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Abstract

Hotspot magmatism in Polynesia represents a unique natural laboratory for investigating the origin of mantle carbon and the role of recycled crustal materials in plume-related settings. Here, we present new carbon ($\delta^{13}\text{C}$) and noble gas isotope data for fluid inclusions (FIs) hosted in mantle xenoliths and volcanic rocks from selected Polynesian localities, including Savai'i and Ta'u Island (Samoa), Moorea Island (Society Islands), and Tubuai Island (Austral–Cook Islands). The Society samples display $\delta^{13}\text{C}$ values ranging from -10.83‰ to -3.12‰ , overlapping the MORB (Middle Oceanic Ridge Basalt) upper mantle range, whereas the Tubuai xenoliths exhibit significantly heavier compositions, between -1.3‰ and $+1\text{‰}$, coupled with elevated CO₂/³He ratios. Helium isotopes further distinguish the regions, with Samoan samples being characterized by high ³He/⁴He values (up to 13.4 Ra, where Ra is the atmospheric He isotope ratio), indicating contributions from a relatively undegassed lower mantle source, while Society (lava) and Tubuai (xenoliths) exhibit lower than MORB ratios (6–7 Ra). The heavy $\delta^{13}\text{C}$ values observed in Tubuai are interpreted as evidence for crustal carbon recycled into the Austral–Cook mantle. In contrast, the Samoa FIs suggest subsequent isotopic re-setting during pervasive silicate metasomatism. These results demonstrate that mantle carbon isotope signatures are controlled not only by crustal recycling, but also by the sequence and style of metasomatic processes affecting the lithospheric mantle, which are strictly dependent on the Earth's geodynamics.

Keywords

36 *Polynesian islands, OIB, ultramafic xenoliths, fluid inclusions, carbon isotopes, carbon recycling,*
37 *helium isotopes.*

38 **Introduction**

39 The long-term exchange of carbon between the Earth's surface and its interior is regulated by the deep
40 carbon cycle, which links surficial reservoirs to Earth's mantle through plate tectonic processes
41 (Dasgupta and Hirschmann, 2010; Kelemen and Manning, 2015). A fundamental component of this
42 cycle is the recycling of carbon-bearing lithologies through subduction, their storage and modification
43 within the mantle, and their eventual return to the surface via mantle plume volcanism (Dasgupta and
44 Hirschmann, 2010; Kelemen and Manning, 2015). Determining the long-term storage and
45 redistribution of recycled materials within the mantle is therefore critical for assessing the fate of
46 subducted carbon. In this context, geophysical and geochemical observations have led to the
47 recognition of two major deep mantle domains associated with the African and Pacific large low-
48 shear-velocity provinces (LLSVPs), which have been proposed to host distinct proportions and types
49 of recycled material (Garnero et al., 2016; McNamara, 2019; Doucet et al., 2020). Variations in
50 radiogenic isotope systematics and noble gas signatures suggest that these domains may have
51 experienced different recycling histories and mantle evolution pathways (Hart et al., 1992; Doucet et
52 al., 2020; Jackson et al., 2021). Investigating carbon isotope variability in plume-derived magmas
53 from the Pacific mantle domain therefore provides a valuable opportunity to evaluate how large-scale
54 mantle heterogeneity and recycling processes influence the deep carbon cycle.

55 Intraplate Oceanic Island Basalt (OIB) volcanism erupts some of the CO₂-richest magmas globally
56 (Dixon et al., 1997; Hauri et al., 2019) and, although contributing little to the global volcanic CO₂
57 flux to the atmosphere (Fischer et al., 2019), provides access to deep, carbon-bearing mantle
58 reservoirs sampled by mantle plumes (Deines, 2002; Dasgupta and Hirschmann, 2010; Hauri et al.,
59 2019; Pank and Maning, 2019). Polynesia, in the central Pacific, is a vast intraplate province, whose
60 hotspot volcanism offers an ideal context for exploring the mantle carbon cycle. Unlike other OIB
61 localities such as Hawaii or the Canary Islands, that are dominated by a relatively narrow range of
62 mantle components (Staudigel and Schmincke, 1984; Stille et al., 1986; Hoernle et al., 1991; Day and
63 Hilton, 2011; Harðardóttir and Jackson, 2025), Polynesia is one of the most geochemically diverse
64 OIB regions on Earth, encompassing a variety of mantle endmembers with contrasting Sr, Nd, Pb and
65 Hf isotopic compositions (Zindler, 1986; Workman et al., 2004; Jackson et al., 2016; Cordier et al.,
66 2021; Harðardóttir and Jackson, 2025). For example, volcanic rocks from the Society, Samoa,
67 Austral–Cook and Pitcairn Islands have been shown to exhibit geochemical affinities ranging from
68 HIMU (high- $\mu = {}^{238}\text{U}/{}^{204}\text{Pb}$, generally attributed to the long-term evolution of recycled oceanic crust
69 with elevated U/Pb ratios) to Enriched Mantle compositions with moderately radiogenic Pb paired
70 with highly elevated ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ and low ${}^{143}\text{Nd}/{}^{144}\text{Nd}$ ratios (EM2), or relatively unradiogenic Pb (EM1)
71 coupled with moderately elevated ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ and very low ${}^{143}\text{Nd}/{}^{144}\text{Nd}$ ratios (EM2) (Zindler and Hart,
72 1986; Nakamura and Tatsumoto, 1988; Chauvel et al., 1992; Duncan et al., 1994; Jackson et al., 2007,
73 2014; Kogiso, 2013; Ashley et al., 2020; Byerly et al., 2021; Cordier et al., 2021; Harðardóttir and
74 Jackson, 2025). Because of this geochemical diversity, Polynesian magmatism offers a unique
75 opportunity to investigate carbon isotope ($\delta^{13}\text{C}$) systematics across isotopically distinct mantle
76 reservoirs, and to understand the role played by recycled crustal materials in determining the carbon
77 isotope budget of HIMU, EM1 and EM2 volcanism.

78 Despite extensive isotopic work on Polynesian volcanism (Zindler and Hart, 1986; Nakamura and
79 Tatsumoto, 1988; Hauri, 1992; Poreda and Farley, 1992; Farley, 1992; Hauri et al., 1993; Hauri and
80 Hart, 1994; Duncan et al., 1994; Hanyu et al., 1999, 2011; Jackson et al., 2007, 2009, 2014, 2016;
81 Lupton et al., 2009; Kogiso, 2013; Horton et al., 2019; Ashley et al., 2020; Byerly et al., 2021; Cordier
82 et al., 2021; Zhang et al., 2024; Harðardóttir and Jackson, 2025), carbon isotopic results remain
83 scarce. The only available $\delta^{13}\text{C}$ results are for submarine volcanic glasses from Society ($\delta^{13}\text{C}$ from -
84 7.2 to -3.8 ‰; Aubaud et al., 2005, 2006) that, although providing important insights into mantle
85 carbon isotope compositions, are limited to a geographically small area, and potentially affected by
86 isotopic fractionation during magmatic degassing (that shift carbon isotopes toward $\delta^{13}\text{C}$ -depleted
87 compositions; e.g., Lo Forte et al., 2024). In addition to analytical challenges that have long prevented
88 measuring carbon isotopes in melt inclusions (Moussallam et al., 2025), our limited understanding of
89 OIB carbon isotope signature(s) reflects the complex behavior of carbon in the mantle. For example,
90 depending on pressure, temperature and redox conditions (Frost and McCammon, 2008; Stagno and
91 Frost, 2010; Stagno et al., 2013; Dasgupta and Hirschmann, 2010), carbon can be stored in the mantle
92 as CO_2 , CH_4 , CO , and COS , in mineral phases such as carbonates, graphite and diamond, or as a trace
93 constituent of silicate minerals (Deines, 2002; Zhang et al., 2009). The coexistence of these different
94 carbon forms can potentially give rise to isotope fractionation (Cartigny et al., 2014) that, in turn, can
95 eventually complicate interpretations of carbon isotope compositions of mantle rock samples.

96 In spite of these complexities, the $\delta^{13}\text{C}$ compositions of mantle fluids and rocks remain powerful
97 tracers of carbon origin, and are especially suitable for resolving the relative contributions of
98 primordial mantle carbon, subducted crustal material, and surface-derived inputs (e.g., Dasgupta and
99 Hirschmann, 2010; Aiuppa et al., 2017; Duncan and Dasgupta, 2017; Li et al., 2019; Plank and
100 Manning, 2019; Bekaert et al., 2021; Sandoval-Velasquez et al., 2021a, 2021b, 2024, 2025; Lo Forte
101 et al., 2024; Plank and Malinverno, 2026). Previous work has found that mantle xenoliths and mantle-
102 sourced magmatic rocks can preserve nearly primary fluids (as FIs) that record any fluid phase present
103 in the mantle, and eventually allow characterizing the composition of volatile-rich melts infiltrating
104 into, and modifying, the composition of the mantle (Vance et al., 1989; Deines, 1992, 2002; von
105 Gehlen, 1992; Matsumoto et al., 2001; Williams and Hemley, 2001; Dunai and Porcelli, 2002;
106 Gautheron and Moreira, 2002; Li et al., 2008; Nuccio et al., 2008; Dasgupta and Hirschmann, 2010;
107 Marty, 2011; Marty, 2012; Boudoire et al., 2018; Rizzo et al., 2018; Hauri et al., 2019; Regier et al.,
108 2020; Sandoval-Velasquez et al., 2021b, 2023, 2024; Remigi et al., 2023; Lo Forte et al., 2024).

109 Here, we focus on the study of carbon isotopes in FIs trapped in mantle xenoliths and volcanic rock
110 samples from Samoa (Savai'i and Ta'u), Society (Moorea) and the Austral-Cook Islands (Tubuai)
111 (Fig. 1). We present new data on CO_2 concentrations and carbon isotopic compositions (coupled to
112 noble gas) in FIs that are used to (i) characterize carbon reservoirs in the Polynesian mantle, (ii) assess
113 the role of recycled crustal carbon in the mantle source(s), and (iii) determine whether the HIMU and
114 EM mantle components sampled by Polynesian hotspot volcanism are associated with distinct carbon
115 isotopic signatures. Our results reveal significant carbon isotope variability across Polynesian mantle
116 domains, with unusually heavy $\delta^{13}\text{C}$ values beneath the Austral–Cook Islands indicating the
117 incorporation of recycled crustal carbon. In contrast, a MORB-like $\delta^{13}\text{C}$ signature beneath Samoa
118 (and possibly the Society Islands) is interpreted to reflect substantial metasomatic overprinting and
119 resetting of carbon isotopic signatures. Our observations indicate that mantle carbon is controlled not
120 only by crustal recycling, but also by the nature and timing of metasomatic processes affecting the
121 mantle.

122 Volcanic and geochemical characteristics of the Samoa, Society, and Austral– 123 Cook Islands

124 OIB volcanism in Polynesia represents one of the most remarkable expressions of intraplate
125 magmatism on Earth. Situated within the South Pacific Superswell, this province includes the Samoa,
126 Society and Austral–Cook chains (Fig. 1; Adam and Bonneville, 2005; McNutt and Fischer, 2013;
127 Adam et al., 2014). Seismic data suggest that the hotspots, which are positioned on top of the South
128 Pacific Superswell, are underlain by a broad region of low velocities and anomalous buoyancy, while
129 geochemical evidence demonstrates that its source is highly heterogeneous (McNutt and Judge, 1990;
130 Fukao et al., 1992; Sichoix et al., 1998; Zhao, 2001; Workman et al., 2004; Jackson and Dasgupta,
131 2008; Kogiso, 2013; McNutt and Fischer, 2013; Adam et al., 2014; Jackson et al., 2016; Cordier et
132 al., 2021). In particular, Polynesia hosts three of the canonical mantle endmembers: HIMU, related
133 to recycled oceanic crust, and both EM1 and EM2, which are associated with recycled subcontinental
134 lithologies (Zindler and Hart, 1984; Chauvel et al., 1992; Workman et al., 2004; Bonneville et al.,
135 2006; Jackson and Dasgupta, 2008; Kogiso, 2013; White and Duncan, 2013; Jackson et al., 2016;
136 Cordier et al., 2021).

137 The Samoa archipelago (Fig. 1) trends east–west just north of the Tonga Trench (Konter and Jackson,
138 2012). Isotopically, Samoan shield lavas are dominated by elevated $^{87}\text{Sr}/^{86}\text{Sr}$, low $^{143}\text{Nd}/^{144}\text{Nd}$, and
139 intermediate Pb isotope ratios, defining a classic EM2 mantle source (Workman et al., 2004; Jackson
140 and Dasgupta, 2008). Submarine lavas from the island of Savai'i, the largest island, represents the
141 clearest expression of this component, suggesting a mantle reservoir enriched by recycled terrigenous
142 sediments (Workman et al., 2004; Jackson et al., 2007). In contrast, Ta'u, at the eastern end of the
143 volcanic segment, departs from the Savai'i compositional field (Workman et al., 2004; Jackson et al.,
144 2014, 2016), and is characterized by more radiogenic Pb isotopes, elevated He isotopes, and
145 geochemically depleted Nd isotopes, suggesting mixing between EM2 and a HIMU- or FOZO (Focal
146 Zone)-like component.

147 The Society Islands volcanic chain extends for approximately 700 km and comprises nine main
148 volcanic islands (Fig. 1), in addition to several atolls and seamounts. The archipelago formed during
149 the last ~5 Ma (Cheng et al., 1993; Cordier et al., 2021). Tahiti displays volcanic ages of
150 approximately ~1.5 Ma (Cheng et al., 1993; Cordier et al., 2021), whereas Moorea volcanism is ~1.85
151 Ma (Bechon et al., 2026). Basalts from the Society Islands display a wide compositional range
152 characterized by low Ce/Pb and Nb/U ratios and relatively high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios relative to most OIBs.
153 These geochemical characteristics also reflect an EM2 mantle source, commonly interpreted as
154 resulting from the contribution of recycled terrigenous sediments into the mantle source region
155 (Cordier et al., 2021).

156 The Austral–Cook chain (MacDonald hotspot) is the archetype of HIMU geochemistry in Polynesia
157 (Chauvel et al., 1992; Hanyu and Kaneoka, 1997; Parai et al., 2009; Hanyu et al., 2011a, 2011b).
158 Tubuai, an island made of two eroded volcanic edifices active between 12 and 6 Ma, preserves
159 sequences of alkaline basalts, basanites, and dike swarms containing ultramafic xenoliths (Caroff et
160 al., 1997). The isotopic compositions of these rocks are markedly different from those of Samoa and
161 the Society: $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are low, while $^{206}\text{Pb}/^{204}\text{Pb}$ values are radiogenic, corresponding to the
162 classic HIMU mantle signature (Chauvel et al., 1992; Hanyu and Kaneoka, 1997; Parai et al., 2009;
163 Hanyu et al., 2011a, 2011b). HIMU is interpreted to reflect a mantle source containing ancient
164 recycled oceanic crust, which has experienced U/Pb fractionation and long-term radiogenic ingrowth

165 of Pb isotopes (Hofmann and White, 1982; Stracke et al., 2005; Jackson and Dasgupta, 2008). Tubuai
166 falls close to the HIMU endmember in radiogenic isotope space. Other islands of the Austral–Cook
167 system, including Mangaia and Rurutu, display similar isotopic traits, confirming that this chain
168 predominantly samples the HIMU reservoir (Hanyu et al., 2011a, 2011b).

169

170 **Materials and methods**

171 We report on results for nine ultramafic xenoliths and three mafic volcanic rocks. These include four
172 harzburgites from Savai'i (Samoa), three magmatic dunites and one picrite from Ta'u (Samoa), one
173 basalt from Moorea (Society) and two harzburgites from Tubuai (Austral-Cook Islands). The Society
174 lavas are part of the NMNH collection provided by the Department of Mineral Sciences of the
175 Smithsonian Institution. Petrographic and geochemical characteristics, together with mineral
176 chemistry data of the xenolith suites from Savai'i, Ta'u, and Tubuai, have been analyzed and reported
177 previously (Hauri et al., 1993; Hauri and Hart, 1994; Jackson et al., 2016; Ashley et al., 2020; Byerly
178 et al., 2021).

179 The analysis of CO₂ and noble gas isotopic in FIs hosted in minerals from this rock suite was carried
180 out at the noble gas and stable isotope laboratories of the Istituto Nazionale di Geofisica e
181 Vulcanologia (INGV), Sezione di Palermo. The preparation and analytical procedures followed
182 established protocols (Gennaro et al., 2017; Rizzo et al., 2018; Sandoval-Velasquez et al., 2023,
183 2024). Bulk samples were crushed and sieved, and individual crystal fractions larger than 0.25 mm
184 were handpicked under a binocular microscope. Between 1 and 3 g aliquots of olivine and pyroxene
185 crystals were selected from all samples and subjected to ultrasonic cleaning in diluted nitric acid
186 (6.5%), deionized water, and acetone. The extraction of CO₂ from fluid inclusions was performed
187 using a high-vacuum crushing system. The apparatus consists of a stainless-steel piston crusher
188 capable of exerting pressures of ~200 bar on a steel plate, a pumping system to maintain vacuum, and
189 a glass collector for cryogenic trapping of released gases. Prior to crushing, the system was heated
190 and evacuated to remove adsorbed species. During crushing, the glass collector was immersed in
191 liquid nitrogen at –196 °C to condense CO₂ and other condensable gases. Subsequent purification
192 was achieved using a glass purification line to minimize adsorption and ensure separation of CO₂
193 from other gases. CO₂ and H₂O were initially condensed at –196 °C, while O₂, N₂, and noble gases
194 were pumped away. Controlled heating to –100 °C allowed CO₂ to be released in gaseous form, while
195 H₂O remained frozen. The purified CO₂ was finally collected in a glass tube, equilibrated to
196 atmospheric pressure with pure helium, and transferred to the stable isotope laboratory (further details
197 are reported in Gennaro et al., 2017). Carbon isotope ratios (¹³C/¹²C) were determined using a Thermo
198 Delta Plus XP isotope ratio mass spectrometer (IRMS) interfaced with a Trace GC and a Thermo
199 GC/C III combustion unit. Results are reported in δ¹³C notation relative to the Vienna Pee Dee
200 Belemnite (V-PDB) standard. The analytical uncertainty for δ¹³C was better than ±0.2 ‰ (1σ)

201 For noble gas analyses, aliquots of cleaned crystals were loaded into an ultra-high vacuum (UHV)
202 crushing device capable of holding up to six samples. The system was baked at 120 °C for 48 h under
203 vacuum. Fluid inclusion gases were released in a single crushing step at ~200 bar and room
204 temperature (~21 °C). This approach minimizes the release of secondary helium (³He cosmogenic
205 and ⁴He radiogenic) from the crystal lattice (Graham, 2002; Rizzo et al., 2018). The released gases
206 entered a purification line, where CO₂ and H₂O were first removed by cryogenic trapping at –196 °C.

The remaining gas mixture underwent further purification to separate noble gases from reactive species (N₂, H₂, H₂O, CO₂), which were removed using heated getter traps. Subsequent cryogenic separation allowed isolation of He, Ne and Ar. Noble gas isotopic compositions were measured using a dedicated mass spectrometric setup. Two split-flight tube mass spectrometers (Helix SFT, Thermo) were used for He and Ne isotopes, while a multicollector Argus mass spectrometer (GVI) was employed for Ar isotopes. ³He, ⁴He, ²⁰Ne, ²¹Ne, ⁴⁰Ar, ³⁸Ar, and ³⁶Ar contents were measured. The ³He/⁴He ratio was further corrected for atmospheric contamination based on the measured ⁴He/²⁰Ne ratio, as follows:

$$R_c/R_a = ((R_M/R_a)(\text{He/Ne})_M - (\text{He/Ne})_A)/((\text{He/Ne})_M - (\text{He/Ne})_A) \quad \text{eq. 1}$$

where subscripts M and A refer to measured and atmospheric theoretical values, respectively (⁴He/²⁰Ne_A = 0.318). The corrected ³He/⁴He ratios are reported as R_c/R_a values.

⁴⁰Ar was corrected for air contamination (⁴⁰Ar*) assuming that the measured ³⁶Ar has an atmospheric origin as follows:

$$^{40}\text{Ar}^* = ^{40}\text{Ar}_{\text{sample}} - [^{36}\text{Ar}_{\text{sample}} \times (^{40}\text{Ar}/^{36}\text{Ar})_{\text{air}}] \quad \text{eq. 2}$$

$$^{40}\text{Ar}_{\text{air}} = ^{40}\text{Ar}_{\text{sample}} - ^{40}\text{Ar} \quad \text{eq. 3}$$

Analytical uncertainties (1σ) for the measured isotope ratios were as follows: <2.7% for ³He/⁴He, <2.0% for ⁴⁰Ar/³⁶Ar, and <1.7% for ³⁸Ar/³⁶Ar.

Results

The elemental and isotopic compositions of CO₂ and noble gases in mantle xenoliths and lavas from Savai'i and Ta'u (Samoa), Moorea (Society) and Tubuai (Austral-Cook) exhibit significant variability (Tables 1–2). In FIs, CO₂ is the dominant volatile phase (Fig. 2A), consistent with previous studies (Hauri et al., 1993; Hauri and Hart, 1994; Ashley et al., 2020). The δ¹³C compositions of fluid inclusions in xenoliths from Samoa and Tubuai identify two distinct isotopic domains (Fig. 2A-B). Samoa and Society δ¹³C values range from –10.83‰ to –3.12‰, extending well into the typical MORB range (–8‰ to –4‰; Sano and Marty, 1995), while Tubuai samples exhibit distinctly heavier signatures, with δ¹³C values between –1.33‰ and +1.00‰ and elevated CO₂/³He ratios (>4·10¹⁰).

Helium isotope ratios corrected for atmospheric contamination (R_c/R_a) in FIs of crystal separates from Samoa, Tubuai and Society generally fall within the range of values reported in the literature (7–25 Ra, 5–10 Ra, 5.5–9 Ra, respectively; Farley et al., 1992; Poreda and Farley, 1992; Hanyu et al., 1999, 2011; Marty and Dauphas, 2003; Jackson et al., 2007, 2009, 2016; Lupton et al., 2009; Parai et al., 2009; Zhang et al., 2024). The lowest ³He/⁴He ratios (Fig. 3A-B) are observed in the Moorea and Tubuai samples (3.3–6.5 Ra), while Samoa samples yield distinctly higher ratios, ranging from 10.0 to 13.4 Ra. As expected, Samoan samples exceeded the typical MORB range (8 ± 1 Ra; Graham, 2002) suggesting the involvement of a more primitive mantle component.

243 Discussion

244 Trace elements in mantle xenoliths from Samoa, Society and Austral-Cook have been taken to
245 demonstrate that metasomatism has played a fundamental role in generating the observed chemical
246 heterogeneity of the Polynesian lithospheric mantle (Hauri, 1992; Poreda and Farley, 1992; Hauri et
247 al., 1993; Farley et al., 1994; Farley, 1995; Dieu, 1995; Burnard et al., 1998; Jackson et al., 2009,
248 2016; Ashley et al., 2020; Byerly et al., 2021). For example, orthopyroxene and clinopyroxene trace
249 element compositions from mantle xenoliths record strong enrichment in incompatible elements and
250 pronounced fractionation patterns, interpreted as due to the interaction with metasomatic melts
251 (Hauri, 1992; Hauri et al., 1993; Ashley et al., 2020; Byerly et al., 2021). The occurrence of LREE-
252 enriched patterns, and the decoupling of high field strength elements (HFSE), provide evidence for
253 the involvement of silicate and carbonatitic melts likely derived from recycled, volatile-rich materials
254 stored within the convecting mantle (Hauri et al., 1993; Farley et al., 1994; Farley, 1995; White and
255 Duncan, 1996; Coltorti et al., 1999; Jackson et al., 2009, 2016; Byerly et al., 2021). Radiogenic
256 isotopes (Sr-Pb-Nd) have provided complementary evidence for the involvement of recycled
257 materials throughout Polynesia, since lavas and mantle xenoliths from Samoa, Society, and the
258 Austral-Cook chain define isotopic arrays involving EM and HIMU mantle components that are
259 commonly interpreted as reflecting the recycling of sedimentary and oceanic crustal materials into
260 the mantle (Fig 4; White and Duncan, 1995; Jackson et al., 2007, 2009, 2016; Ashley et al., 2020;
261 Byerly et al., 2021).

262 Although crustal recycling is consistently observed in the three localities, the metasomatic evolution
263 of each mantle domain seems to differ markedly. In Samoa, trace element data and mineral-scale
264 observations in ultramafic xenoliths indicate a multi-stage metasomatic history, in which early
265 carbonatitic metasomatism is subsequently overprinted by silicate (basaltic) metasomatism in some
266 Savai'i harzburgites (Burnard et al., 1998; Ashley et al., 2020; Byerly et al., 2021). In the case of
267 Tubuai, harzburgites preserve carbonatitic metasomatism (with no signs of silicate melts infiltrating
268 the mantle), while the ultramafic xenoliths from Tahiti (Society) show high Ti/Eu and low Ce/Yb,
269 also consistent with silicate metasomatism or coexisting carbonatitic and silicate metasomatism
270 (Akizawa et al., 2017; Byerly et al., 2021). This temporal sequence of metasomatic events is critical,
271 as it implies that primary metasomatic signatures introduced by carbonatitic melts can be partially or
272 completely overprinted during later melt-rock reactions. As discussed in the following sections, our
273 results highlight the role of metasomatic evolution in determining whether recycled carbon signatures
274 are retained or modified in the mantle.

275

276 $\delta^{13}\text{C}$ systematics in Tubuai: preservation of a HIMU carbon signature

277 Tubuai xenoliths display heavy $\delta^{13}\text{C}$ values (-2 to $+1\text{‰}$) coupled with high $\text{CO}_2/{}^3\text{He}$, strongly
278 suggesting that the local mantle has been contaminated by isotopically heavy, carbon-rich melts/fluids
279 (Fig. 2). Comparable $\delta^{13}\text{C}$ results has been observed in other HIMU-like OIB systems, providing a
280 useful framework for interpreting the Tubuai data. For example, CO_2 -rich FIs in mantle xenoliths
281 from El Hierro and Lanzarote in the Canary Islands show similar $\delta^{13}\text{C}$ values, ranging from -2.4 to
282 $+1.0\text{‰}$ (Sandoval-Velasquez et al., 2021b, 2024). These relatively heavy carbon isotope signatures,
283 which extend well above the typical MORB mantle range, have been interpreted as evidence for the
284 addition of recycled crustal carbon into the local mantle source via metasomatism driven by carbon-

bearing fluids. Consistent with the HIMU-like character of the Canary Islands mantle, this positive $\delta^{13}\text{C}$ signal has been attributed to the incorporation of carbon derived from recycled altered oceanic crust (AOC; Fig. 2B) and/or oceanic lithosphere (OL; Sandoval-Velasquez et al., 2021b). It is worth noting that subducted AOC and OL play a fundamental role in the transfer of crustal carbon into the mantle, as hydrothermal alteration and microbial methanogenesis at mid-ocean ridges leads to extensive carbonate precipitation and CO_2 inorganic reduction ultimately yielding highly variable $\delta^{13}\text{C}$ values in these lithologies (from -24‰ to $+10.3\text{‰}$; Alt et al., 2013; Mason et al., 2017; Plank and Manning, 2019; Lopez et al., 2023; Plank and Malinverno, 2026). As for the Canary Islands, if the incorporation of carbonates in marine sediments, AOC and/or OL is the cause of the $\delta^{13}\text{C}$ variability of Tubuai samples, then the metasomatizing carbonatite melts inferred from the petrological observations should be crustal in origin, which would align with the HIMU-like signature of the local mantle.

297

298 **Metasomatic control on carbon isotope signatures in the Samoa and Society upper mantle**

299 Despite the strong evidence for carbonatitic metasomatism and the existence of recycled materials in
300 the Samoa and Tubuai upper mantle (Hauri et al., 1993; Farley et al., 1994; Farley, 1995; Jackson et
301 al., 2009, 2016; Byerly et al., 2021), the $\delta^{13}\text{C}$ compositions of Savai'i xenoliths remain restricted to
302 -8 to -3‰ , clustering within the range of typical upper-mantle compositions and overlapping those
303 of the Society lavas (Fig. 2). Considering the similarities in trace-element signatures, which indicate
304 the influence of broadly comparable carbonatitic metasomatic processes in both regions (Hauri et al.,
305 1993), it is plausible that the Savai'i mantle initially hosted isotopically heavy carbon analogous to
306 that preserved in the Tubuai xenoliths. If our hypothesis is correct, then the present-day MORB-like
307 upper mantle $\delta^{13}\text{C}$ of Samoa samples may record the effects of subsequent metasomatic overprinting,
308 which is consistent with the petrological evidence for superimposed MORB-like silicate melt
309 infiltration observed in some of the Savai'i harzburgites (Hauri and Hart, 1994; Ashley et al., 2020).
310 In fact, silicate melt–rock interactions are well known to occur through diffusive exchange and
311 dissolution–reprecipitation reactions mainly involving minerals such as clinopyroxene and
312 orthopyroxene (Navon and Stolper, 1987; Hauri, 1992; Kelemen et al., 1997, 1998; Dijkstra et al.,
313 2003; Ashley et al., 2020). During silicate melt migration, repeated equilibration between melt and
314 wall rock may have led to chromatographic evolution of melt compositions and progressive chemical
315 homogenization of the Samoan lithosphere (Navon and Stolper, 1987; Hauri, 1992; Hauri and Hart,
316 1994, 1997; Dijkstra et al., 2003; O'Reilly and Griffin, 2013). Consistent with this interpretation,
317 Byerly et al. (2021) showed that Samoan xenoliths dominated by silicate metasomatism exhibit
318 substantially lower Sr–Nd isotopic heterogeneity than xenoliths affected primarily by carbonatitic
319 metasomatism, suggesting that silicate melts promoted partial isotopic homogenization of pre-
320 existing mantle heterogeneities. Because carbon is highly mobile in silicate melts and can readily
321 exchange between melts, FIs, and mantle minerals (Dasgupta and Hirschmann, 2010; Guillot and
322 Sator, 2011), we propose that these processes may have been particularly effective at modifying
323 carbon isotopes. Thus, whereas silicate metasomatism likely induced only a partial reset of radiogenic
324 isotope systematics, sufficient to reduce mineral-scale heterogeneity without completely erasing the
325 EM2-like characteristics of the mantle source, it may have produced a much more extensive resetting
326 of $\delta^{13}\text{C}$, driving carbon isotope compositions toward mantle-like values (Fig. 4). This interpretation
327 is consistent with the $\delta^{13}\text{C}$ similarity between Savai'i xenoliths and Ta'u dunites (Fig. 2A-B). Because

328 Ta'u dunites represent magmatic cumulates that are in trace-element equilibrium with their host
329 silicate lavas (Ashley et al., 2020) (Fig. 2), their mantle-like $\delta^{13}\text{C}$ is interpreted to reflect the isotopic
330 signature of upper-mantle-derived silicate melts. The similar $\delta^{13}\text{C}$ trend observed in dunites and
331 associated picrites in the Canary Islands (e.g., Sandoval-Velasquez et al., 2024) also corroborate this
332 interpretation. Together, these observations suggest that the present-day Samoan magmatism
333 originated from a mantle source that had already been overprinted and isotopically homogenized by
334 the most recent episode of silicate metasomatism. The overlapping $\delta^{13}\text{C}$ of the Ta'u dunites/basanite
335 and Savai'i xenoliths are then consistent with a common silicate melt influence, suggesting that the
336 Samoa mantle probably inherited the homogenized mantle-like $\delta^{13}\text{C}$ signature of the magmatic
337 component.

338 If correct, this model has broader implications for understanding the contrasting carbon-isotope
339 systematics of the South Pacific mantle. The difference between Tubuai and Samoa/Society mantle
340 is further illustrated by Fig. 4, where Tubuai samples define a trend toward HIMU-like compositions,
341 characterized by radiogenic Pb isotopes and elevated $\delta^{13}\text{C}$ values, indicating a strong contribution
342 from recycled oceanic crust. In contrast, Samoa plot within EM-type (EM1–EM2) isotopic fields
343 (also suggesting that recycled components are present in the Samoa mantle) while retaining MORB-
344 like $\delta^{13}\text{C}$ values. Society samples also occupy EM-type isotopic fields, indicating the presence of
345 recycled components in their mantle source. However, the available $\delta^{13}\text{C}$ data for Society are derived
346 exclusively from submarine volcanic glasses and may have been modified by magmatic degassing
347 (Lo Forte et al., 2024). Consequently, the primary carbon isotope composition of the Society mantle
348 remains poorly constrained. While its radiogenic isotope characteristics suggest similarities with the
349 Samoan mantle domain, it remains unclear whether the apparent overlap between Society and Samoa
350 $\delta^{13}\text{C}$ values reflects primary mantle source characteristics or isotopic fractionation induced by
351 magmatic degassing.

352 Despite these uncertainties, the available data shows a decoupling between carbon isotopes and
353 radiogenic tracers, particularly in Samoa and possibly also in the Society Islands (as similarly
354 observed in mantle xenoliths from Central Mexico and the Canary Islands; Sandoval-Velasquez et
355 al., 2021b, 2022, 2024) and suggests that carbon isotopes are more susceptible to modification during
356 subsequent metasomatic reworking than radiogenic isotope systems. The combined trace element,
357 radiogenic isotopes and $\delta^{13}\text{C}$ data suggest that the variability of carbon isotopes in the mantle is
358 controlled not only by the presence of recycled carbon, but also by the sequence and nature of
359 metasomatic processes. We caution, however, that our interpretations are based on the premise that
360 similar carbonatitic metasomatism affected both the Samoa and Tubuai mantle domains, as evidenced
361 by trace element and the Sr-Pb isotopic systematics observed in clinopyroxene crystals, inclusions,
362 and interstitial glasses in ultramafic xenoliths (Hauri et al., 1993). We also acknowledge that the
363 interpretations for Tubuai and Society are constrained by a limited number of $\delta^{13}\text{C}$ measurements,
364 and that additional studies are required to assess our hypothesis and better characterize the carbon
365 isotopic signature of the Polynesian mantle.

366

367 Helium constraints on mantle source

368 Helium isotopes provide independent constraints on mantle source characteristics and allow direct
369 comparison with the $\delta^{13}\text{C}$ dataset. The Rc/Ra values shown in Fig. 3A reveal that most of our samples

370 fall within the same fields of lavas and ultramafic xenoliths reported in the literature; only samples
371 TBA-5 and NMNH-11672 seem partially decoupled because of their lower Rc/Ra values. In Savai'i
372 and Ta'u, $^3\text{He}/^4\text{He}$ ratios span a relatively wide range (at elevated values from 9 to 14 Ra), confirming
373 the presence of a relatively undegassed or deep mantle component. Society and Tubuai samples,
374 instead, show more radiogenic ratios (4-7 Ra) below the upper mantle range.

375 It is worth noting that samples TBA-5 and NMNH-11672 exhibit not only lower $^3\text{He}/^4\text{He}$ ratios but
376 also markedly reduced ^4He concentrations, suggesting a possible secondary helium loss rather than a
377 primary mantle signature (Burnard, 2004; Yamamoto et al., 2009; Sandoval-Velasquez et al., 2021a).
378 Because helium is extremely mobile, partial melting and metasomatic events (in the case of TBA-4-
379 5 harzburgite) or post eruptive processes may preferentially remove ^3He relative to ^4He owing to their
380 different diffusivities ($D_{^3\text{He}}/D_{^4\text{He}} = 1.15$; Trull et al., 1993; Burnard, 2004; Yamamoto et al., 2009).
381 Thus, and consistent with the trend observed in Fig. 3A, in case of diffusive He loss, a systematic
382 decrease in both $^3\text{He}/^4\text{He}$ and helium contents is expected. Moreover, helium loss would also explain
383 the extremely high $\text{CO}_2/^3\text{He}$ of TBA-4-5 olivines, which approach the crustal limestone range (Fig.
384 3B). Based on the above, we conclude that samples TBA-5 and NMNH-11672 likely record
385 secondary modification of their noble gas systematics and these, therefore, do not represent the
386 primary helium signature of the local mantle.

387 Of particular interest is that the Rc/Ra vs. $\delta^{13}\text{C}$ association (Fig. 3) suggests a decoupling between
388 helium and carbon isotopes, a characteristic previously documented in ultramafic xenoliths from other
389 OIB and continental settings (e.g., Rizzo et al., 2018; Sandoval-Velasquez et al., 2021b, 2021a, 2023,
390 2024; Lo Forte et al., 2024). Notably, samples with the highest Rc/Ra values (Samoa)—interpreted
391 as having the strongest plume contribution—and some with the lowest Rc/Ra (Society)—interpreted
392 as having the strongest ^4He radiogenic contribution—exhibit broadly overlapping $\delta^{13}\text{C}$ values within
393 the mantle range. Moreover, Tubuai samples display systematically heavier $\delta^{13}\text{C}$ (-2 to $+1\text{‰}$) even
394 if Rc/Ra values are comparatively similar to those measured in Society lavas. Further evidence for
395 decoupling between carbon abundance and isotopic composition is provided by the relationship
396 between $\text{CO}_2/^3\text{He}$ and $\delta^{13}\text{C}$ (Fig. 2B). Tubuai samples define a trend toward higher $\delta^{13}\text{C}$ at relatively
397 elevated $\text{CO}_2/^3\text{He}$ ratios, consistent with the addition of a carbon component derived from recycled
398 oceanic crust. In contrast, Samoa and Society samples exhibit a broad range of $\text{CO}_2/^3\text{He}$ ratios
399 (including ratios comparable to those of sample TBA-2-1 from Tubuai) while showing relatively
400 homogeneous $\delta^{13}\text{C}$. This observation suggests that variations in $\text{CO}_2/^3\text{He}$ do not necessarily reflect
401 changes in carbon source and may instead be influenced by other processes capable of fractionating
402 carbon from noble gases.

403 A plausible explanation for the variable $\text{CO}_2/^3\text{He}$ measured in FIs of mantle xenoliths is the high
404 degree of partial melting experienced by the Savai'i mantle. In fact, previous petrological
405 observations on the same samples pointed to $\sim 17\text{--}21\%$ partial melting (Hauri and Hart, 1994; Ashley
406 et al., 2020), which is consistent with the near-complete exhaustion of clinopyroxene in our
407 harzburgites (Hauri, 1994; Ashley et al., 2020). Such extensive melting is expected to significantly
408 modify volatile systematics, particularly for species characterized by different mineral/melt
409 partitioning behavior. Experimental studies have shown that carbon, helium and argon exhibit distinct
410 mineral/melt partition coefficients ($^{(\text{min}/\text{melt})}D_i$) for upper mantle minerals such as olivine and
411 clinopyroxene (Heber et al., 2007; Rosenthal et al., 2015). In olivine, reported partition coefficients
412 are $D_{\text{C}} = 0.0007 \pm 0.0004$, $D_{\text{Ar}} = 0.0011 \pm 0.0006$ and $D_{\text{He}} = 0.00017 \pm 0.00013$ whereas

413 clinopyroxene exhibits $D_C = 0.0005 \pm 0.0004$, $D_{Ar} = 0.0011 \pm 0.0007$ and $D_{He} = 0.0002 \pm 0.0002$.
414 These data indicate that carbon and argon are less incompatible than helium during mantle melting.
415 Consequently, progressive melt extraction is expected to fractionate carbon and argon from helium,
416 leading to systematic increases in $CO_2/{}^3He$ ratios and decreases in ${}^4He/{}^{40}Ar^*$ ratios (Fig. 2B and Table
417 1). Savai'i xenoliths with the highest $CO_2/{}^3He$ consistently show the lowest ${}^4He/{}^{40}Ar^*$ (samples SAV-
418 1-42 Opx, SAV-1-13 Opx and SAV-1-28 Opx; Table 1). In conclusion, elevated and heterogeneous
419 $CO_2/{}^3He$ ratios observed in the Savai'i harzburgites primarily reflects melting-induced volatile
420 fractionation rather than distinct carbon sources.

421

422 Conclusions

423 Our new FIs dataset from Samoa (Savai'i and Ta'u), Society (Tahiti and Moorea), and Austral-Cook
424 (Tubuai) islands demonstrates that carbon isotopes in the Polynesian mantle are strongly
425 heterogeneous. The Tubuai xenoliths preserve evidence of isotopically heavy carbon ($\delta^{13}C = -1.33\text{‰}$
426 and $+1.00\text{‰}$) associated with HIMU-like mantle domains, supporting models in which recycled
427 oceanic lithologies contribute carbon-rich metasomatic fluids/melts to the mantle. The preservation
428 of these signatures suggests relatively limited metasomatic overprinting and indicates that
429 carbonatitic metasomatism can retain identifiable recycled carbon fingerprints. In contrast, the Samoa
430 mantle records substantial decoupling between carbon isotopes and radiogenic tracers. Although Sr-
431 Nd-Pb isotopes and trace element systematics clearly indicate the presence of recycled materials and
432 the influence of similar carbonatitic melts in the upper mantle, their mantle-like $\delta^{13}C$ values (-7.98‰
433 to -3.12‰) suggest that subsequent processes may have reset the carbon isotopic composition. We
434 propose that subsequent metasomatism (as supported by petrological observations), triggered by the
435 infiltration of silicate melts with MORB-like upper mantle carbon signatures, would have promoted
436 efficient isotopic re-setting and homogenization of carbon within the local lithosphere. These
437 observations highlight that carbon isotopes may be more susceptible to metasomatic resetting than
438 radiogenic isotope systems, particularly during prolonged melt-rock interaction involving mobile
439 MORB-like silicate melts.

440 Noble gases further support this interpretation, showing that variations in helium isotopes and
441 $CO_2/{}^3He$ ratios are not systematically coupled with carbon isotope compositions. In particular, the
442 variability of $CO_2/{}^3He$ ratios in Samoa likely reflects partial melting-related volatile fractionation.
443 This decoupling indicates that primordial helium signatures and carbon isotope compositions may
444 respond differently to partial melting, metasomatism, and melt transport processes.

445

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FIGURES

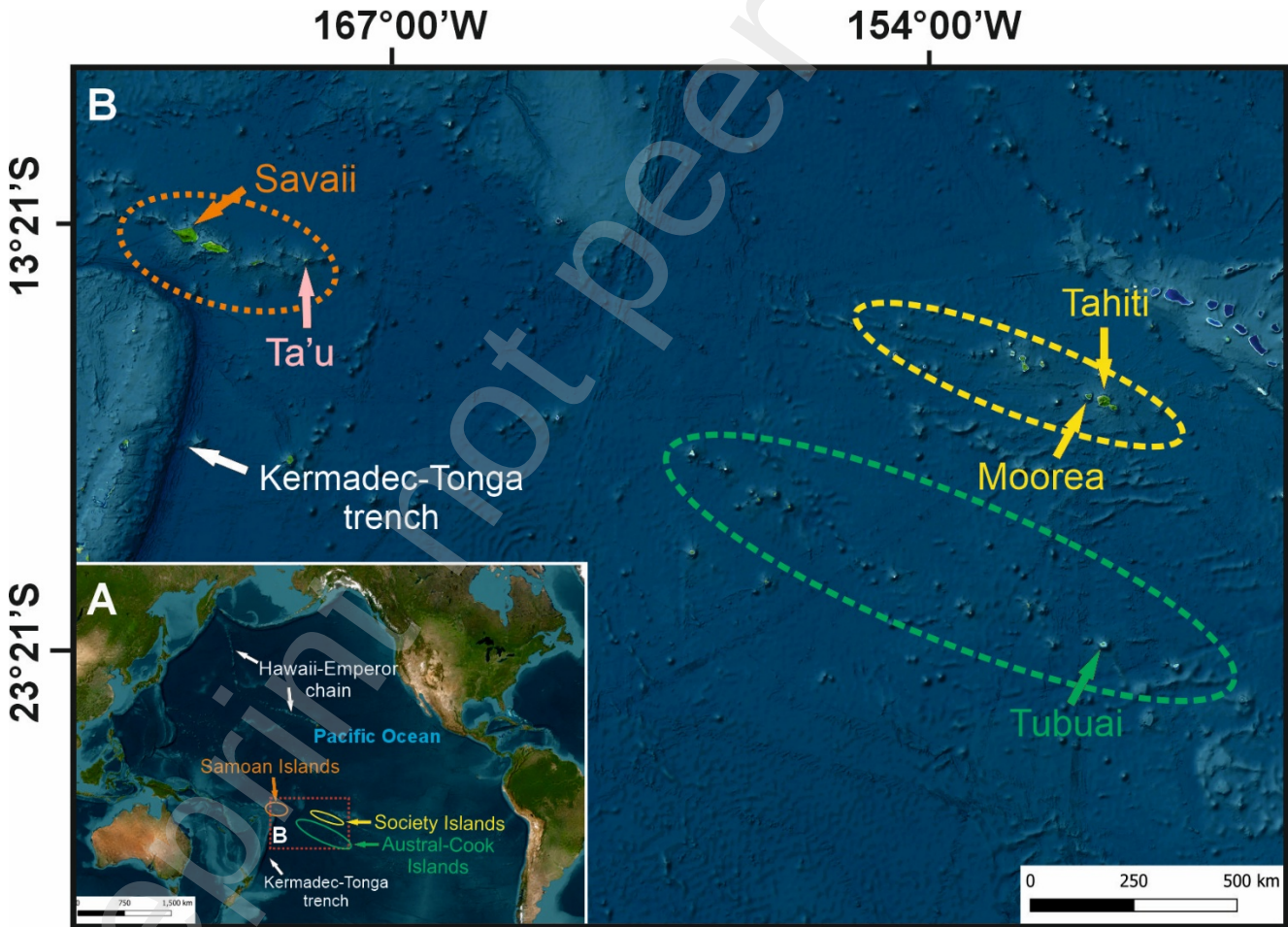


Figure 1. Map showing the Polynesian localities studied in this work. Figure B is a closer view of the Samoa, Society and Austral-Cook islands.

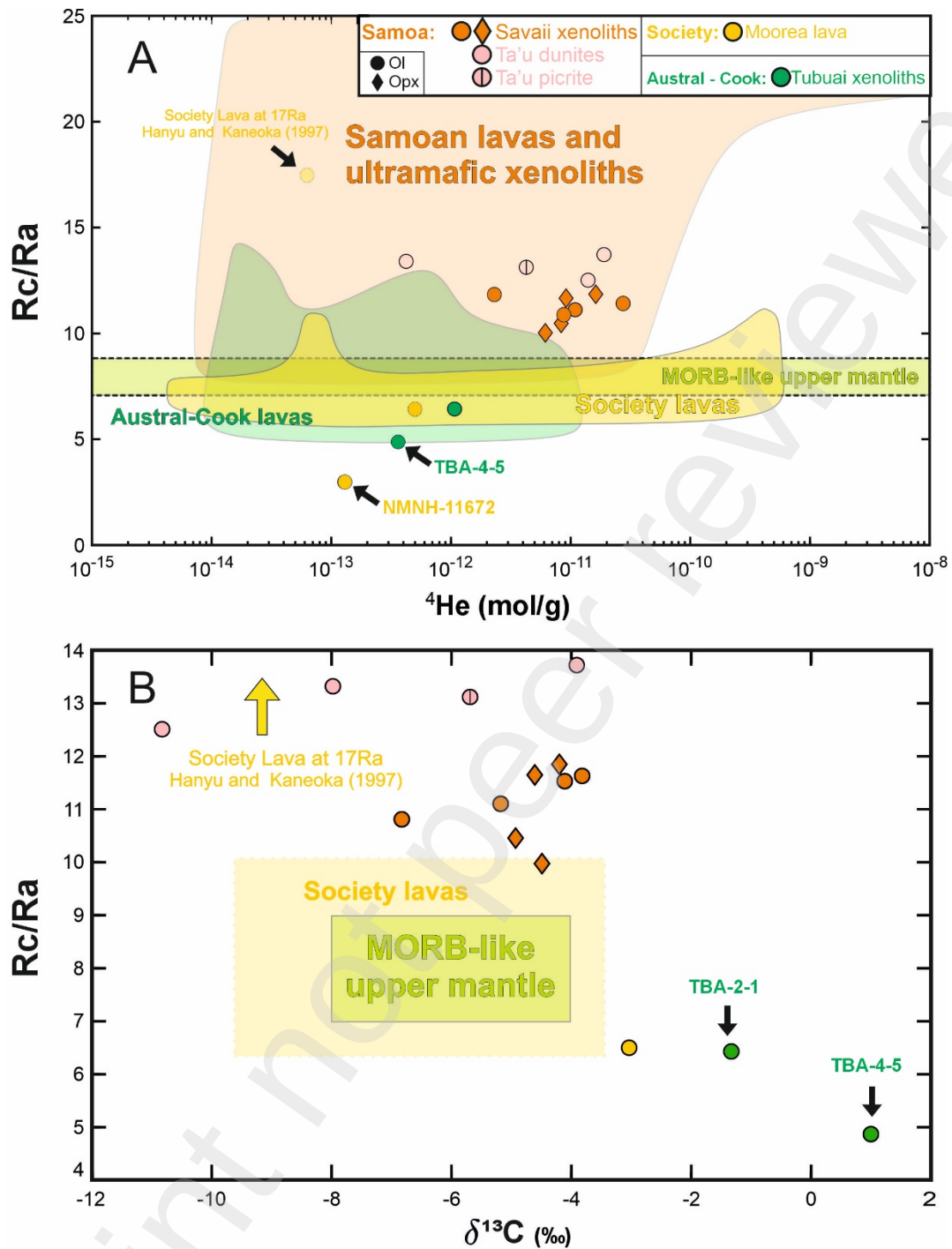


Figure 3. A) ^4He vs R_c/R_a ($^3\text{He}/^4\text{He}$ corrected for atmospheric contamination). MORB range is reported at $R_c/R_a = 8 \pm 1$ (Graham, 2002). Samoa, Society and Austral.Cook fields were designed using helium data from the literature (Farley et al., 1992; Poreda and Farley, 1992; Hanyu and Kaneoka, 1997; Hanyu et al., 1999, 2011; Marty and Dauphas, 2003; Jackson et al., 2007, 2009, 2016; Lupton et al., 2009; Parai et al., 2009; Zhang et al., 2024; Harðardóttir and Jackson, 2025). B) $\delta^{13}\text{C}$ vs R_c/R_a ($^3\text{He}/^4\text{He}$ corrected for atmospheric contamination). MORB range is reported at $R_c/R_a = 8 \pm 1$ (Graham, 2002) and $\delta^{13}\text{C}$ at -8 to -4‰ (Sano and Marty, 1995). Society lavas field was designed after Hanyu and Kaneoka (1997), Marty and Dauphas (2003) and Aubaud et al. (2005).

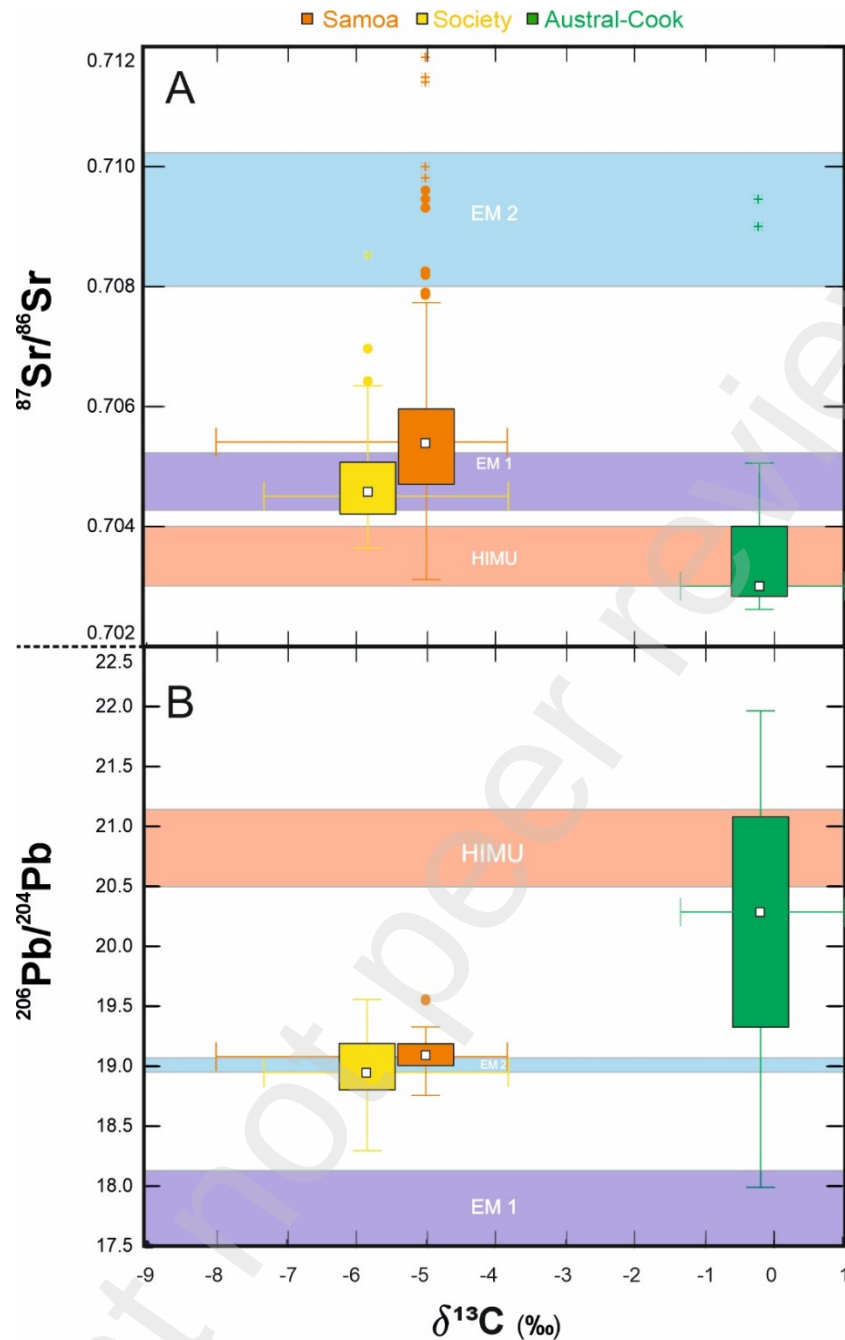


Figure 4. Relationships between $\delta^{13}\text{C}$ values of FIs and radiogenic isotope compositions for mantle-derived samples from Samoa, Society, and Austral-Cook islands. (A) $\delta^{13}\text{C}$ versus (our samples) $^{87}\text{Sr}/^{86}\text{Sr}$ and (B) $\delta^{13}\text{C}$ (our samples) versus $^{206}\text{Pb}/^{204}\text{Pb}$. Colored fields indicate the approximate compositional ranges of the HIMU, EM1, and EM2 mantle $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ end-members from the literature (Workman et al., 2004; Jackson and Dasgupta, 2008). Boxplots illustrate the distribution of the radiogenic isotope datasets, with white squares representing mean values, boxes corresponding to the interquartile range (IQR), and boxes corresponding to the interquartile range (IQR), and vertical whiskers representing the full isotopic range excluding outliers ($^{87}\text{Sr}/^{86}\text{Sr}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ data was obtained from Harðardóttir and Jackson, 2025). The vertical extent of the boxplots therefore reflects the variability of the Sr and Pb isotopic compositions. In contrast, the horizontal width of the boxes is purely schematic and does not represent statistical dispersion in $\delta^{13}\text{C}$ values. The actual range in $\delta^{13}\text{C}$ is represented by the horizontal error bars and corresponds to the range observed in this study together with published literature data (particularly for the Society Islands dataset).

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890 **TABLES**

891 **Table 1.** Carbon and noble gas isotopic composition of Samoa, Society and Tubuai ultramafic xenoliths and lavas. Errors are reported as 1σ
 892 uncertainties.

Sample ID	Locality	Island	Rock	Phase	CO ₂ (mol/g)	CO ₂ /³He	δ¹³C	⁴He (mol/g)	Rc/Ra	error (+/-)	⁴He/⁴⁰Ar*	error (+/-)
SAV 3-11	Samoa	Savai'i	Harzburgite	Ol	5.52E-07	1.47E+10	-3.82	2.32E-12*	11.63*	0.08	-	-
SAV 3-11	Samoa	Savai'i	Harzburgite	Opx	1.00E-06	6.74E+09	-4.61	9.16E-12	11.66	0.1	0.14	0.11
SAV 1-42	Samoa	Savai'i	Harzburgite	Ol	2.69E-06	6.16E+09	-4.11	2.72E-11*	11.53*	0.17	-	-
SAV 1-42	Samoa	Savai'i	Harzburgite	Opx	5.58E-06	6.55E+10	-4.49	6.14E-12	10.03	0.09	0.06	0.1
SAV-1-13	Samoa	Savai'i	Harzburgite	Ol	6.37E-07	5.00E+09	-6.83	8.48E-12*	10.81*	0.16	-	-
SAV 1-13	Samoa	Savai'i	Harzburgite	Opx	3.27E-06	2.70E+10	-4.93	8.34E-12	10.47	0.1	0.13	0.14
SAV 1-28	Samoa	Savai'i	Sp-Harz	Ol	1.97E-07	1.20E+09	-5.18	1.09E-11	11.12	0.1	0.28	2.52
SAV 1-28	Samoa	Savai'i	Sp-Harz	Opx	2.26E-06	8.67E+09	-4.2	1.62E-11	11.85	0.13	0.34	4.09
T25	Samoa	Ta'u	Picritic lava	Ol	3.03E-07	3.90E+09	-5.69	4.26E-12	13.12	0.12	0.8	0.16
Tau-04-28	Samoa	Ta'u	Dunite	Ol	8.15E-07	2.31E+09	-3.91	1.90E-11	13.72	0.13	1.44	0.29
T-17-02	Samoa	Ta'u	Dunite	Ol	1.30E-07	5.47E+08	-10.83	1.40E-11	12.51	0.12	1.55	0.36
T-17-04	Samoa	Ta'u	Dunite	Ol	2.29E-07	2.93E+10	-7.98	4.22E-13	13.4	0.14	0.05	0.25
TBA 4-5	Austral-Cook	Tubuai	Harzburgite	Ol	1.33E-06	5.42E+11	1	3.62E-13*	4.87*	0.09	-	-
TBA 2-1	Austral-Cook	Tubuai	Harzburgite	Ol	4.30E-07	4.49E+10	-1.33	1.07E-12*	6.43*	0.1	-	-
NMNH-11672	Society	Tahiti	Basanite	Ol	-	-	-	1.57E-13	3.26	0.11	0.26	0.27
NMNH-101165	Society	Moorea	Olivine Basalt	Ol	3.32E-07	7.74E+10	-3.12	4.86E-13	6.35	0.09	-	-

*Rc/Ra and ⁴He values are from Workman et al. (2004) and Jackson et al. (2016)

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