Day, J.M.D., and Hilton, D.R., 2020, Heterogeneous mantle-derived helium isotopes in the Canary Islands and other ocean islands: Geology, v. 49, https://doi.org/10.1130/G47676.1

## **1 SUPPLEMENTARY INFORMATION**

## 2 Detailed methods

3 An ultra-high vacuum (UHV) extraction line was used to process all gas and fluid 4 samples (see Kulongoski & Hilton, 2002). Following release, samples were acidified with 5 phosphorus pentoxide to ensure complete release of  $CO_2$  and, as a result, measured  $CO_2$ 6 abundances correspond to the total dissolved inorganic carbon (DIC) content. A glass 7 trap held at acetone-dry ice temperature was used to isolate water vapor, and a stainless-8 steel trap held at liquid nitrogen temperature was used to separate the condensable gas 9 fraction (dominantly  $CO_2$ ). A non-condensable gas fraction split was collected in a Pyrex 10 glass break-seal for transfer to the VG5400 mass spectrometer for neon isotope 11 measurements (Craig et al., 1993). The remaining non-condensable gas was purified 12 using activated charcoal traps (held at -196°C) and a Ti-getter pump at 700°C. A calibrated aliquot (0.5%) of this fraction was expanded into an AR-glass break-seal for 13 14 transfer to a MAP 215 noble gas mass spectrometer for He isotope analysis. The CO<sub>2</sub> 15 fraction was condensed into a Pyrex break-seal for transfer to a dedicated CO<sub>2</sub> clean-up 16 line.

17 Helium, neon and carbon isotopes were measured using previously detailed 18 protocols (Füri et al., 2010; Barry et al., 2014). Briefly, for He isotope analysis, fractions 19 were released into a preparation line for further purification by exposure to a 750°C hot 20 Ti-getter and activated charcoal trap (held at -196°C), a SAES getter, and a cryogenic 21 trap lined with activated charcoal (held at <20K). Helium was released from the cryogenic trap at 35K, with Ne release at 90K. Sample <sup>3</sup>He/<sup>4</sup>He and <sup>4</sup>He/<sup>20</sup>Ne ratios were 22 23 measured in static mode and calibrated against aliquots of air run at least twice a day 24 under identical conditions. The blank contribution to the <sup>4</sup>He signal was less than 2%. For 25 Ne isotope analysis, samples were run in peak jumping mode. Gas released from the 26 Pyrex break-seals was first exposed to a Ti-sponge getter that was cooled from 750 to 27 450°C, after which the heavy noble gases were trapped onto a stainless-steel frit kept at -28 196°C. Remaining volatiles were exposed to a SAES getter, and He and Ne were trapped 29 onto a He-cooled cryogenic trap (<20K). Neon was released from the trap at 90K. 30 Reproducibility of the Ne isotope analyses was monitored using an air standard measured 31 every eight or less sample runs and sample neon isotope compositions were determined 32 normalized to an air standard. All results consider corrections for procedural blanks and contributions to <sup>20</sup>Ne and <sup>22</sup>Ne from doubly charged <sup>40</sup>Ar and CO<sub>2</sub>, respectively. The 33  $40 \text{Ar}^{++}/40 \text{Ar}^{+}$  ratio was 0.25 and the  $\text{CO}^{2++}/\text{CO}^{2+}$  ratio was 0.013. HF and H<sub>2</sub>O are 34 resolved from <sup>20</sup>Ne, so no corrections for these species were required. Procedural blanks 35 on the <sup>20</sup>Ne signal were less than 2%. 36

37 The CO<sub>2</sub> gas fraction was purified on a separate cleanup and quantification line, 38 constructed from Pyrex glass, enabling resolution from any sulfur-bearing species using a 39 variable temperature trap. Following cleanup, the total amount of  $CO_2$  was measured 40 using a capacitance manometer in a calibrated volume, enabling CO<sub>2</sub> abundance and  $CO_2/{}^{3}$ He ratios to be calculated. Finally,  $CO_2$  was refrozen into a Pyrex tube for transfer 41 to a VG Prism mass spectrometer for carbon isotopic ( $\delta^{13}$ C) analysis. Carbon isotope 42  $\delta^{13}$ C (CO<sub>2</sub>) values are reported relative to the international reference standard, VPDB. 43 44 Precision of individual analyses of standards and samples is better than 0.1<sup>\overline</sup>; however,

- 45 we estimate the accuracy of our  $\delta^{13}$ C determinations (±0.5‰) by repeat analyses of NBS-
- 46 19, itself calibrated relative to VPDB. Procedural blanks were processed on the extraction
- 47 line and comprised less than 1% of sample abundance releases.
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## 49 **References not given in the main manuscript**

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- **Figure S1** Sampling on (a) the summit of Teide, Tenerife and (b) at the CO<sub>2</sub>-rich cold spring in the Cumbre Vieja, La Palma. 57 58



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61 Figure S2 – Neon isotopic compositions of Teide and Taburiente hydrothermal samples compared with Air, mid-ocean ridge basalt (MORB) and Solar reservoirs. Reservoirs 62

from Sarda et al. (1988). 63

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65 Figure S3 – Time-series measurements of helium isotopic composition at the summit of Teide (Tenerife) and from the cold-springs of the 'Taburiente', Barranco De Angustias, 66 67 La Palma. There is no correlation for helium isotope measurements at the summit of Teide, and a weak ( $R^2 = 0.6$ ) negative correlation in La Palma where no measurements 68 have been conducted for >10 years; further analyses in the near future will reveal if this 69 70 relationship has significance. Data are from Peréz et al. (1994) [1994]; Hilton et al. 71 (2000) [1996]; this study [2006]; Alonso et al. (2019) [2016]. Mid-ocean ridge basalt 72 estimate from Graham (2002). Shown are two sigma errors on measurements.