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1 The influence of igneous processes on the chromium
2 isotopic compositions of Ocean Island basalts

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26 Abstract:

27 We present the first stable chromium isotopic data for a suite of ocean island basalts (OIB) in order to
28 investigate the Cr isotope fractionation during major igneous processes such as partial melting and
29 fractional crystallisation. Twenty-one basaltic samples from Fangataufa Island (Tuamotu Archipelago,
30 Pacific Ocean) have been analysed for major- and trace-element concentrations, and Sr, Nd and Cr
31 isotopic compositions. They define two distinct series: medium to high-K calc-alkaline and low to
32 medium-K calc-alkaline. The variations in incompatible elements such as La and Yb mostly result from
33 varying degrees of partial melting of a mixed mantle source composed of two lithologies: garnet bearing
34 peridotite and a “fertile” component. The recycled component is also identified with the Sr and Nd
35 isotopic composition of Fangataufa basalts. In contrast, the variations in compatible element contents
36 such as Cr and Ni are governed by fractional crystallisation of a mixture of olivine, clinopyroxene and
37 spinel. The samples analysed in this study are also characterised by small Cr isotope variations from -
38 0.24 to -0.17 ‰. The Cr-poor samples have on average lighter Cr isotopic compositions compared to
39 Cr-rich ones. The observed variations in the low-K series can be modelled by a Rayleigh fractionation
40 model with a fractionation factor ($\Delta^{53}\text{Cr}_{\text{crystals-melt}}$) of -0.010 ± 0.005 ‰. The fractionation is more limited
41 than that observed in lunar basalts and two hypotheses may explain this observation: a change in
42 crystallising phases (cpx + spinel on Earth and spinel on the Moon) and/or the difference in temperature
43 and oxygen fugacity between the crystallisation of lunar and terrestrial basalts. The more primitive
44 basalts from Fangataufa have an average Cr isotopic composition of -0.18 ± 0.01 ‰, lighter than the Cr
45 isotopic composition of pristine mantle xenoliths. Chromium isotopes are therefore slightly fractionated
46 during partial melting with the melts depleted in heavy Cr isotopes. The difference between silicate
47 melts and mantle xenoliths indicates that partial melting could produce small but resolvable shift in
48 mantle xenoliths Cr isotopic composition of up to 0.05 ‰. The covariation between $\delta^{53}\text{Cr}$ and $\epsilon^{143}\text{Nd}$
49 values in Fangataufa basalts can be explained by two processes: (i) lower degree of partial melting
50 produce lighter Cr isotopic composition in basaltic liquids relative to their sources and (ii) melts
51 produced by low degree of partial melting have also experienced more fractional crystallisation.

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61 Keywords: Cr isotopes, Ocean Island Basalts, partial melting, magmatic differentiation, Fangataufa
62 Archipelago.

63 1. Introduction

64

65 The knowledge of the isotopic composition of the Earth and its main reservoirs is fundamental
66 to understand the evolution of our planet through time. For this purpose, radiogenic isotopes have been
67 extensively studied over the last decades. More recently, non-conventional stable isotopes have been
68 also used to constrain the chemical evolution of planetary reservoirs (e.g. Dauphas et al. 2012, Sossi et
69 al. 2018). High-temperature, mass-dependent stable isotope variations have indeed been discovered,
70 opening a new way to interrogate the geological record and to constrain the chemical reactions during
71 differentiation (e.g. Teng et al. 2008). However, contrary to radiogenic isotopes, mass dependent stable
72 isotopes can be fractionated by igneous processes (Xia et al. 2017, Tang et al. 2017). It is therefore
73 essential to constrain the behaviour of stable isotopes during partial melting and fractional
74 crystallisation of basaltic system in order to use them as efficient tracers of the Cr isotopic composition
75 of planetary reservoirs.

76 Chromium has a half-nebular condensation temperature of 1296 K which makes it a moderately
77 volatile element (Lodders 2003) although many studies have classified chromium as “medium
78 refractory” (Allègre et al. 2001). Chromium is a moderately siderophile element with metal-silicate
79 partition coefficient ranging from 2.5 to 3.5 when determined at core formation conditions (Wood et al.
80 2008, Siebert et al. 2011, Clési et al. 2016). The siderophile behaviour of chromium depends on
81 temperature and oxygen fugacity (e.g. Siebert et al. 2011). Finally, chromium behaves compatibly
82 during partial melting of the upper mantle and fractional crystallisation of basaltic systems. The Cr
83 compatibility during partial melting of the mantle strongly depends on temperature and Cr becomes less
84 compatible with increasing temperature (Liang and Elthon 1990). The main Cr-bearing phases in the
85 upper mantle are spinel (or garnet at high pressure) and pyroxenes (Murck and Campbell 1986; Li et al.
86 1995). In natural high temperature systems, chromium can be present in multiple oxidation states: Cr⁰
87 in metal melt, Cr²⁺ and Cr³⁺ in silicate melts and minerals, and Cr³⁺ in oxide minerals (Berry et al. 2004,
88 Berry et al. 2006, Li et al. 1995). At 1400 °C, in MORB glass composition, 50% of the Cr is trivalent
89 at the QFM buffer (Berry et al. 2006). The redox behaviour of chromium makes it a key element to
90 study variations in oxygen fugacity and redox reactions in natural systems. The estimate for the bulk

91 silicate Earth (BSE) and the core Cr concentrations vary from 2500 to 2800 $\mu\text{g g}^{-1}$ (McDonough and
92 Sun 1995, Palme and O'Neill 2014) and from 5500 to 9000 $\mu\text{g g}^{-1}$ (Allègre et al. 1995, Javoy et al. 2010,
93 McDonough 2013), respectively. Typical variations of Cr concentrations in mantle xenoliths have been
94 linked to partial melting and for terrestrial basalts to fractional crystallisation processes (e.g. Li et al.
95 1995). These variations and their causes need to be precisely addressed before determining the initial
96 Cr concentration in the primary liquids of terrestrial basalts and in the primitive mantle material.

97 Chromium isotopes have been studied recently in a number of terrestrial and extra-terrestrial
98 samples (e.g. Schoenberg et al. 2008, Farkas et al. 2016, Wang et al. 2016, Bonnand et al. 2016a,
99 Bonnand et al. 2016b). The variations in Cr isotopes are reported using the notation:

$$100 \quad \delta^{53}\text{Cr} = \left(\frac{{}^{53}\text{Cr}/{}^{52}\text{Cr}_{\text{sample}}}{{}^{53}\text{Cr}/{}^{52}\text{Cr}_{\text{NIST 979}}} - 1 \right) \times 1000$$

101 Most recent analyses on mantle xenoliths and mineral separates have shown that Cr isotopes are
102 fractionated during partial melting (Xia et al. 2017). The residues become heavier while partial melting
103 proceeds and the melts are therefore believed to be lighter than the residues although no melts have yet
104 been analysed at high precision. Furthermore, mineral separates data reveal that at equilibrium, the Cr
105 isotopic composition become heavier in the order olivine \leq cpx = opx < spinel (Shen et al. 2018). It has
106 also been demonstrated that Cr are fractionated during magmatic differentiation on the Moon (Bonnand
107 et al. 2016b). Comparing Cr isotope signatures in samples from different planetary bodies is challenging
108 because planetary processes may have affected the composition of the major reservoirs. The difference
109 in isotopic composition between the Moon and the Earth has been attributed to the volatility of Cr during
110 cooling and accretion on the Moon in the aftermath of the giant impact (Sossi et al. 2018). For HEDs
111 meteorites (Howardite-Eucrite-Diogenite), both volatility and magmatic processes could play a role in
112 their Cr isotopic compositions (Zhu et al. 2019, Bonnand et al. 2016b). Iron meteorites are characterised
113 by heavy Cr isotopic compositions compared to terrestrial samples, which have been explained by
114 fractional crystallisation in the liquid iron (Bonnand and Halliday 2018). The similarity between the Cr
115 isotopic compositions of the bulk silicate Earth estimates and chondritic material suggests that there is
116 no fractionation during core formation on Earth (Bonnand et al. 2016a, Schoenberg et al. 2016).

117 Although there is strong indication that Cr isotopes are fractionated in terrestrial samples during
118 partial melting and fractional crystallisation, direct measurements are still lacking. Two studies have
119 presented Cr isotopic composition in terrestrial basaltic rocks (Schoenberg et al. 2008; Xia et al. 2017).
120 Resolvable variations were observed but the limited number of samples did not allow to determine the
121 behaviour of Cr isotopes during partial melting and/or fractional crystallisation. Here, we investigate
122 the Cr isotopic composition of basalts from one oceanic island: Fangataufa, Pacific Ocean. In order to
123 determine the initial Cr isotopic composition of the parental melt we investigate the isotopic
124 fractionation resulting from fractional crystallisation and assess whether these fractionations are
125 comparable to those observed in other planetary bodies. Once the primary melt has been defined, we
126 also investigate the fractionation happening during partial melting.

127

128 2. Geological setting

129

130 Fangataufa Island is a small atoll located in the Tuamotu Archipelago, South Pacific Ocean.
131 The studied samples come from a 958m drill core called “Terme Sud” which was realised in the
132 seventies by the Commissariat à l’Energie Atomique (CEA). The stratigraphic sequence is composed
133 of both sub-aerial and submarine volcanic rocks topped by carbonate sediments (Schiano et al. 1990).
134 Our samples have been selected to span the entire sequence recorded there (sample depth from -929m
135 to -458m, see supplementary information file). In detail, the samples have been classified into three
136 main lithologies: submarine lava flows, dykes and sills, and monolithological hyaloclastites. Previous
137 studies have shown limited variations in the major element concentrations of volcanic rocks: the
138 samples are mainly basalts or trachy-basalts and a continuous evolution between alkaline series and a
139 transitional series was observed at “Terme Sud” drill core (Fig. S1, Schiano et al. 1990, Dupuy et al.
140 1993, Schiano et al. 1993). The Fangataufa basalts are characterised by typical OIB REE patterns with
141 LREE enrichment relative to HREE ($Ce/Yb_N = 8.8$, Dupuy et al. 1993). They also have incompatible
142 trace element ratios (Th/La, Ba/La, Hf/Sm) in the same range as other OIBs (Dupuy et al. 1993). Partial
143 melting is the dominant igneous process that controls the variations in incompatible trace element
144 contents (Dupuy et al. 1993). By fractional crystallization, Fangataufa submarine lavas evolve also from

145 picrites to basalts whereas subaerial lavas, which correspond to the end of the volcanic activity, evolve
146 from picrites to hawaiites. Some of the samples have also experienced secondary processes that altered
147 their original signatures (Schiano et al. 1990). The main process is seawater-rock interaction (some of
148 the samples have been emplaced under-water), although alteration during late hydrothermal fluid
149 circulation was also reported. Lastly, volcanic products from Fangataufa have been studied for their
150 $^{87}\text{Sr}/^{86}\text{Sr}$ (0.7028-0.703), $^{143}\text{Nd}/^{144}\text{Nd}$ (0.512949-0.512951) and $^{206}\text{Pb}/^{204}\text{Pb}$ (19.292-19.396) isotopic
151 compositions (Dupuy et al. 1993). The results have led to the conclusion that their source is uniform
152 isotopically (Dupuy et al. 1993, Bardintzeff et al. 1994). The relatively radiogenic Pb isotopes
153 associated with unradiogenic Sr and radiogenic Nd suggest that Fangataufa basalts may have been
154 influenced by a HIMU-like component.

155

156 3. Analytical methods

157 3.1. Major elements

158 Major-element concentrations were determined at the Laboratoire Magmas et Volcans (LMV)
159 using Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES, Jobin-Yvon ULTIMA
160 C), after alkaline melting with lithium metaborate/tetraborate and nitric acid dissolution. The BHVO-1
161 reference material was used as an internal standard to determine the reproducibility of the analyses. It
162 is better than 5% except for Na_2O and K_2O (Table SI). Loss on ignition (LOI) was also measured for
163 our samples. Up to 1 g of sample powder was precisely weighed and subsequently placed in an oven at
164 110°C and 1100°C for 24h and 48h respectively. After cooling down, the sample powders were weighed
165 again and the difference between the masses is the Loss on Ignition.

166

167 3.2. Trace elements

168 Up to 300 mg of whole-rock powders were digested using concentrated HNO_3 -HF-HCl acid
169 mixtures. The fully dissolved samples were evaporated to dryness and re-dissolved in 6M HCl. Once
170 they were perfectly clear, the samples were split in three aliquots: one for trace element, one for Sr and
171 Nd isotopes and finally one for Cr isotope measurements. For trace-element analyses, an aliquot of ~1
172 mg of sample was taken and dried down on the hotplate. The sample was then re-dissolved in 0.05M

173 HNO₃ and diluted 2000 times for the inductively coupled plasma mass spectrometer (ICP-MS)
174 measurements. Trace element concentrations were determined using the Agilent 7500 at the LMV. The
175 analyses were standardised against five reference materials (BIR-1, BHVO-2, AGV-1, DNC-1 and W-
176 2). Within-run drift was assessed and corrected internally using an In standard. Precision was assessed
177 with repeated measurements of BEN reference material and is routinely better than 4 % (2 s.d.).
178

178

179 3.3. Sr and Nd isotopes

180 An aliquot of the sample solutions, with approximately 1 µg of Nd, was dried down and re-
181 dissolved in 2ml 1.25M HCl. The samples were passed through the column protocol (Sr Spec, True
182 Spec and Ln Spec columns) after most of the iron had been removed through a cation resin (AG50 X4).
183 Sr and Nd procedural blanks were negligible with respect to the amount of each element processed
184 through chemistry. All measurements were made on the LMV ThermoFisher Triton Thermo-Ionization
185 Mass Spectrometer (TIMS) with single (Sr) and double (Nd) Re filaments. A typical Sr isotope run
186 consisted of 15 blocks of five cycles (8 s integration time) to allow a full rotation of the virtual amplifier
187 system. Strontium isotope ratios were mass-fractionation corrected with $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ and
188 normalized to $^{87}\text{Sr}/^{86}\text{Sr} = 0.710245$ for the NIST-SRM987 standard. Neodymium isotope measurements
189 consisted of 14 blocks of 10 cycles. Neodymium isotopes were mass-fractionation corrected with
190 $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$ and normalized to $^{143}\text{Nd}/^{144}\text{Nd} = 0.512100$ for the JNDi-1 standard. Repeated
191 analyses of the two standards during the course of the study gave $^{87}\text{Sr}/^{86}\text{Sr} = 0.710241 \pm 9$ (2 s.d., n =
192 13) and $^{143}\text{Nd}/^{144}\text{Nd} = 0.512106 \pm 4$ (2 s.d., n = 8), in agreement with the long-term reproducibility at
193 LMV i.e., $^{87}\text{Sr}/^{86}\text{Sr} = 0.710244 \pm 11$ (2σ, n = 96) and $^{143}\text{Nd}/^{144}\text{Nd} = 0.512104 \pm 7$ (2σ, n = 47). The
194 reference material BHVO-2 was also analysed in the same analytical session and gave $^{87}\text{Sr}/^{86}\text{Sr} =$
195 0.703469 ± 5 (2 s.e.) and $^{143}\text{Nd}/^{144}\text{Nd} = 0.512977 \pm 3$ (2 s.e.). These values agree with literature values
196 (Weis et al. 2006).
197

197

198 3.4. Cr isotopes

199 Protocols for the separation of Cr from silicate matrices have been previously published (e.g.
200 Trinquier et al. 2008; Bonnand et al. 2016a). Only a brief summary is given here. Aliquots of ~2 µg of

201 chromium of the solutions prepared for trace element and Sr-Nd isotopes were spiked with the requisite
202 amount of ^{50}Cr - ^{54}Cr double spike (Bonnand et al. 2011). The solutions were dried down and taken up
203 in 6M HCl to ensure isotopic equilibrium between the samples and the spike. The protocol used to
204 separate the Cr fraction from the matrix is a two-column procedure and has been described in details in
205 Bonnand et al. (2016a). Briefly, the first column procedure is designed to remove the major cations
206 while the second column procedure removes the small remaining isobaric interference elements such
207 as Ti. The isotopic measurements were performed on a ThermoFisher Triton TIMS at the Laboratoire
208 Magmas et Volcans. Typical Cr isotope measurements consisted of 54 blocks of 10 cycles. The gains
209 were measured daily and the baselines (30s) were measured before each block. Amplifier rotation was
210 used to cancel out the gain differences between each faraday cups. The external reproducibility obtained
211 for the JP-1 reference material over the course of this study is $\delta^{53}\text{Cr} = -0.108 \pm 0.014 \text{ ‰}$ (2 s.d., n = 8)
212 comparable to previous estimate of this geological reference material (Bonnand et al. 2016a, Li et al.
213 2016, Zhu et al. 2018).

214

215 4. Results

216 The studied samples present relatively limited variations in their major element
217 concentrations. The Fangataufa basalts have high LOI values ranging from 1.4 to 9.9 % (78% of the
218 samples have LOI values lower than 4%, Table SI), in agreement with previous reported values on
219 samples coming from the same location (Schiano et al. 1990). There is a weak relationship between loss
220 on ignition and Ca concentration suggesting that interaction with carbonate rocks and/or seawater-
221 derived fluids are responsible for the secondary alteration observed in the samples. In detail, this
222 covariation is strongly dependent on three samples and the absence of correlation between LOI and Sr
223 isotopes suggest that a simple mixing with carbonate rocks is not the only process at the origin of the
224 high LOI values. Other major element concentrations do not co-vary with LOI and are therefore related
225 to igneous processes. Furthermore, most samples have LOI values between 2 and 4 %, which is coherent
226 with submarine emplacement.

227 Selected major element concentrations, expressed as dry sample, are presented in Table 1 and
228 Figures 1, S1 and S2. The SiO_2 and MgO concentrations range from 43.8 to 50.3 wt. % and from 3.7 to

229 9.0 wt. %, respectively. The TiO₂ concentrations range from 1.8 to 4.3 wt. %. The K₂O concentrations
230 are more variable and range from 0.1 to 1.8 wt. %. Finally, CaO concentrations range from 8.2 wt. %
231 to 21.1 wt. %. Our measurements are in good agreement with previously published results on
232 Fangataufa basalts (Dupuy et al. 1990). Reported in typical TAS diagram (Fig. S1), the studied samples
233 fall in the basalt, picro-basalt, basanite and trachy-basalt fields. Major element compositions show that
234 they have experienced limited magmatic differentiation as previously proposed for Fangataufa. The
235 variations in K₂O and TiO₂ indicate that, in agreement with earlier studies, two magmatic suites have
236 been sampled: one medium to high-K calc-alkaline (K₂O > 0.75 wt. %, TiO₂ > 3 wt. %) and one low to
237 medium-K calc-alkaline (K₂O < 0.75 wt. % and TiO₂ < 2.5 Wt. %), hereafter called high-K and low-K
238 suites, respectively. The high-K (red) and low-K (blue) series are shown in Figure 1 and 2. They can
239 be clearly distinguished by their K₂O content even if a continuum exists.

240 The minor- and trace-element concentrations measured in Fangataufa basalts are presented in
241 Table S1. Chromium and Th concentrations range from 3.4 to 677.7 μg g⁻¹ and from 0.9 to 4.6 μg g⁻¹,
242 respectively (Table 1). The Ni concentrations range from 4.8 to 292.5 μg g⁻¹. The La/Yb ratio
243 determined for the studied samples varies from 10.0 to 22.8 (Figure 2). While the REE concentrations
244 are positively correlated with Th concentration, Cr and Ni concentrations appear to be negatively
245 correlated to Th content. Our results agree well with previous estimates of the trace element contents of
246 Fangataufa basalts (Schiano et al. 1993).

247 The Sr and Nd isotope data are presented in Table 1 and Figure 3. Fangataufa basalts have Sr
248 and Nd isotopic compositions ranging from 0.702865 to 0.703491 and from 0.512883 to 0.512956,
249 respectively. The results presented in this study are in good agreement with previous studies (Dupuy et
250 al. 1993, Bardintzeff et al. 1994). There is no correlation between Sr and Nd isotopes, and between Sr
251 or Nd isotopes and LOI. In the case of secondary alteration, Sr isotopes are more readily affected than
252 Nd isotopes due to the relatively depleted Nd concentration of authigenic sediments. The discussion
253 will be mainly focus on the Nd isotopic composition in the following sections.

254 The Cr isotopic compositions (δ⁵³Cr) of the studied samples are given in Table 1 and range
255 from -0.226 to -0.169 ‰. There is no correlation between Cr isotopes and LOI and other indices of

256 alteration. There is a weak relationship between Cr isotopes and Cr and MgO concentrations and Nd
257 isotopes (Fig. 4 and 5).

258

259 5. Discussion

260 5.1. Magmatic processes and the composition of the mantle source

261 Primary melts produced at the degrees of melting and pressure inferred for OIBs at equilibrium
262 with peridotite have around 12-14% MgO (e.g. Laporte et al. 2004). Typical magmas erupting on ocean
263 islands have MgO concentrations around 7-8%, representing evolved magmas with substantial olivine
264 crystallisation. Most of Fangataufa rocks have 5-6% MgO in agreement with such an explanation. A
265 few samples, however, have higher MgO concentrations (up to 9 wt. %). These concentrations may
266 reflect either less evolved magmas or olivine accumulation during the magma ascent to the surface,
267 although, in other Ocean Island settings, accumulation of olivine increases the MgO concentration in
268 magmas up to ~25 wt. % (e.g. La Reunion Island, 26.3%, Albarède et al. 1997) much higher than the
269 observed variations in Fangataufa basalts. Thus, the relatively small increase in MgO concentration
270 suggests that accumulation of olivine antecrysts and/or xenocrysts is limited in the studied samples. In
271 modelling the variations of incompatible elements, olivine accumulation is a simple linear addition,
272 whereas fractional crystallisation follows an exponential law. Thus, on a log-log plot of an incompatible
273 element in olivine (e.g. Y) versus a compatible one (e.g. MgO), a global covariation with an inflection,
274 corresponding to the composition of the parental liquid, is expected in case of accumulation (e.g. Nebel
275 et al. 2014). In Fangataufa basalts, there is no break in the array (Fig. S6) which suggests that there is
276 indeed limited amount of olivine accumulation.

277 The CaO concentration can help us constraining the crystallisation history experienced by the
278 studied samples. The samples with LOI higher than 4.5% (4 samples) were not considered. In a typical
279 CaO/Al₂O₃ vs FeO/MgO plot (Fig. S2), the remaining samples display a negative correlation. This
280 indicates that during magmatic differentiation the fractionating mineralogy assemblage was mainly
281 composed of olivine and clinopyroxene. This observation is similar to that observed for another Ocean
282 Island basalts (La Réunion, Albarède et al. 1997).

283 It has been proposed that the source of Fangataufa basalts is relatively homogeneous
284 isotopically compared to other OIBs (Bardintzeff et al. 1994). In contrast, results presented in this study
285 show that although the variations are limited, the studied samples clearly have heterogeneous Nd
286 isotope ratios. This heterogeneity is not correlated with sample depths and therefore it cannot be linked
287 to the transition from low-K to high-K series. The weak correlation between $^{143}\text{Nd}/^{144}\text{Nd}$ isotopic ratios
288 and La/Yb rather suggests that variation of the Nd composition results from the variation of the degree
289 of partial melting of a heterogeneous source.

290 Fangataufa basalts plot close to the HIMU end-member region in the $^{143}\text{Nd}/^{144}\text{Nd}$ vs $^{87}\text{Sr}/^{86}\text{Sr}$
291 isotope variation diagram (Figure 3). The HIMU end-member is classically associated with the
292 recycling of old and altered oceanic crust (i.e., readily-fusible component referred as fertile component)
293 into mantle sources (e.g. Chauvel et al. 1992). The Sr and Nd isotope ratios measured in the studied
294 samples together with previous Pb isotope data (Bardintzeff et al. 1994) suggest the presence of a
295 recycled component in the source of Fangataufa. Along with isotopic evidence for a recycled component
296 in OIBs sources is the Ti concentrations. An elevated Ti concentration in basalts usually reflects the
297 presence of an enriched component in the source (Prytulak and Elliot 2007). It is indeed difficult to
298 produce high TiO_2 concentration in silicate melt when melting typical peridotite composition (garnet or
299 spinel). The Fangataufa basalts have TiO_2 concentrations ranging from 1.8 to 4.3 wt. % with an average
300 of 3.17 wt. % in line with other OIBs (Prytulak and Elliot 2007). The high Ti concentration in the basalts
301 analysed in this study, together with their isotopic compositions, indicates that the source of these
302 basalts has been enriched by a “fertile” component. The presence of a “fertile” component in the source
303 of Fangataufa basalts has a strong implication for the behaviour of incompatible trace elements during
304 partial melting.

305

306 5.2. The influence of partial melting on the trace element budget

307 The studied samples show large variations of their trace element concentrations. Fangataufa
308 basalts have REE patterns enriched in Light REE (LREE) and depleted in Heavy REE (HREE), like
309 typical OIBs (Fig. S3). The La/Yb ratio varies from 7.2 to 16.4 in our less altered samples compared to
310 1.5 in the primitive mantle (Sun and McDonough 1995). Variations in REE concentrations can be

311 explained by two igneous processes: partial melting and fractional crystallisation. The positive
312 correlation defined by our samples between REE (and also La/Yb) and Th (a strongly incompatible
313 element) indicates that the observed variations are mainly due to partial melting. It is also confirmed by
314 the positive correlation between Th/Nd ratio and Th concentration for the studied samples.

315 A model for the partial melting of a mantle source is presented in Figure 6. In order to
316 fractionate the La/Yb ratio and also to explain the Nd isotopic composition of basalts (section 5.1), we
317 considered a uniform mixed source composed of 2% “fertile” component and 98% of a peridotite in the
318 garnet stability field (a spinel peridotite would not explain the high La/Yb values). The partition
319 coefficients and the initial concentrations used in the model are presented in Table 2. Hirschmann and
320 Stolper (1996) reported a difference in productivity between “fertile” component and peridotite during
321 partial melting. Based on their experimental results, we assigned to the “fertile” component a
322 productivity four times higher than that of the peridotite. The mineralogy of the peridotite was set to
323 57% olivine (ol), 13% clinopyroxene (cpx), 28% orthopyroxene (opx) and 5% garnet (gt), similarly to
324 the mineralogy used by Putirka et al. (2011). The modal compositions of pyroxenites are extremely
325 variable (e.g. Lambart et al. 2016). In this study, we considered an “eclogite type” component with a
326 mineralogy of 40% cpx, 60% gt (e.g. Sossi and O’Neill 2017). As melting of the mantle is non-modal,
327 the effect of melting stoichiometry on the partition coefficient has to be considered (e.g. Shaw 1970,
328 Sossi and O’Neill 2017, Shen et al. 2018). The melting reactions used in our model are $0.05*Ol +$
329 $0.96*Cpx + 0.14*Gt = 0.15*Opx + 1*Liq$ for the garnet peridotite and $0.8*Cpx + 0.2*Gt = 1*Liq$ for
330 the “fertile” component (Walters 2003, Sossi and O’Neill 2017). At 1% partial melting, the Th
331 concentration in the melt is $8.5 \mu\text{g g}^{-1}$ and the La/Yb ratio is 44. At 10% partial melting, the Th
332 concentration is $0.86 \mu\text{g g}^{-1}$ and the La/Yb ratio is 5.6. The model reproduces the Th and La/Yb
333 variations observed in Fangataufa basalts with a degree of partial melting varying from 2% to 10%. We
334 therefore propose that the high-K series is produced by 2-5% partial melting of such a mixed mantle
335 source, whereas the low-K series was produced by up to 10% partial melting. Our model allows us to
336 reproduce the variations in trace element concentrations and also the small variations in Nd isotopes if
337 we modify the difference of productivity between the “fertile” and peridotite components with partial
338 melting. As melting proceeds, the “fertile” component is melting first but becomes progressively diluted

339 by melts derived from the garnet peridotites (with higher $\epsilon^{143}\text{Nd}$). Importantly, the change in fertility
340 would not significantly change the main observation obtained with our conceptual non-modal melting
341 model of a mixed source presented in Figure 7. Fangataufa basalts are produced by partial melting of a
342 mixed source (garnet peridotite and a fertile component) with the high-K series produced with lower
343 degree of partial melting compared to the low-K series.

344

345 5.3. Variations in Cr concentration: partial melting vs fractional crystallisation

346 Contrary to most trace elements, the variations in Cr and Ni contents do not positively correlate
347 with Th and/or La/Yb ratio (Fig. 7 and S4). As it is typically admitted that Th is an ultra-incompatible
348 element with a $D^{\text{Th}}_{\text{crystal-melt}}$ close to 0 (e.g. McKenzie and O’Nions 1995), this indicates that Cr
349 behaviour is not controlled by partial melting but is strongly affected by another igneous process.
350 During fractional crystallisation of phases such as chromite ($D^{\text{Cr}}_{\text{chromite-melt}} = 170$, Liu and O’Neill 2004),
351 Cr behaves as a compatible element and so its concentration decreases rapidly in the residual silicate
352 melt. It is therefore possible to explain the variations measured in Fangataufa basalts by fractional
353 crystallisation of Cr-bearing phases early in the crystallisation sequence. Turning to nickel, it also
354 behaves compatibly during igneous processes and is compatible into olivine (e.g. $D^{\text{Ni}}_{\text{olivine-melt}} = 9.4$,
355 McKenzie and O’Nions 1995), one of the early phases to crystallise from a basaltic melt. The Ni
356 behaviour in basaltic system is strongly dependent on melt composition and temperature (e.g. Matzen
357 et al. 2013). The positive correlation between Cr and Ni (slope of 2.7, Fig. S5) shows that both elements
358 are affected during the crystallisation of the parental melts.

359 The behaviour of chromium in basaltic systems has attracted a lot of interest. It is however
360 difficult to model its behaviour in the presence of spinel because Cr is highly compatible in this mineral
361 and therefore its presence controls the way chromium will be partitioned between the solid and the
362 silicate melt. The solubility of chromium in silicate melts has been extensively studied and, in the
363 presence of spinel, the main factors controlling the Cr concentration in the melts are temperature and
364 oxygen fugacity (Hanson and Jones, 1998, Roeder and Reynolds 1991). In general, lower temperature
365 favours Cr^{3+} in the silicate melts as does higher $f\text{O}_2$ (Li et al. 1995). At fixed $f\text{O}_2$, a decrease in
366 temperature is associated with a decrease in the saturated Cr concentration in the silicate melts (e.g.

367 Murck and Campbell 1986, Hanson and Jones 1998, Roeder and Reynolds 1991). The saturated Cr
368 concentration in silicate melt vary from $\sim 3000 \mu\text{g g}^{-1}$ at 1450°C to $\sim 300 \mu\text{g g}^{-1}$ at 1200°C (Murck and
369 Campbell 1986). Importantly, the $\text{Cr}^{3+}/\text{Cr}_{\text{TOT}}$ ratio in natural basalts are unknown due to the reaction
370 $\text{Fe}^{3+} + \text{Cr}^{2+} = \text{Fe}^{2+} + \text{Cr}^{3+}$ that happens during quenching (Berry et al. 2004). Depending on the physical
371 conditions during melting, the Cr concentration in silicate melts can vary from $5000 \mu\text{g g}^{-1}$ (lunar
372 basalts) to $500 \mu\text{g g}^{-1}$ (MORB). If initial melts in Fangataufa have formed in typical upper mantle
373 conditions ($f\text{O}_2 = \text{QFM} \pm 1$ and $T = 1300^\circ\text{C}$), we could have assumed that regardless of partial melting
374 the Cr concentration is between 500 and $600 \mu\text{g g}^{-1}$. However, our Th-REE modelling shows that partial
375 melting happened in the garnet stability field. Using garnet as a phase residue during melting implies
376 that the overall D decreases. As a consequence, the Cr concentration in the parental melt is controlled
377 by the partition coefficient of Cr between garnet, clinopyroxene and orthopyroxene and the silicate melt
378 (Table 2), and this results in a concentration of Cr slightly higher in the primary melts, between 630 and
379 $700 \mu\text{g g}^{-1}$.

380 The models presented in Figures 7 and S4 combine partial melting of a mixed mantle source
381 (peridotite + “fertile” component) and fractional crystallisation of olivine, clinopyroxene and spinel.
382 During partial melting, the average D values between crystals and melt for Cr and Ni are 5.5 and 8.5 ,
383 respectively (Table 2). Thus, the Cr and Ni concentrations in the primary melts vary little with changing
384 degree of partial melting: as stated above, the concentration of Cr in the melt ranges from 630 to 700
385 $\mu\text{g g}^{-1}$, whereas the concentration of Ni is buffered by the presence of olivine in the source and is about
386 $200 \mu\text{g g}^{-1}$. For fractional crystallisation, we assume it happened at low pressure and we consider a
387 crystallizing assemblage composed of olivine (50%), clinopyroxene (45%) and spinel (5%). The large
388 variations in Cr and Ni concentrations can be then explained by up to 30 % fractional crystallisation. In
389 this model, during crystallisation the Cr budget is 58% in spinel, 37% in cpx and 5% in olivine. It has
390 to be mentioned that values for Cr partition coefficients available in the literature are strongly variable
391 and therefore this can affect the models presented in this study. It is especially true for clinopyroxene
392 and spinel with experimentally determined partition coefficients varying over one order of magnitude
393 (e.g. Hauri et al. 1994, Liu and O’Neill 2004, Mallmann and O’Neill 2009). However, the main
394 conclusions reached in this discussion would not change.

395 In summary, a varying degree of partial melting does not explain the variations in Cr and Ni
396 contents measured in Fangataufa basalts as opposed to incompatible elements. Modelling shows that
397 fractional crystallisation of a Cr-bearing phases (clinopyroxene and spinel) and olivine is the main
398 process controlling the behaviour of chromium. Moreover, the melts produced by low degree of partial
399 melting are also characterised by more evolved liquid in term of fractional crystallisation.

400

401 5.4. Implication for Cr isotope fractionation during magmatic differentiation

402 The variations in Cr isotopic compositions in Fangataufa basalts are presented in Figure 4. The
403 observed variations are small which suggests that igneous processes have a limited impact on the Cr
404 isotopic composition of terrestrial basalts. First, it is important to note that there is no correlation
405 between Cr isotopes and LOI which indicates that the processes responsible for the high LOI values in
406 Fangataufa basalts do not affect their Cr isotopic compositions. We therefore argue that the observed
407 variations in Cr isotopes are due to igneous processes. The samples with high MgO have on average a
408 heavier isotopic composition and the samples with less Cr have on average a lighter Cr isotopic
409 composition compared to Cr rich samples. As explained above, the variations in Cr and MgO
410 concentrations can be explained by a variation in the rate of fractional crystallisation during magmatic
411 differentiation. In Figure 8, we present a Rayleigh fractionation model to reproduce the small but
412 nonetheless resolvable Cr isotopic variations. For the low-K series, we assume an initial Cr
413 concentration and isotopic composition of $670 \mu\text{g g}^{-1}$ and -0.18 ‰ , respectively. The initial Cr
414 concentration was obtained from the modelling done in Figure 7, where we calculated the Cr
415 concentration in the silicate liquid at equilibrium with a mixed source (98% garnet peridotite and 2%
416 “fertile” component) at 7% partial melting. We decided to use the average of the Cr-rich samples ($>$
417 $500 \mu\text{g g}^{-1}$) as a starting point in the model with an isotopic composition of $-0.18 \pm 0.02 \text{ ‰}$. The
418 variations observed can be modelled with a Rayleigh fractionation model with a bulk isotopic
419 fractionation ($\Delta^{53}\text{Cr}_{\text{melt-crystals}}$) of $-0.010 \pm 0.005 \text{ ‰}$ (Fig. 8). For the high-K series, the initial Cr
420 concentration is more difficult to constrain. If the high-K series has the same modal proportion in the
421 source as the low-K series, then its initial Cr concentration is likely to be very similar. In this case,
422 assuming the same initial $\delta^{53}\text{Cr}$, the Cr isotopic composition in the high-K series can be modelled with

423 a fractionation factor ($\Delta^{53}\text{Cr}_{\text{melt-crystals}}$) of -0.020 ± 0.010 ‰, slightly higher than that of the low-K series.
424 If the initial Cr concentration in the high-K series was lower than the low-K series because of a change
425 in the bulk partition coefficient between the residue and the melt, then the fractionation factor needed
426 to explain the observed variations would be even higher.

427 The fractionation factors used in our models for Fangataufa basalts are smaller than that of
428 lunar basalts. In Figure 8, we also consider a fractional crystallisation model for the lunar basalts. This
429 model shows that the isotopic fractionation ($\Delta^{53}\text{Cr}_{\text{melt-crystals}} = -0.07$ ‰) for the lunar samples is higher
430 than that of Fangataufa basalts. This difference can have several origins. First, the mineralogy of the
431 crystallising assemblage may be different in both terrestrial and lunar conditions. As described above,
432 clinopyroxene is a major component of the crystallising assemblage in terrestrial basalts, and in the
433 models presented in Figure 7, Cr in cpx accounts for about ~50% of total Cr. Clinopyroxene has a
434 lighter isotopic composition compared to chromite (Shen et al. 2018) and crystallising large amount of
435 cpx will decrease the bulk fractionation factor between melt and crystals. The second option is linked
436 to the fact that the redox conditions on the Moon and the Earth are very different. The Cr^{3+} concentration
437 in the silicate melts is mainly controlled by two equilibria: (i) the homogeneous equilibrium $\text{CrO} + \frac{1}{4}$
438 $\text{O}_2 = \text{CrO}_{1.5}$ and (ii) chromite saturation (e.g. $2\text{CrO}_{1.5} + \text{MgO} = \text{MgCr}_2\text{O}_4$). In lunar conditions (reduced),
439 Cr is believed to be mainly Cr^{2+} (Sutton et al. 1995) and the $\text{Cr}^{3+}/\text{Cr}_{\text{TOT}}$ ratio is much lower in lunar
440 basalts (0.1 at IW-1) than in terrestrial basalts (0.6-0.7 at QFM). It is likely that the bulk fractionation
441 factor during fractional crystallisation is controlled by the difference in $\text{Cr}^{3+}/\text{Cr}_{\text{TOT}}$ ratio between the
442 melts and the crystallising phases. On the Moon, there is a considerable difference between $\text{Cr}^{3+}/\text{Cr}_{\text{TOT}}$
443 ratio in the melt ($\text{Cr}^{3+}/\text{Cr}_{\text{TOT}} = 0.1$) and the crystallising phases (e.g. $\text{Cr}^{3+}/\text{Cr}_{\text{TOT}} = 1$ in chromite) which
444 result in larger isotopic shifts. Under terrestrial conditions, the difference in $\text{Cr}^{3+}/\text{Cr}_{\text{TOT}}$ between silicate
445 melts and crystallising phases is smaller resulting in limited Cr isotopic variations.

446

447 5.5. Cr isotopes behaviour during partial melting

448 It has been proposed in the literature that Cr isotopes are fractionated during partial melting
449 of the mantle (Xia et al. 2017, Shen et al. 2018). While partial melting proceeds, the residue would
450 become isotopically heavier. Accordingly, the isotopic composition of silicate melts at equilibrium with

451 mantle peridotites (that is to say, primary melts) should be isotopically lighter compare to their sources.
452 The difficulty, however, is to determine the Cr isotopic composition of the primary melts. In order to
453 determine the Cr isotopic composition of the parental melt of Fangataufa basalts, we consider the
454 average of the Cr-rich samples analysed in this study. The obtained value (Cr isotopic composition of
455 $\delta^{53}\text{Cr} = -0.18 \pm 0.02 \text{ ‰}$ (2sd, n = 3)) is on the low end of the BSE values of $\delta^{53}\text{Cr} = -0.12 \pm 0.10 \text{ ‰}$
456 (Schoenberg et al. 2008) and $\delta^{53}\text{Cr} = -0.11 \pm 0.06 \text{ ‰}$ (Sossi et al. 2018). Interestingly, it is lighter than
457 most Cr isotopic composition reported for mantle xenoliths. This observation confirms that Cr isotopes
458 are fractionated during low degree partial melting in mantle conditions and that the melts are enriched
459 in light Cr isotopes compared to the source. Using our primary Cr isotopic composition and the BSE
460 value of Sossi et al. 2018, we propose a bulk fractionation factor ($\Delta^{53}\text{Cr}_{\text{melt-residue}}$) during low-degree
461 partial melting of $-0.07 \pm 0.06 \text{ ‰}$. This fractionation is small and cannot solely explain the large Cr
462 isotopic variations reported in mantle xenoliths ($-0.51 < \delta^{53}\text{Cr} < 0.74\text{‰}$, Xia et al. 2017). Assuming an
463 isotopic fractionation of $\Delta^{53}\text{Cr}_{\text{melt-residue}}$ of -0.07 ‰ ($\Delta^{53}\text{Cr}_{\text{melt-residue}} = \delta^{53}\text{Cr}_{\text{melt}} - \delta^{53}\text{Cr}_{\text{residue}}$), partial melting
464 of the mantle can only explain up to 0.05 ‰ variations in mantle xenoliths. As stated by Shen et al.
465 (2018), the variations caused by partial melting is small compared to the reported 1.2 ‰ variations in
466 mantle xenoliths (Xia et al. 2017).

467 It has been previously proposed that the fractionation factor between the residues and products
468 of partial melting changes with the degree of partial melting (Shen et al. 2018). Interestingly, Fangataufa
469 basalts form two distinct groups in $\delta^{53}\text{Cr}$ -Nd isotope space (Fig. 5). We have previously proposed that
470 the variations in Nd isotopes reflect a difference in productivity between the “fertile” and peridotite
471 components with partial melting. Indeed, the correlation between Nd isotopes and La/Yb ratio suggests
472 that low degree of partial melting is associated with less radiogenic Nd isotopic composition. At face
473 value, the covariation between Cr and Nd isotopes could be explained by a change in the fractionation
474 factor between products and residues during partial melting. This finding would agree with the model
475 proposed by Shen et al. (2018) performed with spinel peridotite. However, the shift in Cr isotopic
476 composition between the high-K and the low-K series is opposite to what the model predicts. Using the
477 Shen et al. (2018) model and assuming the same initial Cr isotopic composition for the source, it can be
478 shown that melts produced at low degree of partial melting should be isotopically heavier than melts

479 produced at higher degree of melting. Instead, our results show that melts produced by low degree of
480 partial melting have lighter Cr isotopic composition because the Cr isotopic composition of the high-K
481 series is lighter compare to the low-K series.

482 It is however important to note that the samples with less radiogenic Nd isotopic composition
483 have experienced more fractional crystallisation than the melts produced by higher degrees of partial
484 melting. Indeed, as described above, the samples with high Th and/or La/Yb ratios have lower Cr and
485 Ni concentrations. It is therefore possible that the covariation between Cr and Nd isotopes is due to the
486 fact that samples with lower degrees of partial melting are more evolved basalts and the lighter Cr
487 isotopic composition is due to fractional crystallisation rather than a change in the rate of partial melting.
488 This is supported by the similar isotopic composition between the samples with the highest Cr
489 concentration in each suite. The difficulty with determining the initial Cr concentration in the high-K
490 series do not allow us to say whether partial melting or fractional crystallisation are responsible for the
491 grouping between the high-K and the low-K series in the Nd vs Cr isotopes plot. More work is needed
492 to test this hypothesis. It is also interesting to note that until the mechanisms responsible for the
493 fractionation during partial melting are better understood it is impossible to reconstruct the Cr isotopic
494 composition of terrestrial reservoirs using melts produced by partial melting of the mantle.

495

496 6. Conclusions

497 We have investigated the Cr isotopes behaviour during igneous processes. To this end, 21
498 samples from Fangataufa Island were measured for their major and trace elements concentrations, Sr
499 and Nd isotopic composition and finally for their Cr isotopic composition. Fangataufa basalts are
500 characterised by relatively small variations in major elements but two series are identified (low-K and
501 high-K) based on their K_2O and TiO_2 concentrations. The major element concentrations also indicate
502 that Fangataufa samples have experience some magmatic differentiation and that both olivine and
503 clinopyroxene crystalized.

504 The variations in incompatible trace element content (e.g. REE, Th) in Fangataufa basalts can
505 be explained by changes in degree of partial melting of the mantle source. The variations in La/Yb ratio
506 indicate that the melting producing Fangataufa basalts happened in the garnet stability field. The

507 variations in Nd isotopes suggest that the mantle source is heterogeneous and composed of a peridotite
508 and a small amount of “fertile” component. The variations in compatible elements such as Cr and Ni in
509 Fangataufa basalts can be explained by fractional crystallisation of olivine, clinopyroxene and chromite.
510 The models presented in this study suggest that relatively small amount of crystallisation can explain
511 the large range of variation in Cr and Ni concentrations measured in natural samples.

512 Fangataufa basalts are characterised by variations in Cr isotopes from -0.23 to -0.17 ‰. Overall,
513 the Cr-rich samples have heavier Cr isotopic composition compared to Cr-poor samples. The small but
514 nonetheless distinguishable variations in Cr isotopes are better explained by fractional crystallisation.
515 The low-K and high-K suites can be modelled with fractionation factors ($\Delta^{53}\text{Cr}_{\text{melt-residue}}$) of $-0.010 \pm$
516 0.005 ‰ and -0.020 ± 0.010 ‰, respectively. Compared to the lunar basalts, the isotopic fractionation
517 between the melt and the crystallising phases is smaller in terrestrial samples and two hypotheses could
518 explain this observation. The mineralogy with chromite (on the Moon) to clinopyroxene + chromite in
519 the studied setting could change the bulk isotopic fractionation and possibly the more oxidised
520 conditions on Earth could also limit the fractionation factor. Finally, the more primitive basalts have
521 slightly lower Cr isotopic composition compared to mantle xenoliths suggesting that Cr isotopes are
522 fractionated during partial melting with the melts being lighter than the residues. However, the small
523 difference between basalts and mantle xenolith cannot explain the large variations observed in mantle
524 xenoliths. The covariation between $\delta^{53}\text{Cr}$ and $\epsilon^{143}\text{Nd}$ values in Fangataufa basalts can be explained by
525 two processes: (i) lower degree of partial melting have lighter Cr isotopic composition and (ii) melts
526 produced by low degree of partial melting have also experienced more fractional crystallisation.

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716 Conflicts of interest:

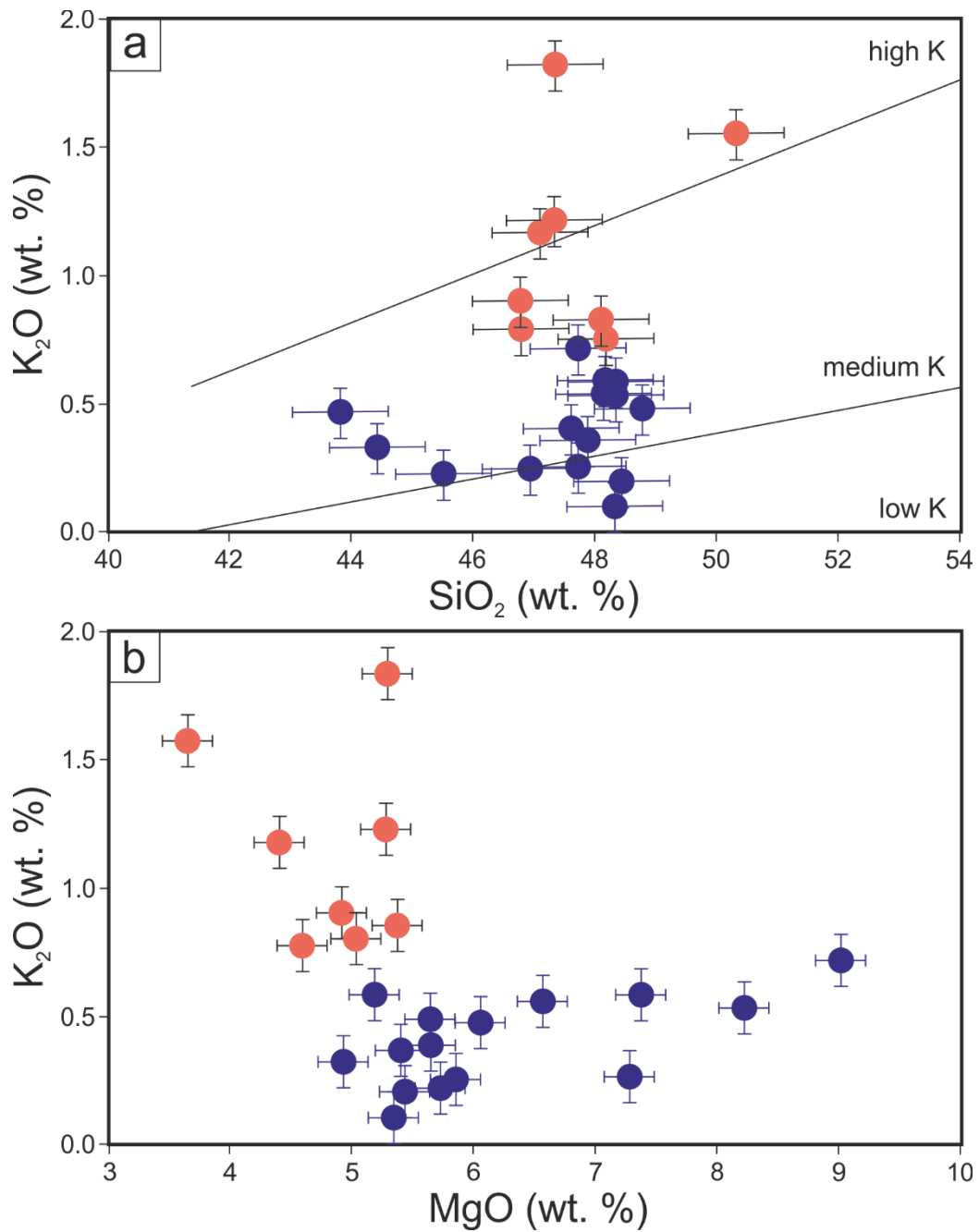
717 There are no conflicts of interest.

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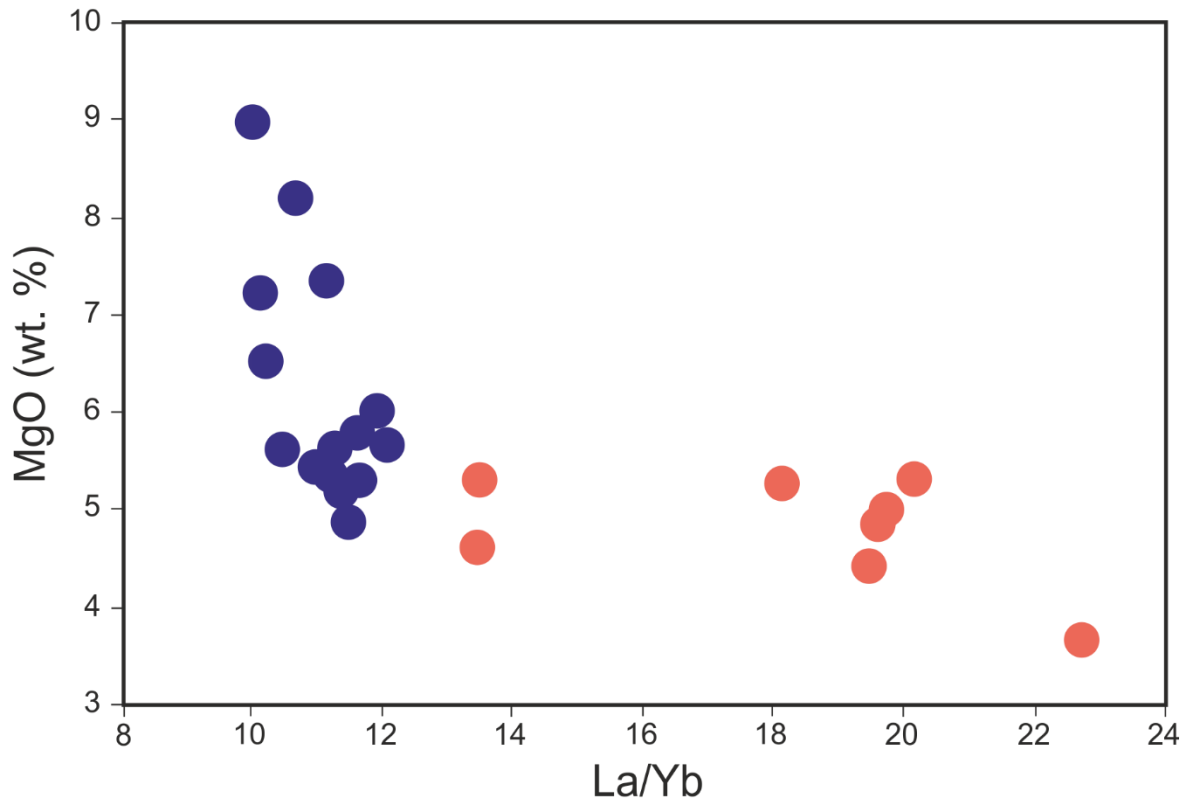


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723 Figure 1: K₂O vs (a) SiO₂ and (b) MgO concentrations in the samples analysed in this study. The
724 colours are for the two series (red for high-K and blue for low-K) observed in the Fangataufa samples
725 (see text for details).

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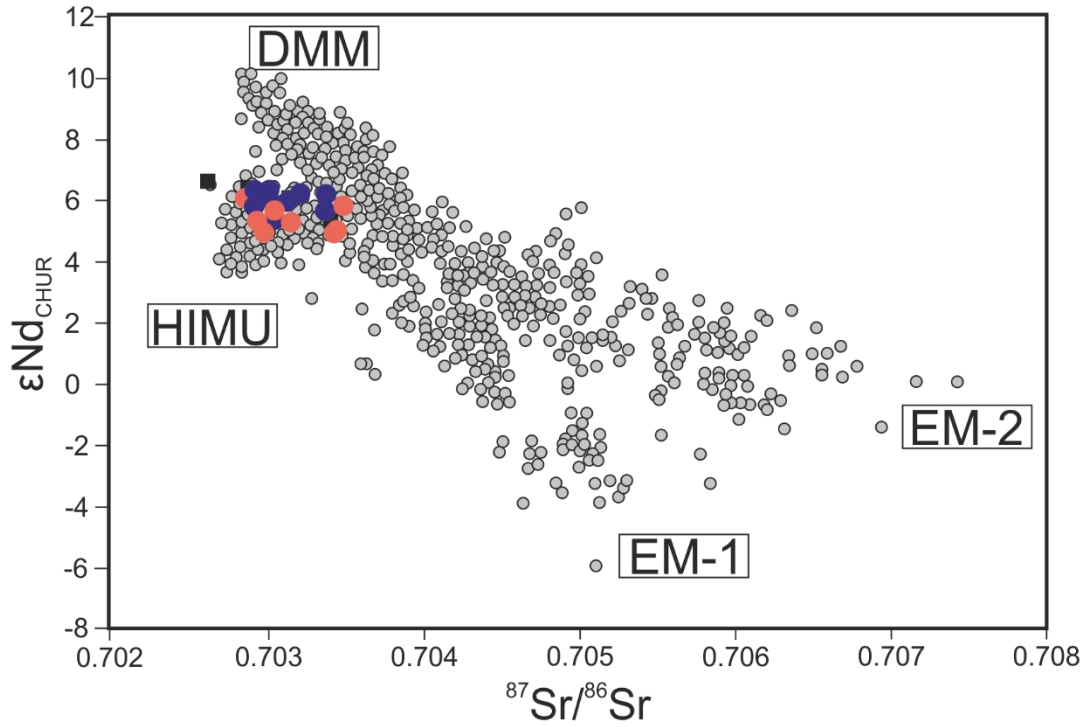
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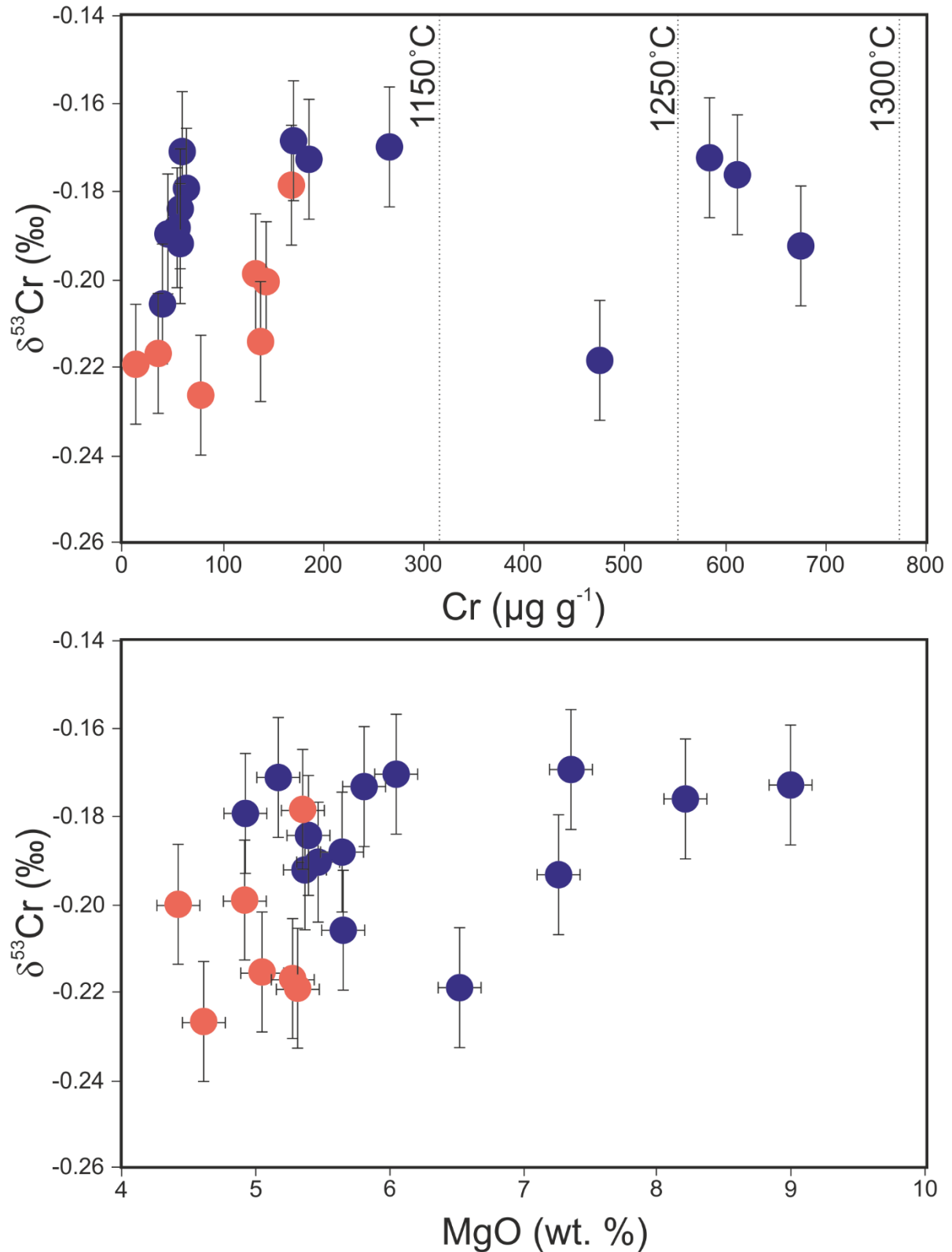
Figure 2: MgO vs La/Yb ratios for the samples analysed in this study. Colours defined in Fig. 1.

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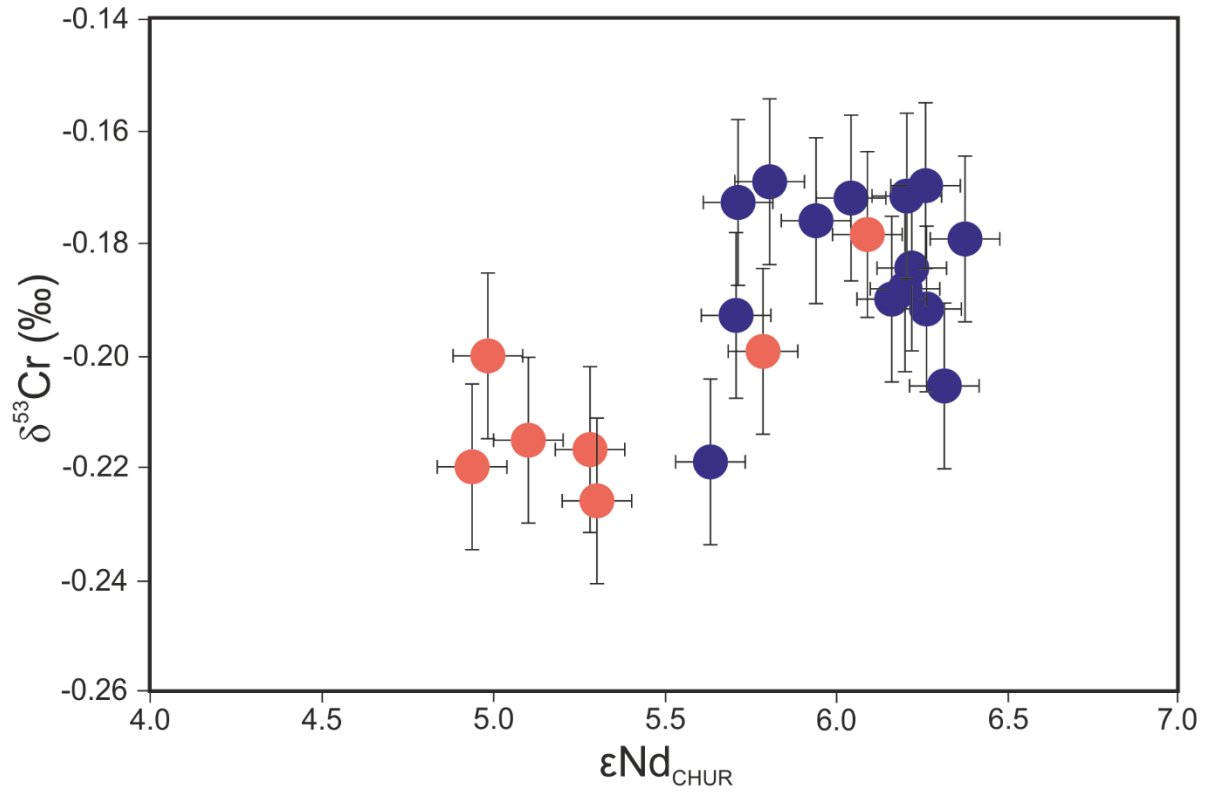


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Figure 3: $\epsilon\text{Nd}_{\text{CHUR}}$ versus Sr isotopic composition for Ocean Island basalts. The blue and red circles are the data from this study (colours defined in Figure 1). The black squares are Fangataufa samples from Bardintzeff et al. (1994). The grey circles are OIB samples from the literature. Modified after Hoffman 2007.

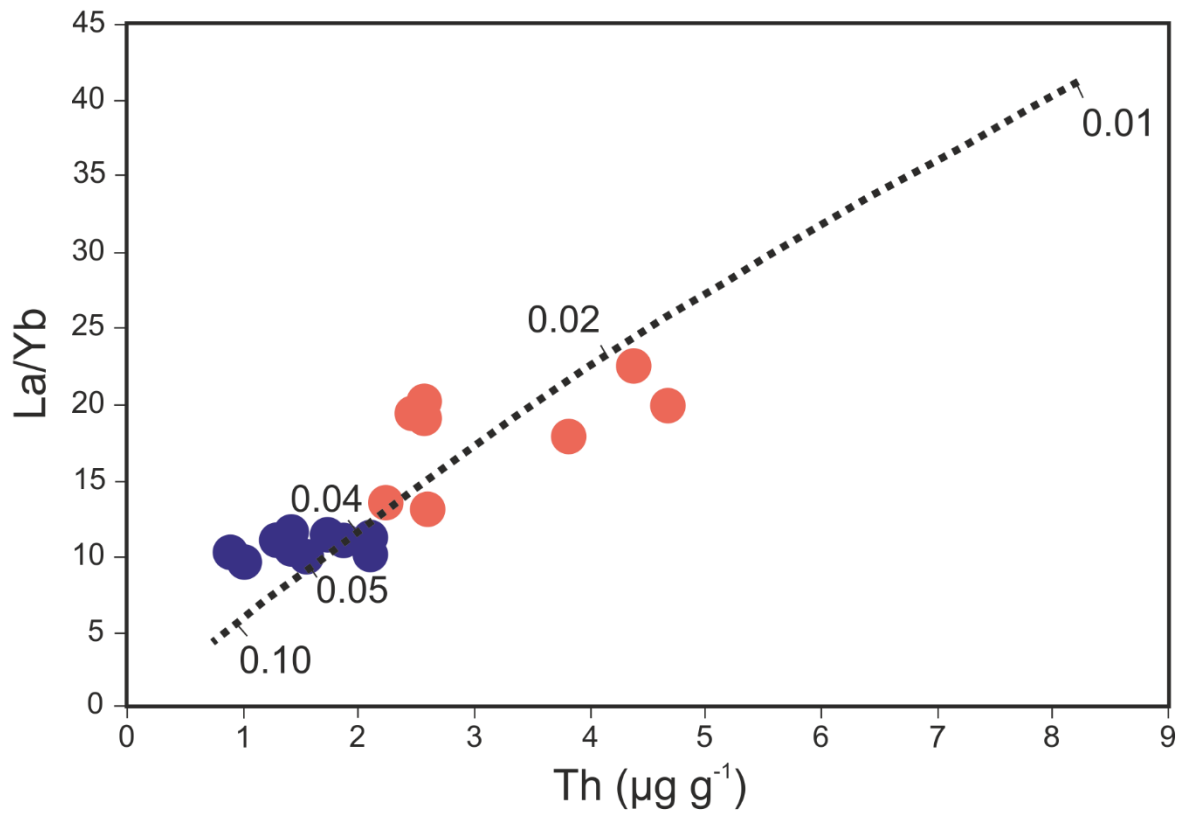


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 773 Figure 4: $\delta^{53}\text{Cr}$ versus (a) Cr and (b) MgO concentrations for the samples analysed in this study. Errors
 774 bars are the external reproducibility obtained in this study (see text for details). Colours defined in
 775 Figure 1. The vertical dotted lines in (a) are the Cr saturation concentrations in silicate melts at the given
 776 temperature at QFM (Murck and Campbell 1986).
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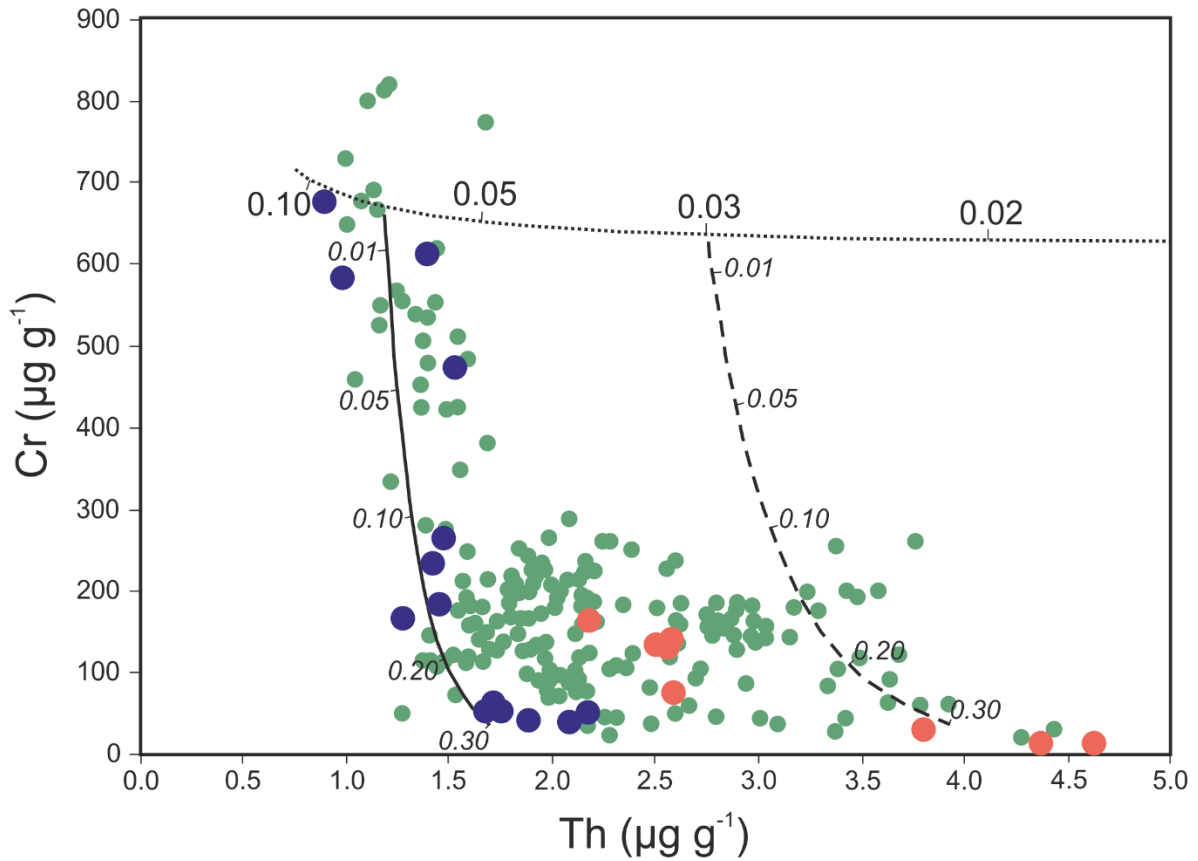


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Figure 5: Cr versus Nd isotopic compositions for the Fangataufa basalts. The colour coding is defined in Figure 1.



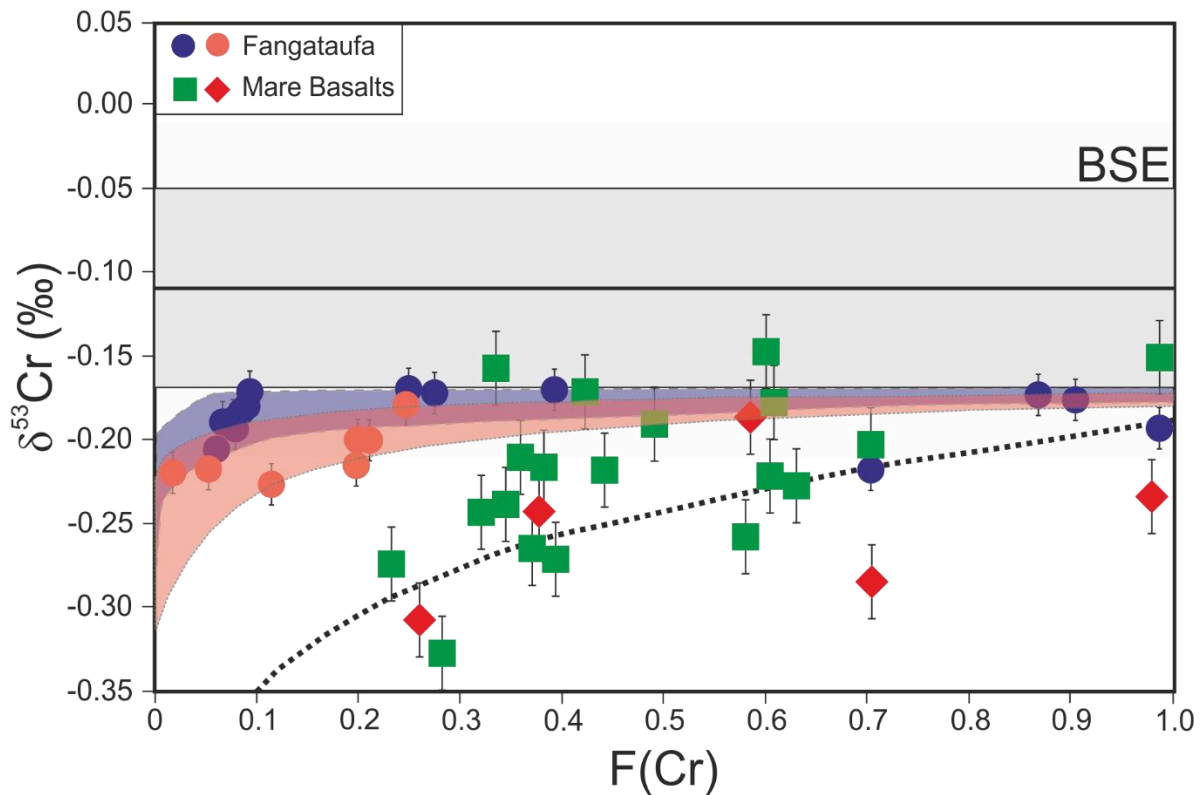
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 794 Figure 6: La/Yb ratios versus Th concentrations in Fangataufa basalts. The colours are the same as in
 795 Figure 1. The dotted line is the partial melting model discussed in the text in the section 5.3. The
 796 numbers are the degrees of partial melting (1 = 100% melt).
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 823 Figure 7: Chromium versus Th concentrations in Fangataufa basalts. The blue and red circles are the
 824 samples analysed in this study. The green circles are literature data together with unpublished data
 825 (Schiano et al. 1993, unpublished data, P. Schiano). The dotted line is the partial melting model
 826 discussed in the text. The solid and dashed lines are fractional crystallisation models for 7 and 3 %
 827 partial melts, respectively. The numbers are the proportion of partial melting and fractional
 828 crystallisation (*italic*). Colours defined in Fig. 1.

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857 Figure 8: $\delta^{53}\text{Cr}$ versus the amount of Cr left (F) in the melt during fractional crystallisation for the
858 Fangataufa basalts and the lunar basalts from Bonnand et al. (2016) and Sossi et al. (2018). F was
859 calculated using the equation: $F = \text{Cr}_{\text{sample}} / \text{Cr}_{\text{max}}$, where Cr_{max} is the highest concentration in both
860 Fangataufa and lunar basalts. The grey horizontal bars are the current estimates of the BSE (dark grey:
861 Sossi et al. 2018, light grey: Schoenberg et al. 2008). The dotted line is a Rayleigh fractionation model
862 for lunar basalts. The blue and red shaded areas are the fractional models for the low-K and high-K
863 suites, respectively. The isotopic fractionation ($\Delta^{53}\text{Cr}$) used for Fangataufa basalts are -0.010 ± 0.005
864 ‰ (low-K suite) and $-0.020 \pm 0.010 \text{‰}$ (high-K suite). For the lunar basalts, the fractionation factor is
865 -0.07‰ (see text for details). Error bars are the external reproducibility reported in each study.

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Table 1: Selected major and trace element concentrations, La/Yb ratio and Sr, Nd and Cr isotopic compositions for the samples analysed in this study.

Sample name	MgO* wt. %	Cr $\mu\text{g g}^{-1}$	Ni $\mu\text{g g}^{-1}$	La $\mu\text{g g}^{-1}$	Yb $\mu\text{g g}^{-1}$	Th $\mu\text{g g}^{-1}$	$^{87}\text{Sr}/^{86}\text{Sr}$	$\epsilon\text{Nd}_{\text{CHUR}}$	$\delta^{53}\text{Cr}$ ‰	La/Yb
F107	7.36	169.1	144.1	15.2	1.4	1.3	0.703052	5.81	-0.169	11.2
F120	5.82	186.8	92.7	17.3	1.5	1.5	0.702908	6.04	-0.172	11.7
F122	7.28	677.7	292.5	10.1	1.0	0.9	0.703388	5.71	-0.193	10.2
F123	5.72	236.3	98.2	17.1	1.4	1.4	0.702923	6.17	n.d.	12.2
F124	4.61	77.9	56.3	25.5	1.9	2.6	0.702923	5.30	-0.226	19.4
F125	5.36	53.8	46.2	19.7	1.7	1.7	0.703218	6.27	-0.192	11.7
F126	5.45	45.3	43.2	20.0	1.8	1.9	0.702930	6.15	-0.190	11.0
F127	5.17	63.3	57.1	18.9	1.7	1.7	0.702902	6.21	-0.171	11.4
F128	9.00	586.7	246.2	11.2	1.1	1.0	0.702915	5.71	-0.173	10.0
F129	4.93	61.4	49.9	19.5	1.7	1.7	0.702927	6.37	-0.179	11.5
F130	5.39	58.1	46.9	19.4	1.7	1.7	0.702897	6.22	-0.184	11.3
F131	5.64	41.2	34.1	20.4	1.9	2.1	0.703018	6.31	-0.205	12.0
F132	5.65	52.5	41.7	22.0	1.9	2.2	0.703384	6.20	-0.188	10.2
F134	6.04	265.5	129.3	17.0	1.4	1.5	0.702998	6.26	-0.170	10.7
F141	6.55	476.7	168.9	15.5	1.5	1.5	0.703045	5.63	-0.219	10.2
F165	8.22	612.4	193.7	14.4	1.3	1.4	0.703129	5.94	-0.176	10.7
F17	5.04	134.3	56.3	29.2	1.5	2.6	0.703457	5.10	-0.215	13.5
F172	5.29	12.6	39.0	47.1	2.3	4.6	0.702989	4.94	-0.219	19.6
F176	5.27	35.2	52.8	39.0	2.2	3.8	0.703158	5.28	-0.217	19.7
F188	5.35	167.3	81.7	21.5	1.6	2.2	0.702865	6.09	-0.179	20.1
F19	4.91	134.5	61.0	28.9	1.5	2.5	0.703491	5.79	-0.199	22.8
F7	4.42	142.8	54.5	29.0	1.5	2.6	0.703429	4.99	-0.200	10.5
F80	3.68	3.4	4.8	47.3	2.1	4.4	0.703032	5.87	n.d.	13.4

CHUR value for $\epsilon\text{Nd} = 0.51263$;

n.d. : not determined

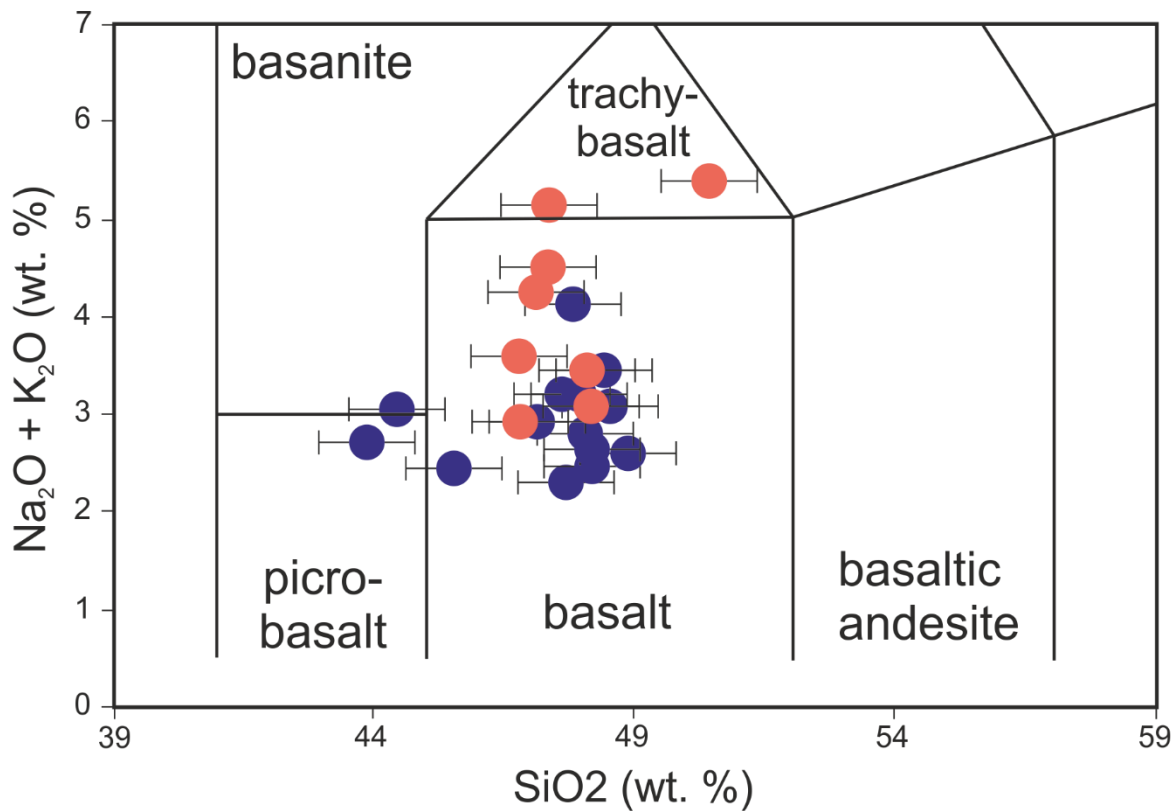
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906 Table 2: Concentrations and partition coefficients used in the partial melting and fractional
 907 crystallisation models presented in the study. See text for details. The mantle and “fertile” component
 908 concentrations are from McDonough and Sun (1995) and Gale et al. (2013), respectively. The partition
 909 coefficients are from Liu and O’Neill 2004, McKenzie and O’Nions 1995, Villemant et al. 1981 and
 910 Salters and Stracke 2004.

	Mantle	fertile component	$D_{\text{olivine-melt}}$	$D_{\text{opx-melt}}$	$D_{\text{cpx-melt}}$	$D_{\text{garnet-melt}}$	$D_{\text{spinel-melt}}$
Cr	2625	250	0.8	9	7	5.5	170
Ni	1970	92	9.4	9.4	2.5	4.93	0
La	648	5210	0.0005	0.004	0.015	0.0007	0
Ce	1675	14860	0.0005	0.004	0.038	0.017	0
Nd	1250	12030	0.00042	0.012	0.0884	0.064	0
Sm	406	3820	0.0011	0.02	0.1509	0.23	0
Gd	544	4990	0.0011	0.065	0.16	1.2	0
Dy	674	6080	0.0027	0.065	0.17	2	0
Er	438	3790	0.013	0.065	0.18	3	0
Yb	441	3630	0.02	0.08	0.25	5.5	0
Lu	67.5	530	0.02	0.12	0.276	7	0
Th	79.5	404	0.00005	0.002	0.00566	0.009	0

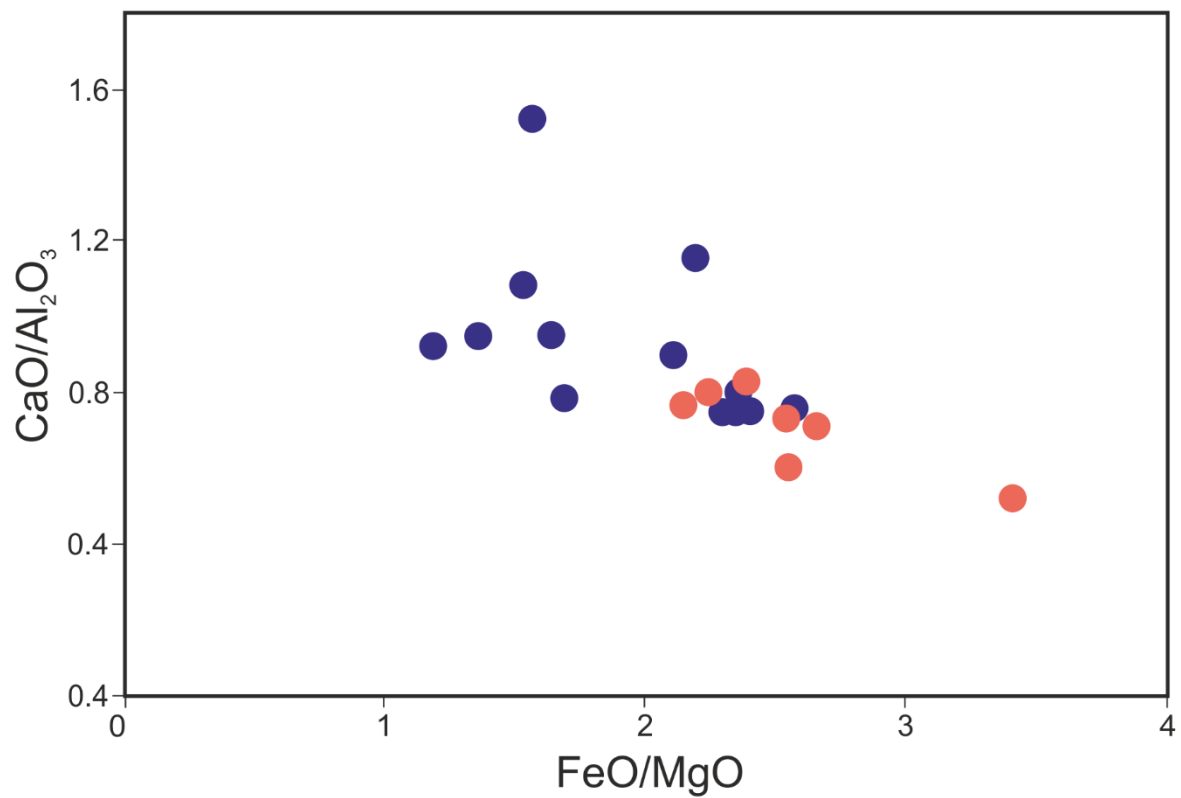
911 Cr and Ni concentrations are in ($\mu\text{g} \cdot \text{g}^{-1}$) and REE and Th concentrations are in ($\text{ng} \cdot \text{g}^{-1}$).

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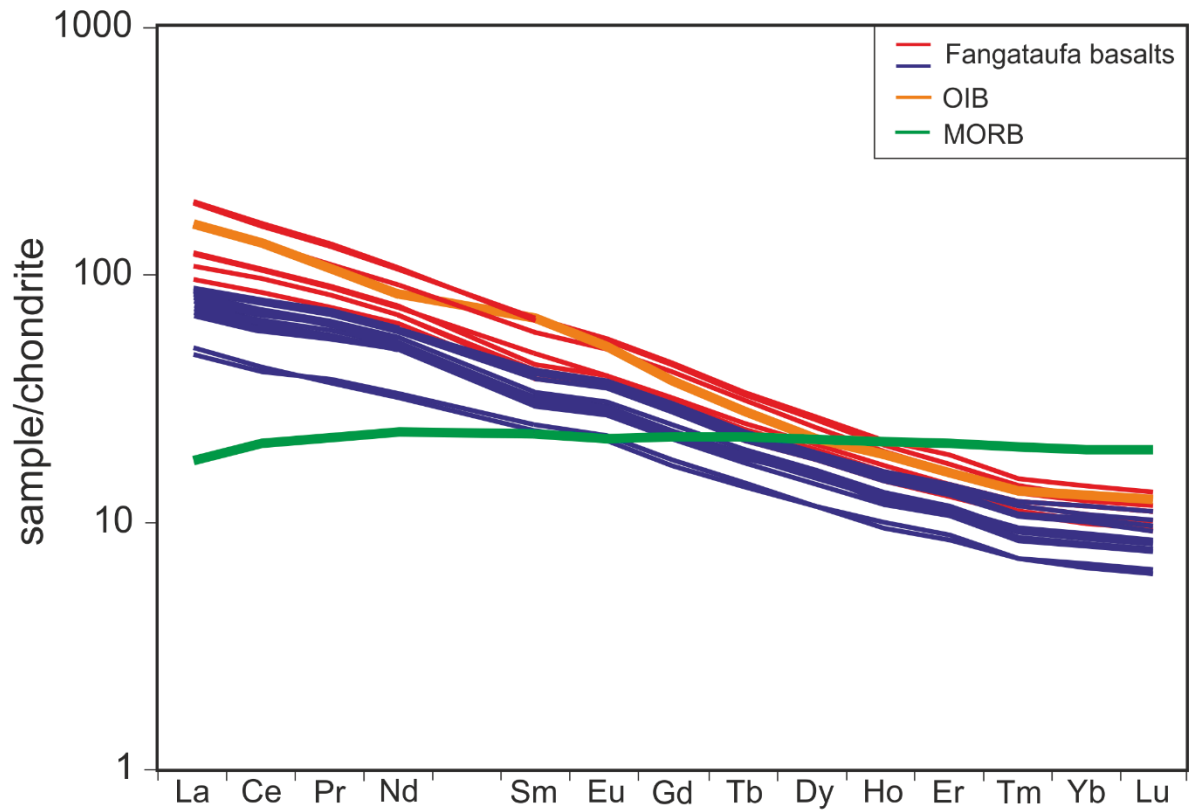
943
944 Figure S1: $\text{Na}_2\text{O} + \text{K}_2\text{O}$ as a function of SiO_2 concentrations for the Fangataufa samples analysed in
945 this study. The colours are for the two series (red for high-K and blue for low-K) observed in the
946 Fangataufa samples (see text for details).

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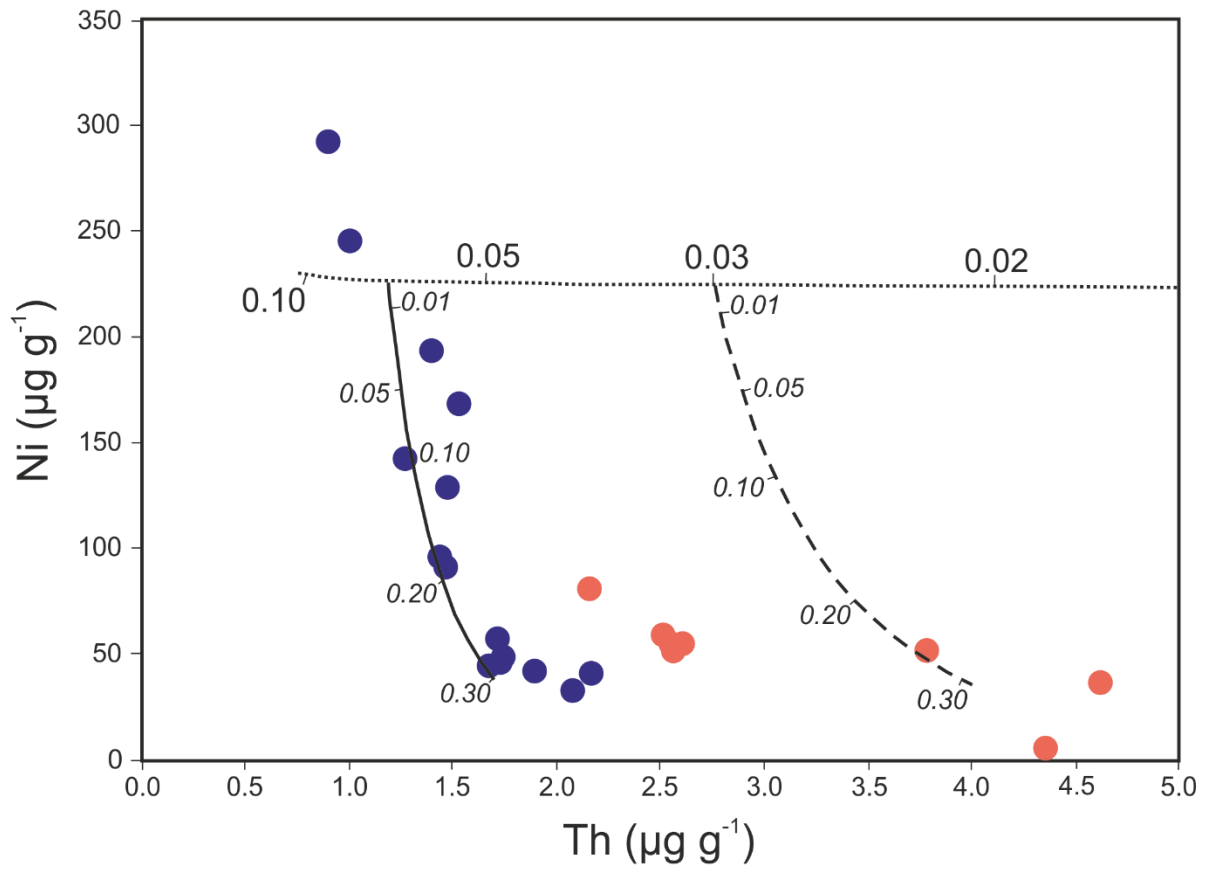


950 Figure S2: CaO/Al₂O₃ versus FeO/MgO ratios in Fangataufa basalts. The colour coding is defined in
951 Figure S1.
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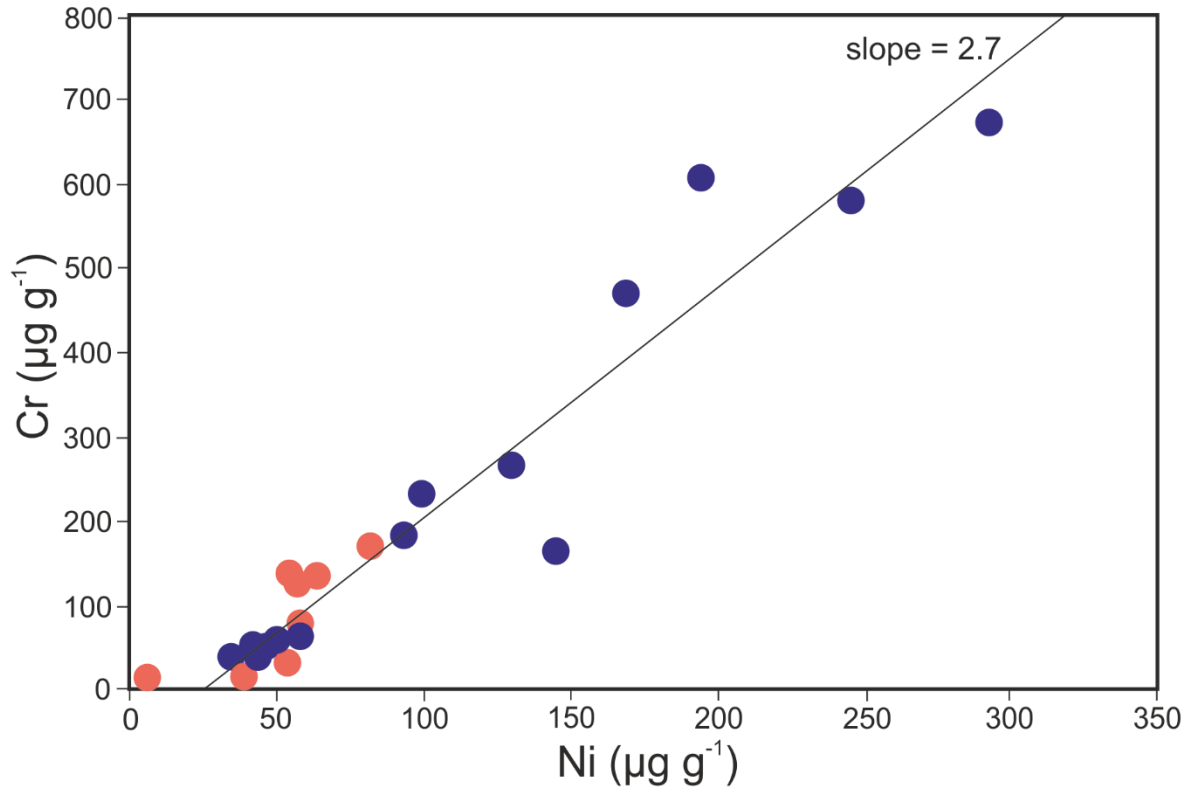
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 963 Figure S3: Chondrite normalised REE patterns for Fangataufa basalts. The OIB data is from Sun and
 964 McDonough (1989) and the MORB concentrations are from Gale et al. (2013). The chondrite values
 965 used for normalisation is from Barrat et al. (2012). The colour coding is defined in Figure S1.
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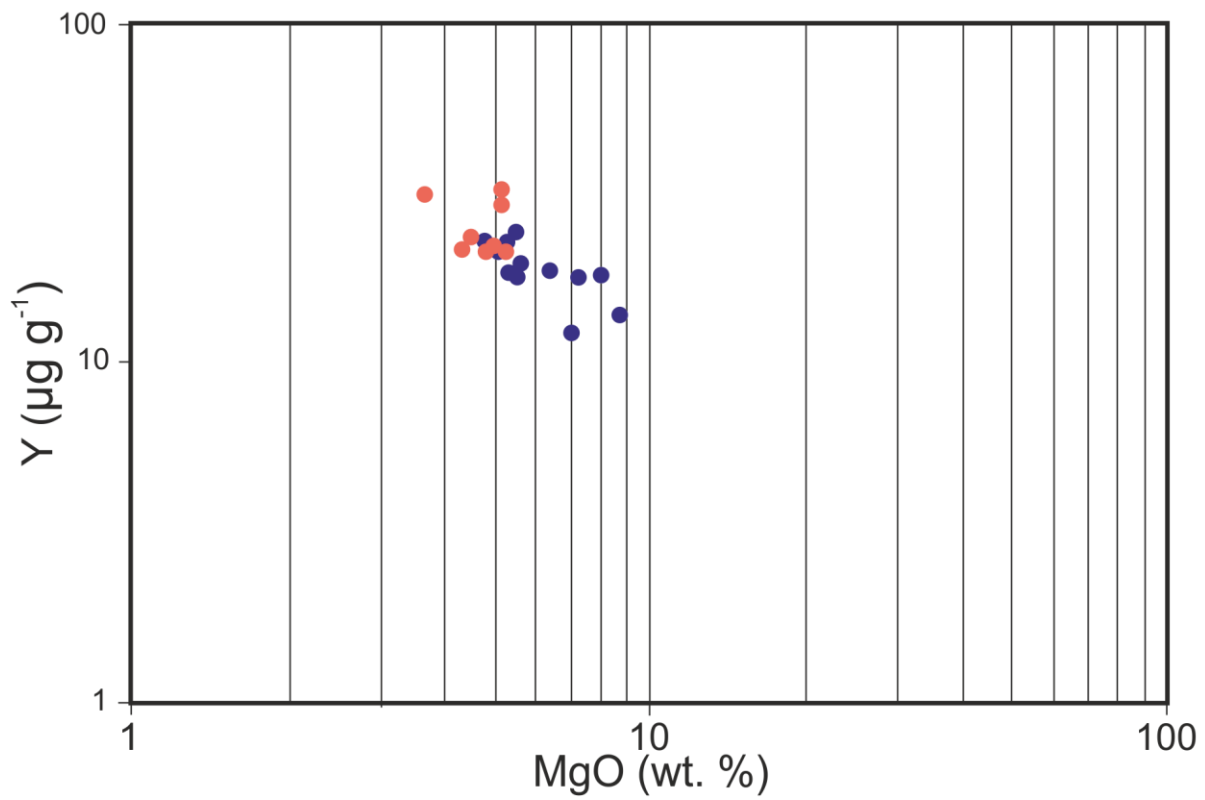


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 974 Figure S4: Nickel versus Th concentrations in Fangataufa basalts. The blue and red circles are the
 975 samples analysed in this study. The dotted line is the partial melting model discussed in the text. The
 976 solid and dashed lines are fractional crystallisation models for 7 and 3 % partial melts, respectively. The
 977 numbers are the proportion of partial melting and fractional crystallisation (*italic*). The colour coding is
 978 defined in Figure S1.
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Figure S5: Cr concentrations versus Ni concentrations in the samples analysed in this study. The slope between the two concentrations is 2.7. The colour coding is defined in Figure S1.



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996 Figure S6: The variation of MgO and Y plotted on a logarithmic scale. The colour coding is defined in
997 Figure S1.

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