178–1181.

623 Moreira M., et al., 2001. Solar neon in the Icelandic mantle: new evidence for an undegassed
624 lower mantle. *Earth Planet. Sci. Lett.* 185, 15–23.

625 Mukhopadhyay, S., 2012. Early differentiation and volatile accretion recorded in deep-mantle

626 neon and xenon. *Nature* 486, 101–104.

627 Ozima M., Podosek F., Igarashi G., 1985. Terrestrial xenon isotope constraints on the early
628 history of the Earth. *Nature* 315, 471–474.

629 Parman S.W., 2007. Helium isotopic evidence for episodic mantle melting and crustal growth.
630 *Nature* 446, 900-903.

631 Paonita A., Martelli M., 2007. A new view of the He-Ar-CO₂ degassing at mid-ocean ridges:
632 Homogeneous composition of magmas from the upper mantle. *Geochim. Cosmochim*
633 *Acta* 71, 1747-1763.

634 Pepin R.O., 1991. On the origin and early evolution of terrestrial planet atmospheres and
635 meteoritic volatiles. *Icarus* 92, 2–79.

636 Pepin R. O., Porcelli D., 2006. Xenon isotope systematics, giant impacts, and mantle degassing
637 on the early Earth. *Earth Planet. Sci. Lett.* 250, 470–485.

638 Porcelli D., Wasserburg G., 1995. Mass transfer of helium, neon, argon, and xenon through a
639 steady-state upper mantle. *Geochim. Cosmochim. Acta* 59, 4921–4937.

640 Poreda R. J., Farley K. A., 1992. Rare gases in Samoan xenoliths. *Earth Planet. Sci. Lett.* 113,
641 129-144.

642 Poreda R. J., Craig H., 1991. He and Sr isotopes in the Lau basin mantle – Depleted and
643 primitive mantle components. *Earth Planet. Sci. Lett.* 113, 487-493.

644 Pujol M., Marty B., Burgess R., 2011. Chondritic-like xenon trapped in Archean rocks: A
645 possible signature of the ancient atmosphere. *Earth Planet. Sci. Lett.* 308, 298–306.

646 Raquin A., Moreira M., 2009. Atmospheric ³⁸Ar/³⁶Ar in the mantle: Implications for the nature
647 of the terrestrial parent bodies. *Earth Planet. Sci. Lett.* 287, 551–558.

648 Raquin A., Moreira M., Guillon F., 2008. He, Ne and Ar systematics in single vesicles: Mantle
649 isotopic ratios and origin of the air component in basaltic glasses. *Earth Planet. Sci. Lett.*
650 274, 142–150.

651 Regelous M., et al., 2008. Mantle dynamics and mantle melting beneath Niufo’ou Island and
652 the northern Lau back-arc basin, *Contrib. Mineral. Petrol.* 156, 103–118.

653 Sarda P., Staudacher T., Allegre C. J., 1985. ⁴⁰Ar/³⁶Ar in MORB glasses - constraints on
654 atmosphere and mantle evolution. *Earth Planet. Sci. Lett.* 72, 357-375.

655 Shaw A. M., et al., 2001. Nucleogenic neon in high ³He/⁴He lavas from the Manus back-arc
656 basin: a new perspective on He-Ne decoupling. *Earth Planet. Sci. Lett* 194, 53-66.

657 Smith G. P., et al., 2001. A complex pattern of mantle flow in the Lau backarc. *Science* 292,
658 713-716.

659 Staudacher T., Allegre C. J., 1982. Terrestrial xenology. *Earth Planet. Sci. Lett.* 60, 389-406.

660 Stuart F. M., et al., 2003. High $^3\text{He}/^4\text{He}$ ratios in picritic basalts from Baffin Island and the role
661 of a mixed reservoir in mantle plumes. *Nature* 424, 57-59.

662 Sumino H., et al., 2010. Seawater-derived noble gases and halogens preserved in exhumed
663 mantle wedge peridotite. *Earth Planet. Sci. Lett.* 294, 163–172.

664 Tackley P. J., 2011. Living dead slabs in 3-D: The dynamics of compositionally-stratified slabs
665 entering a “slab graveyard” above the core-mantle boundary. *Phys. Earth Planet. Interiors*
666 188, 150-162.

667 Tan E., Gurnis M., 2005. Metastable superplumes and mantle compressibility. *Geophys. Res.*
668 *Lett.* L20307, doi: 10.1029/2005GL024190.

669 Tolstikhin I., Hofmann A.W., 2005. Early crust on top of the Earth's core. *Phys. Earth Planet.*
670 *Interiors* 148, 109–130.

671 Tolstikhin I., O'Nions R., 1996. Some comments on isotopic structure of terrestrial xenon. *Chem.*
672 *Geol.* 129, 185–199.

673 Tolstikhin I., Kramers J. D., Hofmann A. W., 2006. A chemical Earth model with whole mantle
674 convection: The importance of a core-mantle boundary layer (D'') and its early formation.
675 *Chem. Geol.*, 226, 79-99.

676 Trieloff M., Kunz J., 2005. Isotope systematics of noble gases in the Earth's mantle: possible
677 sources of primordial isotopes and implications for mantle structure. *Phys. Earth Planet.*
678 *Interiors* 148, 13–38.

679 Trieloff M., Kunz J., Allègre C. J., 2002. Noble gas systematics of the Reunion mantle plume
680 source and the origin of primordial noble gases in Earth's mantle. *Earth Planet. Sci. Lett.*
681 200, 297–313.

682 Trieloff M., et al., 2000. The nature of pristine noble gases in mantle plumes. *Science* 288, 1036.

683 Tucker J. M., Mukhopadhyay S., Schilling J-G., In press. The heavy noble gas composition of
684 the depleted MORB mantle (DMM) and its implications for the preservation of
685 heterogeneities in the mantle. *Earth Planet. Sci. Lett.*, doi:10.1016/j.epsl.2012.08.025.

686 Turner S., Hawkesworth C., 1998. Using geochemistry to map mantle flow beneath the Lau
687 basin. *Geology* 26, 1019-1022.

- 688 Valbracht P.J., et al., 1997. Noble gas systematics of deep rift zone glasses from Loihi Seamount,
689 Hawaii. *Earth Planet. Sci. Lett.* 150, 399–411.
- 690 Yatssevich I., Honda M., 1997. Production of nucleogenic neon in the Earth from natural
691 radioactive decay. *J. Geophys. Res.* **102**, 10291-10298.
- 692 Yokochi R., Marty B., 2004. A determination of the neon isotopic composition of the deep
693 mantle. *Earth Planet. Sci. Lett.* 225, 77–88.
- 694 Yokochi, R., Marty, B., 2005. Geochemical constraints on mantle dynamics in the Hadean. *Earth*
695 *Planet. Sci. Lett.* 238, 17–30.

696 **Figure Captions**

697 **Figure 1:** Map showing the location of the four NLD samples along the Rochambeau Rift in the
698 northern Lau back-arc basin.

699 **Figure 2:** Neon three isotope plot for samples from the Rochambeau Rift. Each point represents
700 the Ne isotopic composition of a step crush. Error bars are 1σ . The mantle $^{20}\text{Ne}/^{22}\text{Ne}$ is set at
701 accretion while $^{21}\text{Ne}/^{22}\text{Ne}$ evolves as a function of the degree of degassing of a mantle reservoir
702 with low ratios indicative of a less degassed reservoir. Because mantle-derived basalts have
703 vesicles with variable degrees of air contamination, step crushing produces a linear array that lies
704 between air and the mantle composition. Projecting the best fit line through the step crushes to
705 the mantle $^{20}\text{Ne}/^{22}\text{Ne}$ ratio value yields the mantle $^{21}\text{Ne}/^{22}\text{Ne}$. While the MORB source has a
706 $^{20}\text{Ne}/^{22}\text{Ne}$ of 12.5 (Ballentine et al., 2005; Ballentine and Holland, 2008; Raquin et al., 2008), the
707 Iceland and Kola plumes have higher $^{20}\text{Ne}/^{22}\text{Ne}$, close to the solar composition (Mukhopadhyay,
708 2012; Yokochi and Marty, 2004). We projected the best fit line through the step crushes to the
709 OIB-MORB mixing line, which subsequently defines the mantle source $^{21}\text{Ne}/^{22}\text{Ne}$ ($^{21}\text{Ne}/^{22}\text{Ne}_E$)
710 of the basalts. The OIB endmember is based on the least radiogenic $^{21}\text{Ne}/^{22}\text{Ne}$ measured at
711 Galapagos (Kurz et al., 2009) and the MORB composition is from $^{21}\text{Ne}/^{22}\text{Ne}$ in depleted MORBs
712 from the equatorial Atlantic (Tucker et al., In press). Best fit lines were calculated using x and y
713 error weighted fits forced through the atmospheric composition. L2012 is the Ne isotopic data
714 for NLD 13 and NLD 27 from Lupton et al. (2012) and these data are used in calculating the
715 error-weighted best fit lines.

716 **Figure 3.** $^4\text{He}/^3\text{He}$ ratios in the Rochambeau Rift samples plotted against the mantle source
717 $^{21}\text{Ne}/^{22}\text{Ne}$ ratio ($^{21}\text{Ne}/^{22}\text{Ne}_E$). All of the Rochambeau samples have lower $^{21}\text{Ne}/^{22}\text{Ne}_E$ compared

718 to the North Atlantic popping rock (2ΠD43; Moreira et al., 1998) and to depleted MORBs from
719 the Equatorial Atlantic (Tucker et al., In press). The Rochambeau samples appear to show a
720 similar trend to the five Samoan xenoliths from Savai and PPT seamount (Poreda and Farley,
721 1992), but have a higher $^{21}\text{Ne}/^{22}\text{Ne}_E$ compared to basalts from the Samoan islands of Ofu and
722 Tau (Jackson et al., 2009). The OIB endmember is based on lowest measured $^4\text{He}/^3\text{He}$ ratio at
723 Baffin Island (Stuart et al., 2003) and the least nucleogenic $^{21}\text{Ne}/^{22}\text{Ne}$ from Galapagos (Kurz et
724 al., 2009). The depleted mantle composition was selected based on the extrapolation of the trend
725 observed in depleted MORBs from the equatorial Atlantic (Tucker et al., In press) to a $^4\text{He}/^3\text{He}$
726 ratio of 73,000, which corresponds to the He isotopic composition in the most depleted MORBs
727 from the Garret fracture zone (see Mahoney et al., 1993, discussion in Graham et al. 2001). $R =$
728 $(^3\text{He}/^{22}\text{Ne})_{\text{MORB}}/(^3\text{He}/^{22}\text{Ne})_{\text{plume}}$. For reference, the fields for Galapagos (Kurz et al., 2009),
729 Iceland (Moreira et al., 2001; Mukhoapdhyay 2012; Trieloff et al., 2000), Loihi (Honda et al.,
730 1993; Valbracht et al., 1997), and Manus basin (Shaw et al., 2001) are shown.

731 **Figure 4. A)** Ne-Ar compositions of individual step crushes for the NLD 27 sample from the
732 Rochambeau Rift. ^{40}Ar is generated by radioactive decay of ^{40}K and low $^{40}\text{Ar}/^{36}\text{Ar}$ ratios are
733 indicative of a less degassed mantle. Popping rock from the North Mid-Atlantic Ridge is shown
734 for comparison (Moreira et al., 1998) and the Bravo Dome well gas data is from Holland and
735 Ballentine (2006). The vesicle compositions in basaltic glass are a combination of magmatic
736 gases and shallow-level post-eruptive air contamination. Step crushing leads to sampling of
737 vesicles with varying degrees of air contamination, which in Ne-Ar space should lead to a
738 hyperbolic trend. A least-squares hyperbolic fit through the data indicate that the mantle source
739 for NLD 27 (Rochambeau source) has a $^{40}\text{Ar}/^{36}\text{Ar}$ of $16,763 \pm 1,144$ for a mantle $^{20}\text{Ne}/^{22}\text{Ne}$ of
740 13.22 (see text for discussion). **B)** Step crushes from samples NLD 13 and NLD 14. The

741 hyperbolic best fit regression for NLD 27 is overlain on the data. The $^{40}\text{Ar}/^{36}\text{Ar}$ ratios for the
742 NLD 13 and NLD 14 mantle sources appear to be comparable to that of NLD 27.

743 **Figure 5. A)** Hyperbolic mixing between $^{40}\text{Ar}/^{36}\text{Ar}$ and $^{129}\text{Xe}/^{130}\text{Xe}$ for the NLD 27 sample from
744 the Rochambeau Rift. Like for Ne-Ar, step crushing leads to sampling of vesicles with varying
745 degrees of air contamination, which will generate a hyperbolic trend between the atmospheric
746 composition and the mantle composition. The correlation shows scatter, likely reflecting the
747 presence of a second fractionated shallow-level air contaminant. A least squares hyperbolic best-
748 fit curve through the data when projected to a mantle $^{20}\text{Ne}/^{22}\text{Ne}$ of 13.22 yields a mantle source
749 $^{129}\text{Xe}/^{130}\text{Xe}$ value of 6.92 ± 0.07 , significantly lower than measured values in MORBs but similar
750 to the Iceland source of 6.98 ± 0.07 . Note that given the curvature in Ar-Xe space, the defined
751 $^{129}\text{Xe}/^{130}\text{Xe}$ in the Rochambeau mantle source is not particularly sensitive to the exact choice of
752 the mantle $^{40}\text{Ar}/^{36}\text{Ar}$ ratio. **B)** Step crushes showing the Ar-Xe relation for NLD 13 and NLD 14.
753 The hyperbolic best fit regression for NLD 27 is overlain on the data.

754 **Figure 6.** Elemental abundance ratios plotted against radiogenic isotope ratios for NLD 27
755 (Rochambeau Rift), DICE 10 (Iceland), Galapagos plume and popping rock (MORB). **A)**
756 $^3\text{He}/^{36}\text{Ar}$ vs. $^{40}\text{Ar}/^{36}\text{Ar}$ and **B)** $^3\text{He}/^{130}\text{Xe}$ vs. $^{129}\text{Xe}/^{130}\text{Xe}$. Iceland data is from Mukhopadhyay
757 (2012), and the Galapagos data is from Raquin and Moreira (2009), and the popping rock data is
758 from Moreira et al. (1998). Good linear relationships are observed between isotope ratios and
759 elemental ratios, which reflect mixing between mantle-derived noble gases and post-eruptive
760 atmospheric contamination. Note that both the Rochambeau and Iceland plumes define the same
761 trend but are quite distinct from popping rock (MORB source). The mixing lines denote the
762 trajectory along which the mantle source will evolve towards the air composition as subducted
763 air is mixed into the mantle source. Therefore, the low $^{40}\text{Ar}/^{36}\text{Ar}$ and low $^{129}\text{Xe}/^{130}\text{Xe}$ ratios in

764 plumes cannot be generated by adding subducted air. Hence, (at least) two distinct mantle
765 reservoirs with different $^{129}\text{Xe}/^{130}\text{Xe}$ ratios are required. Since ^{129}Xe is produced from ^{129}I decay,
766 the $^{129}\text{Xe}/^{130}\text{Xe}$ ratio stopped evolving after ^{129}I became extinct 100 Myr after the start of the
767 solar system. As a result, the difference in MORB and plume $^{129}\text{Xe}/^{130}\text{Xe}$ ratio must have been
768 set up early and the timescale of last major equilibration between the two reservoirs must predate
769 4.45 Ga.

770 **Figure 7. A)** Differences in measured $^{129}\text{Xe}/^{132}\text{Xe}$ - $^{136}\text{Xe}/^{132}\text{Xe}$ between plumes (Iceland and
771 Rochambeau) and depleted MORBs (equatorial Atlantic; Tucker et al., In press). Step crushes in
772 MORBs define a slope of 0.3898 ± 0.0081 (MSWD=0.78) while the plume data define a slope of
773 0.2937 ± 0.0065 (MSWD=0.68). Thus, the depleted MORBs and the Rochambeau and Iceland
774 plumes sources have clear differences in the proportion of radiogenic to fissiogenic Xe; the
775 MORB and plume sources cannot be related to each other solely through recycling of
776 atmospheric Xe. **B)** Differences in measured $^{129}\text{Xe}/^{136}\text{Xe}$ between the two plumes (Iceland and
777 Rochambeau) and MORBs from the Southwest Indian Ridge (n=104; Parai et al., In revision)
778 and depleted MORBs from the equatorial Atlantic (n=25; Tucker et al., In press). RR ALL stands
779 for the error-weighted average derived from all the step crushes on the NLD 13, NLD 14 and
780 NLD 27 (n= 67; Supplemental Table 2). The measured values have not been corrected for post-
781 eruptive air contamination. However, both post-eruptive contamination and recycling of
782 atmospheric Xe will move the mantle source composition linearly towards the atmospheric
783 composition. Therefore, the small Xe isotopic difference between the Rochambeau-Iceland
784 plumes and MORBs cannot be related solely through recycling atmospheric Xe or by adding
785 fissiogenic ^{136}Xe to MORB Xe.