Abstract

We present the first neon data, as well as new helium data, on Terceira Island (Azores archipelago, Portugal). Clear $^{20}$Ne and $^{21}$Ne excesses compared to air are observed ($^{20}$Ne/$^{22}$Ne $>$ 11.2) and moreover, the samples show a more primitive $^{21}$Ne/$^{22}$Ne ratio than MORB, confirming that the Azores hotspot can be considered as sampling a "primitive", relatively undegassed, reservoir. Most $^4$He/$^3$He isotopic ratios range between 80,000 and 63,500 (~ 9 to 11.5 R/Ra), being similar to those previously reported by [1] [M. Moreira, R. Doucelance, B. Dupré, M. Kurz, C.J. Allègre, Helium and lead isotope geochemistry in the Azores archipelago, Earth Planet. Sci. Lett. 169 (1999) 189–205]. A more primitive $^4$He/$^3$He ratio of ~ 50,000 (R/Ra ~ 15) was obtained in one sample, but we cannot completely exclude the possibility of a cosmogenic $^3$He contribution for this sample. Our study illustrates that the neon systematics can be more capable than helium to constrain the ultimate origin of hotspots in geodynamic settings dominated by plume–ridge interaction.

Keywords: neon; helium; Azores; plume–ridge interaction

1. Introduction

From the collection of helium isotopic data in the last 20 yrs, significantly different $^4$He/$^3$He signatures emerged for the MORB and OIB sources (e.g. [2,3] and references therein). $^4$He production results from radioactive decay of U and Th and different time-integrated isotopic ratios can be produced by heterogeneous mantle degassing, through distinct depletion of He as compared with U and Th. MORB samples, considered to come from the upper mantle, present a narrow range of $^4$He/$^3$He ratios with a mean value around 85,000 to 90,000 (R/Ra = 8 to 8.5; where Ra is the atmospheric $^3$He/$^4$He ratio of $1.384 \times 10^{-6}$).
significantly larger variation is found for OIB, but $^4\text{He}/^3\text{He}$ ratios as low as 20,500 ($R/R_\alpha \sim 35$) and 15,000 ($R/R_\alpha \sim 50$) measured in samples from Loihi seamount and Iceland hotspot (Baffin island) respectively, are interpreted as reflecting the contribution of a more primitive and less degassed source believed to be the lower mantle [4–8]. This interpretation is in agreement with the geophysical evidence linking the hotspot activity in these areas to deeply rooted mantle plumes [9–11]. The existence of these $^4\text{He}/^3\text{He}$ contrasts led to the proposition of a two-layer mantle model where the upper mantle is degassed, while a relatively undegassed reservoir is sampled by deep mantle plumes generating OIB [8,12].

More recent studies on noble gases have shown that the two-layer mantle model is confirmed by neon isotopic data (e.g. [13–16]). $^{20}\text{Ne}$ and $^{22}\text{Ne}$ are almost primitive isotopes (the nucleogenic productions of $^{20}\text{Ne}$ and $^{22}\text{Ne}$ can be neglected in mantle rocks) and similar $^{20}\text{Ne}/^{22}\text{Ne}$ ratios were found for MORB and OIB samples [13–15,17–21]. Different $^{21}\text{Ne}/^{22}\text{Ne}$ ratios are, however, assigned to MORB and OIB sources. Since $^{21}\text{Ne}$ derives from $\alpha$ particle-induced nuclear reactions such as $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ [22], its production is connected with radiogenic He formation. The higher $^{21}\text{Ne}/^{22}\text{Ne}$ ratio observed in MORB as compared with OIB samples (for a given $^{20}\text{Ne}/^{22}\text{Ne}$ ratio), is compatible with a higher time-integrated (Th+U)/He ratio as previously predicted from the $^4\text{He}/^3\text{He}$ signatures for the more degassed upper mantle (e.g. [14,16]). Moreover, in oceanic islands where the mixing between primitive mantle and MORB sources or magmas is believed to occur, it has been shown that Ne can be more sensitive than He in detecting contributions from relatively undegassed sources [16,20,23–26].

Notwithstanding that Azores archipelago emerges in the vicinity of the Mid-Atlantic Ridge, it has been considered as a hotspot resulting from a mantle plume [27–29] being characterized by a buoyancy flux similar to that reported, for example, for the Canary Islands (e.g. [30]). Nevertheless, mantle plumes have been the locus of an intense debate and controversy still exists concerning their ultimate origin: D” layer, bottom of mantle transition zone and even the asthenosphere have been considered as possible solutions (e.g. [31]). Noble gas isotope geochemistry is a key tool to clear up this question given its ability to identify the incorporation of relatively undegassed lower mantle portions in plumes. The Azores was firstly classified as one of the “low $^3\text{He}$ hotspots” characterized by $^4\text{He}/^3\text{He}$ ratios higher than the mean MORB ratio [32]. However, Moreira et al. [1] showed that Azores Islands have $^3\text{He}/^4\text{He}$ ratios both higher and lower than MORB values. Higher values ($^4\text{He}/^3\text{He}>140,000$; $R/R_\alpha<5.2$) are represented by rocks from the Eastern part of S. Miguel, while more primitive ratios were recorded in Terceira samples (around 64,000; $R/R_\alpha=11.3$). Using Pb isotopic data, these authors considered the S. Miguel end-member ($^{206}\text{Pb}/^{204}\text{Pb}=20.00$; $^{207}\text{Pb}/^{204}\text{Pb}=15.75$; $^{208}\text{Pb}/^{204}\text{Pb}=40.33$) resulting from the presence in the mantle source of subcontinental lithosphere delaminated during the opening of the Atlantic. For the Terceira end-member, the radiogenic lead isotopic signatures ($^{208}\text{Pb}/^{204}\text{Pb}=20.02$; $^{207}\text{Pb}/^{204}\text{Pb}=15.64$) coupled with the unradiogenic $^4\text{He}/^3\text{He}$ signatures, were assigned to mixing between ancient recycled oceanic crust and high $^3\text{He}$ material from the lower mantle. The influence of such low $^4\text{He}/^3\text{He}$ plume signature on the Mid-Atlantic ridge basalts was demonstrated by [33], who reported a significant decrease of the MORB $^4\text{He}/^3\text{He}$ ratio in the vicinity of the Azores archipelago. However, given that the helium anomaly ($^4\text{He}/^3\text{He}=64,000$; $R/R_\alpha=11.3$) measured in [1] was not sufficient to clearly constrain the origin of the Azores hotspot, we decided to analyze both helium and neon isotopes in new olivine phenocrysts sampled in Terceira lavas. Concerning Ne, these are the first data published for Azores archipelago.

2. Geology of Terceira and sample locations

The Azores archipelago is located in the North Atlantic near the triple junction where the Eurasian, African and American plates meet (Fig. 1). The boundary between the Eurasian and African plates is currently considered as represented by the NW–SE lineament of S. Miguel–Terceira–Graciosa or, alternatively, by the direction defined by S. Miguel–S. Jorge Islands. Several models have been proposed to describe the plate kinematics along this boundary: pure extension [34–37], pure strike-slip [38] and combined extension and strike-slip (e.g. [39–43]). A
discussion concerning the validity of these models is far beyond the aim of our work, but an important extensional component is obvious taking into account the volcanism associated with that boundary. As stated above, the locus of this extension remains controversial, but some authors argued that it is well
expressed in Terceira Island, in the so-called Terceira rift (see Fig. 1; [34–37]).

Terceira Island is slightly elongated in the E–W direction with an emerged area of about 382 km². The relief increases towards west, where the highest elevation reaches 1021 m, and the topography is dominated by four main strato-volcanoes (Fig. 2; [44–46]): Cinco Picos, Guilherme Moniz, Pico Alto and Santa Bárbara. At least the later two are considered to be active and a fissure zone connecting Guilherme Moniz and Santa Bárbara calderas has been regarded as the surface expression of Terceira rift. Cinco Picos area comprises the oldest volcanic centre, currently dominated by a partially eroded caldera with a diameter of almost 7 km, the largest of the Azores archipelago. Smaller cinder cones located in this area materialize minor and more recent eruptions that partially fill this caldera; their deposits are particularly well exposed in the southeast coast of Terceira [45].

From the geological map (Fig. 2) it can be concluded that both pyroclasts and lava flows are common, ranging in composition from primitive basalts to comendites and pantellerites [46,47]. Despite the work of Self [46], where stratigraphic relations between several volcanic units younger than 23,000 yrs are described, absolute age data is still very scarce. Recent basaltic lavas are recognized to have mainly erupted from the fissure zone (the youngest eruption occurred in 1761) sometimes forming extensive flows towards both northern and southern coasts, but little is known about the early evolution of the island. Nevertheless, K/Ar measurements from Feraud et al. [48] have shown that Cinco Picos caldera was formed less than 300,000 yrs ago. Sample locations are given in Fig. 2 and a brief petrographic description of the samples can be found in Appendix A. Most samples resulted from eruption events described by Self [46] (younger than 23,000 yrs). For samples PMT-24 and PMT-71A to 74A the ages are more uncertain.

3. Analytical procedure and results

3.1. Analytical procedure

Noble gases were mainly measured from green to yellow olivine phenocrysts, representing 18 samples of basaltic to mugearitic composition (see Table 1 and Appendix A). For one sample (PMT-24), a fraction of clinopyroxene was also analysed. Olivine was preferentially chosen given its significantly lower U and Th contents as compared with clinopyroxene (e.g. [49]) preventing the in-situ production of radiogenic ³He and nucleogenic ²¹Ne. Moreover, clinopyroxenes are more likely to retain crustal He (radiogenic) incorporated into magma en route to the surface [50], given the higher diffusivity of He in pyroxenes than in olivines at magmatic temperatures [51]. Different lava flows were sampled during the first field campaign in 2001. Some olivine poor specimens were re-sampled in 2002 and referenced with the same code (appended with the suffix A). In order to prevent a hypothetical influence of cosmogenic helium and neon on the determined isotopic ratios, most samples were collected from road-cuts or young sea-cliff faces (see Appendix A). Fresh whole rock samples were gently crushed and sieved (sieves with apertures of 1, 1.4, 2 and 4 mm) regarding the different dimensions of olivine crystals. Grains were then handpicked with the aid of a table light and a binocular microscope. All crystals with iddingsitic alteration rims, visible intragranular fractures and/or attached basaltic matrix were excluded for analysis. Moreover, homogeneous olivine separates concerning their petrographic characteristics were, whenever possible, always prepared. Mineral separates were ultrasonically cleaned in distilled water and submitted to HF reaction for 15 to 30 min to remove the external surface that could be in contact with the basaltic matrix. The final step of cleaning was achieved with ethanol and acetone. Most phenocryst separates were prepared with a minimum weight of ~ 0.7 g, in order to obtain a good accuracy in the analytical results.

Volatiles were extracted by crushing in vacuum (150 to 220 strokes) because this method mainly releases magmatic gases trapped in phenocryst melt inclusions (e.g. [52]). Helium and neon were analyzed in Paris during 2003 with the mass spectrometer ARESIBO I [53], equipped with a new fully automated line with low He and Ne blanks. The new analytical procedure will be given elsewhere [54]. Mean ⁴He and ²²Ne blanks were 3.6 ± 0.7 × 10⁻¹⁰ cm³STP and 1.5 ± 0.3 × 10⁻¹³ cm³STP respectively (uncertainties are 1σ), with isotopic ratios similar to the air ratios. ²²Ne blank proportions were between 2% and 75%
During the period of analyses, the signal of CO\textsuperscript{2+} and 40Ar\textsuperscript{+} were ~ 1000 cps and 150 cps respectively. This implied ~ 8 cps of CO\textsuperscript{2+} and ~ 15 cps of 40Ar\textsuperscript{2+} that corresponds generally to only few percent of the neon signal (usually 20Ne>1000 cps in samples).

Table 1
Helium and neon isotopes in Terceira samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>Diameter (mm)</th>
<th>Weight (g)</th>
<th>4He (ncm\textsuperscript{3}STP/g)</th>
<th>R/Ra ±</th>
<th>4He\textsuperscript{3}/He ±</th>
<th>20Ne\textsuperscript{22}/Ne ±</th>
<th>21Ne\textsuperscript{22}/Ne ±</th>
<th>22Ne (pcm\textsuperscript{3}STP/g)</th>
<th>21Ne/22Ne\textsubscript{corr}</th>
</tr>
</thead>
<tbody>
<tr>
<td>PMT8</td>
<td>1–1.4</td>
<td>0.88</td>
<td>31.0</td>
<td>9.8 0.2</td>
<td>73729</td>
<td>1505 9.92</td>
<td>0.04 0.0292</td>
<td>0.0004 3.8</td>
<td></td>
</tr>
<tr>
<td>PMT9</td>
<td>1–1.4</td>
<td>0.80</td>
<td>46.0</td>
<td>9.8 0.1</td>
<td>73729</td>
<td>752 9.87</td>
<td>0.05 0.0299</td>
<td>0.0003 2.9</td>
<td></td>
</tr>
<tr>
<td>PMT24-1</td>
<td>1.4–2</td>
<td>0.89</td>
<td>7.2</td>
<td>11.4 0.7</td>
<td>63381</td>
<td>3892 9.89</td>
<td>0.09 0.0276</td>
<td>0.0007 0.6</td>
<td></td>
</tr>
<tr>
<td>PMT24-2</td>
<td>2–4</td>
<td>0.67</td>
<td>37.0</td>
<td>10.5 0.1</td>
<td>68814</td>
<td>655 9.97</td>
<td>0.06 0.0301</td>
<td>0.0004 3.0</td>
<td></td>
</tr>
<tr>
<td>PMT24-3</td>
<td>1–3</td>
<td>0.45</td>
<td>13.0</td>
<td>10.2 0.7</td>
<td>70768</td>
<td>4713 9.89</td>
<td>0.05 0.0293</td>
<td>0.0006 1.5</td>
<td></td>
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<tr>
<td>PMT24-4</td>
<td>1–1.4</td>
<td>0.79</td>
<td>10.0</td>
<td>9.0 0.7</td>
<td>80283</td>
<td>6244 9.84</td>
<td>0.04 0.0300</td>
<td>0.0007 1.3</td>
<td></td>
</tr>
<tr>
<td>PMT24 Pyr</td>
<td>1.4–2</td>
<td>0.83</td>
<td>13.0</td>
<td>9.1 0.5</td>
<td>79400</td>
<td>4363 9.85</td>
<td>0.04 0.0297</td>
<td>0.0003 5.7</td>
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</tr>
<tr>
<td>PMT79</td>
<td>1–1.4</td>
<td>0.99</td>
<td>35.0</td>
<td>9.8 0.1</td>
<td>73729</td>
<td>752</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PMT19</td>
<td>1–1.4</td>
<td>1.00</td>
<td>32.0</td>
<td>9.7 0.1</td>
<td>74489</td>
<td>768 10.11</td>
<td>0.08 0.0307</td>
<td>0.0005 0.9</td>
<td></td>
</tr>
<tr>
<td>PMT4</td>
<td>1–1.4</td>
<td>0.89</td>
<td>26.0</td>
<td>10.0 0.1</td>
<td>72254</td>
<td>723 10.68</td>
<td>0.18 0.0321</td>
<td>0.0017 0.2</td>
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</tr>
<tr>
<td>PMT61</td>
<td>1–1.4</td>
<td>1.81</td>
<td>39.0</td>
<td>9.3 0.1</td>
<td>76686</td>
<td>818 10.19</td>
<td>0.07 0.0327</td>
<td>0.0004 0.7</td>
<td></td>
</tr>
<tr>
<td>PMT89</td>
<td>1–1.4</td>
<td>0.98</td>
<td>63.0</td>
<td>9.4 0.1</td>
<td>72984</td>
<td>737 11.28</td>
<td>0.23 0.0373</td>
<td>0.0020 0.2</td>
<td></td>
</tr>
<tr>
<td>PMT27</td>
<td>0.5–1</td>
<td>1.60</td>
<td>40.0</td>
<td>9.3 0.3</td>
<td>77693</td>
<td>2506 9.84</td>
<td>0.02 0.0291</td>
<td>0.0002 7.7</td>
<td></td>
</tr>
<tr>
<td>PMT71A</td>
<td>1–1.4</td>
<td>0.32</td>
<td>16.6</td>
<td>10.0 0.9</td>
<td>68164</td>
<td>6816 9.93</td>
<td>0.07 0.0303</td>
<td>0.0006 1.0</td>
<td></td>
</tr>
<tr>
<td>PMT72</td>
<td>1–1.4</td>
<td>0.86</td>
<td>24.0</td>
<td>14.8 1.1</td>
<td>48820</td>
<td>3629 9.92</td>
<td>0.06 0.0300</td>
<td>0.0007 0.9</td>
<td></td>
</tr>
<tr>
<td>PMT73</td>
<td>1–1.4</td>
<td>0.72</td>
<td>14.8</td>
<td>10.8 0.4</td>
<td>66902</td>
<td>2478 9.87</td>
<td>0.05 0.0289</td>
<td>0.0003 2.7</td>
<td></td>
</tr>
<tr>
<td>PMT74A</td>
<td>1–1.4</td>
<td>0.40</td>
<td>14.0</td>
<td>9.1 0.5</td>
<td>79400</td>
<td>4363 9.74</td>
<td>0.04 0.0292</td>
<td>0.0002 16.4</td>
<td></td>
</tr>
<tr>
<td>PMT57</td>
<td>1–1.4</td>
<td>0.71</td>
<td>15.0</td>
<td>9.4 0.1</td>
<td>73654</td>
<td>751 9.87</td>
<td>0.03 0.0290</td>
<td>0.0003 3.9</td>
<td></td>
</tr>
<tr>
<td>PMT56</td>
<td>1–1.4</td>
<td>1.03</td>
<td>15.0</td>
<td>9.6 0.3</td>
<td>75265</td>
<td>2352 9.85</td>
<td>0.06 0.0287</td>
<td>0.0006 0.9</td>
<td></td>
</tr>
</tbody>
</table>

*Samples from Santa Bárbara sequence are given from base to top.*

Table 2
Helium isotopes in Terceira samples analyzed by M.D. Kurz and M. Moreira at the Woods Hole Oceanographic Institution (WHOI)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Diameter (mm)</th>
<th>Weight (g)</th>
<th>4He (ncm\textsuperscript{3}STP/g)</th>
<th>R/Ra ±</th>
<th>4He\textsuperscript{3}/He ±</th>
<th>21Ne\textsuperscript{22}/Ne ±</th>
<th>22Ne (pcm\textsuperscript{3}STP/g)</th>
<th>21Ne/22Ne\textsubscript{corr}</th>
</tr>
</thead>
<tbody>
<tr>
<td>PMT24</td>
<td>1.4–2</td>
<td>0.32</td>
<td>16.6</td>
<td>10.0 0.0</td>
<td>71988</td>
<td>617</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PMT71A</td>
<td>1.4–2</td>
<td>0.33</td>
<td>14.4</td>
<td>10.5 0.1</td>
<td>68618</td>
<td>613</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PMT72A</td>
<td>1–3</td>
<td>0.22</td>
<td>4.7</td>
<td>9.9 0.3</td>
<td>73139</td>
<td>1866</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PMT73</td>
<td>1–3</td>
<td>0.15</td>
<td>5.6</td>
<td>10.1 0.2</td>
<td>71264</td>
<td>1736</td>
<td></td>
<td></td>
</tr>
<tr>
<td>PMT74A (Brown oliv)</td>
<td>1–3</td>
<td>0.14</td>
<td>19.2</td>
<td>10.0 0.1</td>
<td>72596</td>
<td>1028</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ACO95-17</td>
<td>1–2</td>
<td>0.27</td>
<td>9.5</td>
<td>9.9 0.1</td>
<td>72676</td>
<td>716</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ACO95-18</td>
<td>1–2</td>
<td>0.23</td>
<td>17.6</td>
<td>9.4 0.1</td>
<td>76622</td>
<td>528</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*ACO95-17 and ACO95-18 are new data from collection of [1].*
Seven samples were also analyzed for helium at the Woods Hole Oceanographic Institution (WHOI) with the MS2 instrument. The analytical procedure can be found in Kurz et al. [55]. $^4$He blank was $\sim 9 \times 10^{-11}$ cm$^3$STP and represented between 2% and 10% of the signal.

3.2. Helium

Helium results from all samples analyzed in Paris are shown in Table 1 and a plot of He concentration versus $^4$He/$^3$He is given in Fig. 3. Total $^4$He concentrations vary from 4.8 to 63 ncm$^3$STP/g. The isotopic ratios range between typical lower MORB values (e.g. sample PMT-74A, $^4$He/$^3$He = 83,050 ± 3820; R/Ra = 8.7 ± 0.4) and a more primitive $^4$He/$^3$He ratio of 48,820 ± 3630 for sample PMT-72 (R/Ra = 14.8 ± 1.1). There is not a simple correlation between isotopic ratios and He concentration, because the later depends on the volume of melt inclusions trapped in phenocrysts as well on the crushing efficiency. However, there is a tendency for low $^4$He/$^3$He ratios to appear in low He concentration samples. Also relevant are the different ratios obtained for distinct olivine separates coming from the same sample or lava flow. Although most of these ratios can be considered similar at the $1\sigma$ uncertainty level, a detailed petrographic and electron-microprobe study [56] shows that, within a single rock sample, olivine crystals with different major element compositions can be characterized by different He isotopic signatures.

Concentrations of helium in the samples analyzed at WHOI are between 4.7 and 19.2 ncm$^3$STP/g (Table 2), consistent with the measurements made in Paris. The $^4$He/$^3$He ratio varies between 68,620 ± 610 and 76,620 ± 530 (R/Ra = 10.5 ± 0.1 and 9.4 ± 0.1 respectively). Data are also reported in Fig. 3. Similar isotopic ratios were obtained for samples PMT-24, PMT-71A and PMT-73 in both laboratories. However, a significant difference in $^4$He/$^3$He ratio can be observed for sample PMT-74A (83,050 vs. 72,600) and, particularly, for the most primitive sample (PMT-72) analyzed in Paris (48,820 vs. 73,140). For sample PMT-74A, the difference is of the same order as that obtained for some olivine groups coming from one single rock sample and can also be ascribed to some distinctive petrographic features (namely the color, see Table 2). Sample PMT-72 is a helium poor sample (4.8 ncm$^3$STP/g) and therefore the difference in

![Fig. 3. $^4$He concentration versus $^4$He/$^3$He in Terceira samples. Data from this study reach a minimum $^4$He/$^3$He ratio of 48,820 ± 3630 (see text). $1\sigma$ uncertainties are shown.](image-url)
between the two laboratories could be attributed to an analytical problem. Our glass machine ARESIBO I has a low resolution power (~ 200) that does not allow a peak separation between HD⁺ and ³He⁺. Nevertheless, the HD⁺ peak is always lower than 2–3 cps (H₂ is lower than 8000 cps in the tube) and the HD contribution is corrected in all the samples, blanks and standards by measuring H₂ together with ³He and ⁴He. We introduced in the mass spectrometer ~ 7 cps of ³He for sample PMT-72 and the ⁴He blank corresponded to ~ 9% of the ⁴He signal. However, sample PMT-71A, that gives the exactly same ratio in both laboratories (68,160 vs. 68,620) has roughly the same introduced ³He (9.5 cps) with also ~ 9% of ⁴He blank. Moreover, the two samples were analyzed in the same week and the machine was very stable during the period of analyses. Therefore we exclude an analytical problem for the large difference in ⁴He/³He measured in samples PMT-72 and PMT-72A.

Despite the possibility for the lowest ⁴He/³He isotopic ratio to represent the Azores plume signature, it is important to emphasize that the low He concentration and the uncertainty relative to the age of PMT-72 lava flow make the ⁴He/³He ratio particularly prone to possible ³He cosmogenic contributions. Indeed, experiments led by Yokochi et al. [57] show that a significant proportion (up to 25%) of crystalline matrix-sited He can be released during crushing trough lattice distortion and shock- and dislocation-induced diffusion. Notwithstanding that an extremely low ³He concentration (lower than 2.2 × 10⁻¹⁴ cm² STP/g, similar to blank values) was measured after melting the olivine powder, we cannot completely discard the possibility of ³He cosmogenic contribution for the low ⁴He/³He isotopic ratio obtained in Paris for this sample.

### 3.3. Neon

Neon results are shown in Table 1. ²²Ne concentrations vary between 0.2 and 16.4 pcm³ STP/g and the three Ne isotope diagram is given in Fig. 4. Six of the olivine separates, representing three different lava flows from the fissure zone (Table 1 and Fig. 2), are placed above the atmospheric values at the 1σ uncertainty level. These samples plot above the MORB line as determined by [58], confirming the contribution of a lower mantle reservoir to the Terceira magma source. The ²⁰Ne/²²Ne ratios are lower than the mantle ²⁰Ne/²²Ne ratio (≥ 12.5) and can be currently interpreted as reflecting atmospheric contamination (e.g. [14,58]).

A regression line (R² ~ 0.9) was calculated for Terceira samples and projected in Fig. 4. The obtained trend line passes through the point with the highest ²⁰Ne/²²Ne ratio (sample PMT-98), which simultaneously represents the best analytical accuracy as a result of minor atmospheric contamination. It is possible to extend this line up to the primitive mantle ²⁰Ne/²²Ne ratio in order to obtain a ²¹Ne/²²Ne ratio corrected for atmospheric contamination (²¹Ne/²²Ne_{corr}) [15]. A corrected ratio of 0.052 was obtained using a solar ²⁰Ne/²²Ne ratio of 13.8 [59]. This value is significantly lower than the MORB ratio (0.075) [60]. It should be noticed that the trend line was calculated after excluding the olivine separate PMT-101-2 as a result of its large uncertainty at the 1σ level.

### 4. Discussion

#### 4.1. Origin of the Azores hotspot

Anomalous topography, gravity, crustal thickness, S-wave velocities and geochemical signatures have
been used as evidence for the existence of a plume in the Azores region ([11,27,61–63] and references therein). However, the relatively shallow character of the S-wave negative anomalous zone (< 300 km), the lack of a well defined hotspot track, the recent 3D reinterpretation of admittance in the area [64] and some geochemical arguments (e.g. [65]) have been used against a deep plume hypothesis applied to Azores. Moreover, Anderson [66] proposed that hotspots could be explained by asthenospheric upwelling as a passive response to breakup which, given the tectonic complexity of the Azores region, could be considered as an attractive working hypothesis.

Noble gas signatures, enabling the discrimination between involvement of degassed and undegassed (primitive) mantle reservoirs in the magma genesis must be considered of utmost importance to unravel the ultimate origin of the Azores magmatism. The previously existent noble gas determinations in the Azores basalts have shown the existence of some unradiogenic He signatures as compared with MORB. For Terceira Island, 4He/3He ratios as low as 64,000 (R/Ra = 11.3) were interpreted as reflecting the existence of a contribution from the deep mantle [1]. A deep plume component is strengthened by the first Ne data now obtained for the Azores archipelago through the study of Terceira Island (21Ne/22Ne corr = 0.052), which are clearly indicative of the involvement of the lower mantle in the Azores magmas. In this context, the shallow character of the S-wave anomaly in the area shall be considered as reflecting the presence of a dying and nowadays untailed plume, the head of which underplates the Azores Plateau [63].

In the next section we will use He–Ne systematics to better constrain the Terceira mantle source.

4.2. Mixing vs. heterogeneous plume source

Given that 3He and 22Ne isotopes are considered to be primordial one might expect that the mantle 3He/22Ne ratio of a mantle reservoir is constant over time (they are both highly incompatible elements), provided that the system remained closed and no He–Ne fractionation occurred. On the other hand, mantle sources characterized by a given 4He/22Ne will be affected by coupled changes in the 4He/3He and 21Ne/22Ne relative to their initial values, since both 4He and 21Ne are produced from decay of U and Th (e.g. [14]). In the following, we will discuss two possibilities for the variations of the He and Ne isotopic ratios. One is a closed system evolution of a heterogeneous mantle plume, whereas a second possibility is mixing between two different sources.

4.2.1. Heterogeneous 3He/22Ne plume source

The assessment of mantle signatures can be made, if an initial isotope composition of the solid Earth is assumed, through the determination of the time-integrated (degassing independent) ratio (designated as 3He/22Ne calc by [24]) inferred from isotope compositions of He and Ne [67]:

$$\frac{3\text{He}}{22\text{Ne}} \text{calc} = \left( \frac{21\text{Ne}}{22\text{Ne}} \text{corr} - \frac{21\text{Ne}}{22\text{Ne}} \text{init} \right) \left( \frac{4\text{He}}{21\text{Ne}} \text{prod} \right) \times \left( \frac{1}{4\text{He/3He}} \text{obs} - \frac{4\text{He}}{3\text{He}} \text{init} \right)$$

where $4\text{He/21Ne}_{\text{prod}}$ represents the mantle production ratio, considered constant and estimated to be equal to $2.2 \times 10^7$ [68], and $4\text{He/3He}_{\text{obs}}$ stands for the measured He isotope ratio. The primordial mantle $4\text{He/3He}_{\text{init}}$ and $21\text{Ne/22Ne}_{\text{init}}$ ratios can be assigned to the solar nebula or to the meteoritic composition Ne–B [21,69]. It is beyond the scope of the paper to discuss the exact primitive ratios of the mantle and, to be consistent, we will consider solar-like lower mantle ratios. Nevertheless, it should be noticed that considering the primitive ratios as Ne–B, the conclusions of this paper do not change.

The mean $3\text{He/22Ne}_{\text{calc}}$ ratios estimated for Terceira samples with $20\text{Ne/22Ne}$ greater than 10 at 1σ uncertainty level, range between 10.6 ± 1.4 and 3.4 ± 2.6 with a mean value around 5 (Fig. 5). These values are intermediate to those estimated for MORB and Iceland (7 and 4 respectively) [20,60,67] and could be interpreted as reflecting the presence of long-lived heterogeneities in the mantle plume source in terms of the $3\text{He/22Ne}$ and (Th+U)/3He ratios (Fig. 5).

However, considering the proximity of Terceira Island to Mid-Atlantic Ridge, we will now evaluate the hypothesis of mixing to generate the low $21\text{Ne/22Ne}$ ratios in association with MORB-like helium ratios.

4.2.2. Binary mixing

The binary mixing model can be tested in a plot of $4\text{He/3He}$ vs. $21\text{Ne/22Ne}_{\text{corr}}$, where the end-members are
considered to be MORB upper mantle and a primitive plume component with solar-like He and Ne isotopic ratios (Fig. 6). The degree of relative $^{3}$He/$^{22}$Ne elemental fractionation is described by the parameter $r = (^{3}$He/$^{22}$Ne)$_{MORB}/(^{3}$He/$^{22}$Ne)$_{plume}$ [20,24].

Hyperbolic mixing lines with $r$-values close to 10 encompass the majority of data implying that, during mixing, the MORB $^{3}$He/$^{22}$Ne ratio was ~10 times higher than in the plume component, here considered as represented by the solar value. $r$-values similar to those here determined for Terceira were reported by [15,20,26,69] for Iceland, Shona, Discovery and East Pacific Rise (17°S) and interpreted in terms of magma mixing. These authors explained the significantly higher $^{3}$He/$^{22}$Ne MORB ratio as the result of He–Ne fractionation promoted by magma degassing in a relatively near surface environment. Indeed, since He and Ne have different melt solubilities (higher for He) the elemental He/Ne ratio of a basaltic melt increases with progressive volatile loss [71]. In the case of Loihi, located in a clear within-plate setting, near linear mixing ($r = 1$) was interpreted as taking place at greater mantle depths, inhibiting the magma degassing processes to occur. Considering that, like Iceland and Shona-Discovery, the Azores hotspot is also located close to an oceanic ridge, a similar model can be considered to explain the high $^{3}$He/$^{22}$Ne MORB ratio during mixing. This idea is endorsed by a three order of magnitude degassing estimated by Moreira and Allègre [33] for MORB in the vicinity of the Azores Plateau. The $^{3}$He/$^{22}$Ne ratios determined for Terceira magmas are thus envisaged as resulting from mixing between a degassed MORB magma and an undegassed plume-like magma (or distillated, see below).

It remains to find an explanation for the apparently less degassed nature of the Terceira plume magma when compared with local MORB. This can be argued in terms of the model proposed by Dixon et al. [72] and Clague et al. [73] for Kilauea and Puna ridge (Hawaii). The model states that the degassed and, as a consequence, denser shallow magmas can sink through a long (~4 km) magma column, thus enabling magma mixing at deeper levels. More recently, this model was also envisaged by Natland [74] for Juan Fernandez Islands (SE Pacific). Such a model can explain the less fractionated nature of plume related magmas, providing a mechanism for magma mixing to occur at a sufficient depth to prevent significant fractionation induced by volatile loss. In our study, we cannot decide if the plume-like magma is not degassed or only degassed following a distillation process as it is the case for Shona and Discovery ridge.

![Fig. 5. $^{21}$Ne/$^{22}$Ne$_{corr}$ ratios plotted against $^{4}$He/$^{3}$He. Straight lines represent the temporal evolution of reservoirs with different $^{3}$He/$^{22}$Ne ratios (indicated on the figure) [20].](image)

![Fig. 6. Binary mixing model between the MORB (for the North Atlantic) and a solar type mantle end-members. To prevent large corrections and uncertainties, only samples with $^{20}$Ne/$^{22}$Ne ratios greater than 10 are plotted as well samples with uncertainties lower than 30%. Terceira data can be fitted by a hyperbolic mixing line with an $r$ value ($= (^{3}$He/$^{22}$Ne)$_{MORB}/(^{3}$He/$^{22}$Ne)$_{solar}$) close to 10. Small dots are from [15,20,26,69] and represent different plume-ridge interactions. Loihi point is plotted from [19].](image)
anomalies (that are also plume-ridge interactions) [75]. Argon measurements are required to evaluate the most feasible hypothesis. We did not analyze argon in our samples in order to decrease the $^{40}$Ar memory during neon analyses.

Another explanation for the high $r$-value is that the plume source component has a $^{3}$He/$^{22}$Ne ratio lower than 2 (i.e. solar-like), 5 to 10 times lower than the ratio in the MORB source reservoir (7–10) [60]. Therefore, in this hypothesis, there is no need to mix degassed MORB magma and the mixing probably occurs at depth. The acceptance of such a model, which attributes a very low $^{3}$He/$^{22}$Ne ratio to the plume source, has fundamental consequences for the origin, distribution and evolution of volatile elements in the Earth as well as for the dynamics of the terrestrial mantle.

5. Conclusions

We presented the first neon isotopic ratios on the Azores archipelago together with new helium data for Terceira Island. Ne and He isotopic data argue for a primitive mantle contribution to Terceira magmas, thus endorsing the models invoking the role of a deep mantle plume in the generation of the Azores Islands. We propose that the Terceira He and Ne signatures result of mixing between two types of magmas: MORB and solar-like plume. As indicated by the positioning of Terceira samples close to the MORB end-member (Fig. 6) this component can be considered as prevalent in the mixing process, particularly concerning helium. The large influence of the MORB end-member can be envisaged as a consequence of the proximity to Mid-Atlantic ridge, explaining the majority of $^{4}$He/$^{3}$He ratios found for Terceira samples only slightly lower than the typical MORB value. Our study highlights the importance of neon data in getting the plume signature of a mixture dominated by MORB.

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Appendix A. Supplementary data


References


