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- New constraints from Central Chile on the origins of enriched continental 5 compositions in thick-crusted arc magmas 6 7 Penny E. Wieser^{1,2*}, Stephen J. Turner³, Tamsin A. Mather¹, David M. Pyle¹, Ivan P. Savov⁴ and Gabriel Orozco⁵ 8 9 ¹ Department of Earth Sciences, University of Oxford, South Parks Road, Oxford OX1 3AN, UK 10 ² Now at Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge, UK, 11 12 CB2 3EQ. 13 ³ Washington University in St. Louis, One Brookings Drive, St. Louis, MO 63130 14 ⁴ School of Earth and Environment, Institute of Geophysics & Tectonics, University of Leeds, Leeds 15 LS2 9JT, UK. 16 ⁵ Red Nacional de Vigilancia Volcánica, Servicio Nacional de Geología y Minería, Av. Santa María 0104, Providencia, Chile. 17 18 19 * Corresponding author: pew26@cam.ac.uk. Phone: 07593670818 20 Abstract 21 Previous work has shown that arc volcanics from thick-crusted continental arcs are compositionally distinct from those erupted in island arcs or continental arcs with thinner crust (<30km). However, 22 the relative influence of the slab, mantle, and upper plate on this variability remains poorly 23
- 24 constrained. Subduction zones have been the loci of continental crust creation for over 2 Ga, so the

25 processes responsible for this variability have important implications for the chemical evolution of 26 the Earth. The Andean Southern Volcanic Zone (SVZ; 33-46° S) is a particularly suitable setting in 27 which to examine the mechanisms leading to enriched magma compositions in continental arcs, 28 because both crustal thickness and magma chemistry vary consistently along strike. However, the 29 scarcity of primitive samples from the northern SVZ, where the continental crust is thickest (50km), 30 has precluded unbiased comparisons between the southernmost and northernmost volcanoes, and 31 may have contributed to an overemphasis of the role of crustal processing in along-arc trends. This study investigates the geochemistry (major and trace element abundances, ⁸⁷Sr/⁸⁶Sr and 32 33 ¹⁴³Nd/¹⁴⁴Nd) of new samples from Don Casimiro and Maipo volcanoes, which lie within the 34 Diamante-Maipo Caldera Complex of the northern SVZ. While evolved Diamante-Maipo samples 35 show evidence for crustal assimilation, the trace element and isotopic enrichment of the primitive 36 samples analysed in this study cannot result from crustal processing, because plausible basement 37 lithologies are not uniformly enriched in all of the necessary elements. Recycling of certain crustal 38 lithologies via subduction erosion could potentially generate the isotopic composition of the 39 Diamante-Maipo magmas, but would result in highly irregular trace element patterns that are not 40 observed in the lavas. We argue that the relative enrichment of the northern SVZ is best explained 41 by the presence of an enriched ambient mantle component (similar to EM1-type ocean island 42 basalts), superimposed on a northward decline in melt extent. Rather than crustal recycling, the 43 EM1-like signatures in arc volcanics may arise from recycling of metasomatized subcontinental 44 lithospheric mantle (M-SCLM). This hypothesis is consistent with the isotopic composition of M-45 SCLM melts across South America, as well as the isotopic compositions of samples from the 46 Argentinian rear arc adjacent to and south of Diamante-Maipo. These rear-arc centres not only 47 corroborate the findings at the arc front, they also demonstrate extensive variability in the supply 48 of slab fluids to regions well behind the arc. Our results caution that if ambient mantle enrichment

49	is not taken into account, petrogenetic models of evolved lavas may exaggerate the role of crustal
50	assimilation, and models for the growth of continental crust may overestimate the amount of
51	continental material that must be recycled back into the mantle to satisfy mass balance.
52	
53	Key Words
54	Mantle heterogeneity
55	Andean Southern Volcanic Zone
56	Highlights
57	• NSVZ isotopic and trace element enrichment indicate an EM1-like mantle source.
58	• EM1-like signatures originate from recycling of M-SCLM from nearby cratons.
59	• Slab melting, low mantle melt extents, and M-SLCM may enrich continental arc lavas
60	• Slab fluids are not supplied vertically from their point of origin.
61	1. Introduction
62	
63	Continental arc magmas, which erupt through thick over-riding crust, are compositionally distinct
64	from oceanic arc magmas. They tend to have elevated incompatible element abundances, steeper
65	incompatible trace element patterns (Fig. 1), and isotopic signatures offset to higher ⁸⁷ Sr/ ⁸⁶ Sr and
66	lower ¹⁴³ Nd/ ¹⁴⁴ Nd than oceanic arc magmas (Leeman et al., 1983; Plank and Langmuir, 1988;
67	Hildreth and Moorbath, 1988; Turner et al., 2015a; Farner and Lee, 2017). Many of these
68	compositional characteristics correlate strongly with the thickness of the arc crust (Turner et al.,
69	2015a), and have even been used as proxies to estimate changes in crustal thickness and rates of
70	surface uplift though time (Profeta et al., 2015; Chiaradia, 2015; Chapman et al., 2015; Scott et al.,
71	2018). Much of the compositional offset between thick and thin-crusted arcs can be accounted for

72 by higher extents of crystal fractionation and crustal assimilation in continental settings (e.g. Farner 73 and Lee, 2017). However, some compositional differences persist even when considering volcanics 74 that have not been extensively overprinted in the crust (e.g. Plank and Langmuir, 1988; Turner and 75 Langmuir, 2015a; Turner et al., 2017; Schmidt and Jagoutz, 2017), which is suggestive of a mantle 76 origin. Compositional differences among "primary" magmas from continental vs. oceanic settings 77 have been attributed to variations in slab temperature (Ruscitto et al., 2012; Turner and Langmuir, 78 2015b; Schmidt and Jagoutz, 2017), different extents of mantle melting (e.g. Plank and Langmuir, 79 1988; Tormey et al., 1991; Turner et al., 2016), subduction erosion (e.g. Stern, 1989), or mantle 80 heterogeneity that is not the direct result of subducting materials (e.g. Hickey et al., 1986; Ewart 81 and Hawkesworth, 1987; Rogers and Hawkesworth, 1989; Hochstaedter et al., 2001; Pearce et al., 82 2007; Turner et al., 2017). Our understanding of the thermal structures of subducting plates, the 83 growth of the continental crust, and the petrogenesis of evolved arc magmas depends critically on 84 constraining the relative influence of each of these factors in producing the enriched compositions 85 of continental arc magmas.

86

87 The Andean Southern Volcanic Zone (SVZ) is an excellent natural laboratory in which to investigate 88 how the thick crusts and lithospheres of active compressional plate margins affect the compositions 89 of magmas added to the continents. The SVZ (33-46° S) is one of four volcanically active segments 90 within the Andean Cordillera, where the Nazca Plate subducts under South America. It is commonly 91 subdivided into three segments: the northern, transitional, and southern SVZ (NSVZ, TSVZ and SSVZ 92 respectively; Dungan et al., 2001; Fig. 2a). From the SSVZ to the NSVZ, crustal thickness increases 93 from ~30-50 km (Fig. 2c), slab depth from 70-120 km (Tassara and Echaurren, 2012), the lower 94 plate age from 16-37 Ma (at 42° S and 33° S respectively, Volker et al., 2011), and trench sediment 95 thickness ranges from 1.3-2.1 km (c.f. Hildreth and Moorbath, 1991). There is also a well-

96 established geochemical gradient from the SSVZ to the NSVZ (Tormey et al., 1991; Hildreth and 97 Moorbath, 1988; Hickey et al., 2016, Fig. 2d-e). Trace element abundances of SSVZ volcanoes, 98 where the crust is thinnest, have similar trace element patterns, and fall within the compositional 99 range of island arcs (Fig. 1). The scarcity of primitive lavas from the NSVZ, which overlies the 100 thickest crust, has made it difficult for prior work to assess the chemical characteristics of NSVZ 101 parental magmas, though an average of the more mafic TSVZ lavas ("Central Chile"; Turner and 102 Langmuir, 2015a) indicates substantially higher trace element abundances, which are similar in 103 character to the bulk continental crust and the average of the more mafic volcanics of Cascades 104 stratovolcanoes (Fig. 1). Thus, the chemical variability between the SSVZ and NSVZ are analogous to 105 the chemical offsets between island arcs and continental arcs.

106

107 While many prior studies have addressed the mechanisms producing along-strike compositional 108 variability of the SVZ arc-front volcanoes (Lopez-Escobar et al., 1977; Hickey et al., 1986; Hildreth 109 and Moorbath, 1988; Tormey et al., 1991; Jacques et al., 2014; Hickey et al., 2016), the role of 110 enrichment and heterogeneity within the asthenospheric mantle feeding the arc has received 111 relatively little attention. Such ambient mantle heterogeneity is commonly inferred from trace 112 element and isotopic compositions of mid-oceanic ridge basalts (MORBs) and ocean island basalts 113 (OIBs; Zindler and Hart, 1986; Willbold and Stracke, 2010). In OIBs, this variability can be accounted 114 for by a handful of isotopically distinct end-member components, such as the "enriched mantle" 115 end-members (EM1, EM2; e.g. Zindler and Hart, 1986), which are commonly modelled as the 116 products of recycled lithospheric mantle, sediment, and ancient oceanic/continental crust (c.f. 117 Willbold and Stracke, 2010). There is also evidence for ambient mantle heterogeneity in arcs, 118 despite additional tectonic complexity. For example, a global compilation of samples from rear-arc 119 provinces filtered to minimize slab contributions (Turner and Langmuir, 2015b) form a linear array,

120	extending to EM1-type OIB compositions, which are significantly more isotopically enriched (higher
121	⁸⁷ Sr/ ⁸⁶ Sr and lower ¹⁴³ Nd/ ¹⁴⁴ Nd) than the average Depleted MORB Mantle (DMM; Fig. 3a). Models
122	of corner flow find that rear-arc mantle domains are generally transported towards the arc-front.
123	Therefore, this rear-arc data may indicate that ambient sub-arc mantle compositions also vary
124	substantially (e.g. Woodhead et al., 2012). The Andean Southern Volcanic Zone (SVZ) rear arc
125	exemplifies this enriched ambient mantle signature (Kay et al., 2013; Jacques et al. 2013, 2014;
126	Søager et al., 2015a; Turner et al. 2017), with more enriched rear-arc ⁸⁷ Sr/ ⁸⁶ Sr and ¹⁴³ Nd/ ¹⁴⁴ Nd
127	values than any other arc globally (Fig. 3a). A major goal of this work is thus to assess the relative
128	influence of ambient mantle heterogeneity in a targeted region of the NSVZ.
129	
130	This study investigates the petrogenesis of new mafic lava samples from the volcanoes Don
131	Casimiro and Maipo within the Diamante-Maipo Caldera complex (34°S, Fig. 2b), to better
132	determine the geochemical character of NSVZ parental magmas. The lavas from Don Casimiro and
133	the basal portion of Maipo are among the most primitive ever sampled within the NSVZ (Fig. 4a).
134	This may be linked to their eruption immediately after the catastrophic formation (~150ka) of the
135	Diamante Caldera (Orozco et al., 2015), prior to the re-organization and maturation of crustal
136	magma reservoirs. These primitive NSVZ magma compositions allow us to characterise and explore
137	the origins of magmatic enrichment within a thickened continental subduction zone.
138	
139	This study also incorporates samples from seven Argentinean monogenetic cones, located up to
140	350 km behind the NSVZ arc front. This rear-arc volcanism initiated in the earliest Miocene,

141 associated with a period of shallow slab subduction (Kay and Copeland, 2006). Steepening of the

slab towards the present-day dip of 33° (Tassara and Echuarren, 2012) started at 5 Ma (Kay et al.,

143 2005). The samples included in this study are from monogenetic cones that erupted within the last

144 1 Myr, after the cessation of slab steepening. The post-Miocene rear-arc volcanism may be due to
145 the influx of hot asthenosphere during slab steepening from a Middle-Late Miocene flat slab regime
146 (Ramos and Folguera, 2011), possibly in a mantle plume-like configuration (Burd et al., 2014).
147 Regardless of the mode of magmagenesis, these samples provide additional constraints on the
148 composition of the ambient sub-arc mantle beneath the SVZ, and enable examination of slab
149 additions beyond the arc-front.

150

151 2. Samples and methods

152 Of the 15 arc-front samples studied (Fig. 4a-b), six represent the basal sequence of Maipo Volcano, 153 the active stratovolcano at the centre of the Diamante-Maipo Caldera Complex (Fig. 2b). Nine were 154 collected from Don Casimiro Volcano, a small eroded stratovolcano located ~10 km SW of Maipo with activity restricted to preglacial times (Charrier, 1979). ⁴⁰Ar/³⁹Ar dates indicate that early 155 156 activity at these centres was contemporaneous (Orozco et al., 2015). All lavas contain ubiquitous 157 olivine and clinopyroxene phenocrysts within a glassy groundmass. The more evolved samples also 158 contain plagioclase, oxides, and orthopyroxene (details in SIA2). The rear-arc samples are from 159 small monogenetic scoria cones (Fig. 2a). Olivine, clinopyroxene, and oxide phenocrysts dominate, 160 with microcrysts of plagioclase and alkali feldspar, and minor apatite. Whole-rock major-element 161 and trace-element analyses were conducted on all samples, with ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd data and 162 olivine major and trace analyses also collected for a subset (methods in SIA3).

163

164 **3**. Results

The new Don Casimiro-Maipo samples are primitive calc-alkaline basaltic andesites to andesites and
 the rear-arc samples are primitive basalts-trachybasalts (Fig. 4a). Olivine phenocryst compositions

range from Fo_{75.7}-Fo_{83.6} at the arc-front, and Fo_{80.6}-Fo_{90.1} in the rear-arc (Appendix 3; Fig. SB1a). The
olivine compositions of the three analysed arc-front samples and most rear-arc samples lie below
the olivine – whole rock equilibrium line, indicating that minor olivine accumulation has occurred
(up to 7 wt. %; Fig. SB1; Hickey et al., 2016).

171

The whole-rock Mg#s of Don Casimiro-Maipo samples mostly range from 60-70, indicating that these samples have undergone only minor crystal fractionation (Fig. 4a). There is little variation in SiO₂ (55.3-57.9 wt.%) within the sample set. Previous studies within the Diamante-Maipo Caldera (Hickey et al., 1986; Futa and Stern, 1988, Sruoga, 2005; Holm et al., 2014) have primarily sampled volcanics with significant europium anomalies (Eu*= [Sm_N x Gd_N]^{0.5}; Fig. 4b), indicating that samples have undergone fractionation of plagioclase or mixing between primitive and evolved magmas (e.g. Turner and Langmuir, 2015). In contrast, our higher Mg# samples have Eu/Eu*>0.9 (Fig. 4b).

179

180 Like most arc magmas, the mafic Don Casimiro-Maipo lavas and rear-arc scoria have high 181 abundances of Cs, Rb, Ba, U, Th, Pb, and Sr relative to other elements of similar incompatibility 182 during mantle melting, while Nb and Ta are relatively depleted (Fig. 1). The highly incompatible 183 trace element abundances of the Don Casimiro-Maipo arc-front samples are elevated relative to 184 typical mafic to intermediate samples from oceanic arcs or the SSVZ. Abundances in the rear-arc 185 samples are generally even higher than the arc front. As noted by Holm et al. (2016), the rear-arc 186 samples also have relative depletions in the high field strength elements Zr and Hf. The magnitude 187 of this depletion is on the upper end of what is observed within the SVZ arc front, with Hf/Sm=~0.6. 188 The Don Casimiro-Maipo samples, however, have smaller relative Zr-Hf depletions than any other 189 primitive samples from the SVZ arc-front, with Hf/Sm=~0.8 (Fig. SB2). As noted by previous studies

(e.g. Hildreth and Moorbath, 1988), Don Casimiro-Maipo and NSVZ samples have lower Cs/Rb than
Villarrica and other SSVZ samples.

192

193	Samples from Don Casimiro-Maipo have significantly more enriched isotope ratios (higher ⁸⁷ Sr/ ⁸⁶ Sr
194	and lower ¹⁴³ Nd/ ¹⁴⁴ Nd) than Villarrica and other mafic products from SVZ arc-front centres (Fig. 3b).
195	The isotopic compositions of the most primitive Don Casimiro samples in this study generally have
196	lower ⁸⁷ Sr/ ⁸⁶ Sr and higher ¹⁴³ Nd/ ¹⁴⁴ Nd than previously reported samples from the Diamante-Maipo
197	caldera (Fig. 3b). The Maipo samples from this study lie at higher ⁸⁷ Sr/ ⁸⁶ Sr and lower ¹⁴³ Nd/ ¹⁴⁴ Nd
198	than Don Casimiro Samples. The ⁸⁷ Sr/ ⁸⁶ Sr and ¹⁴³ Nd/ ¹⁴⁴ Nd of the rear-arc centres in this study fall
199	between the literature SVZ data and Don Casimiro-Maipo.

200

201 4. Discussion

202 While the gradient of northerly-increasing crustal thickness in the SVZ theoretically provides the 203 ideal setting to understand the relationship between primary magma compositions and crustal 204 thickness, the scarcity of mafic lavas in the NSVZ has made prior assessment of the origin of 205 geochemical variation difficult. The volcanological setting of early Don Casimiro-Maipo lavas 206 allowed the magmas to rapidly ascend through the thick crust of the NSVZ and avoid extensive 207 crystal fractionation. Samples from this unique geological setting can thus be used to improve our 208 understanding of the relative contributions from crustal processing, mantle melting, slab fluxes, 209 subduction erosion, and ambient mantle heterogeneity to the production of compositionally 210 enriched magmas erupted from volcanoes in continental arcs.

212 4.1. Crustal processing at Don Casimiro-Maipo

213 When lavas from throughout the Diamante-Maipo caldera complex are considered, there is a clear 214 role for crustal assimilation or mixing during fractional crystallization (AFC). ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd values correlate with indices of fractionation (Fig. SB3), and the assimilation of known 215 216 SVZ basement compositions can recreate the variation in trace element and isotope ratios between 217 the most primitive and most evolved samples (Fig. 4c-d). 218 219 Hildreth and Moorbath (1988) suggested that mantle-derived magmas are processed in a melting, 220 assimilation, storage, and homogenization (MASH) zone in the lower crust. In their model, mantle-221 derived magmas throughout the SVZ initially have similar compositions, and the enriched 222 "baseline" composition of NSVZ magmas results from increased MASH processing due to the 223 thicker crust. Thus, the isotopic compositions of even the most primitive NSVZ volcanics would be 224 the result of mixing between SSVZ-like mantle melts and some crustal basement component. 225 226 It is not possible to conclusively determine whether the most primitive Don Casimiro-Maipo 227 samples are entirely free of crustal contamination. It is possible, however, to assess the plausibility 228 of a scenario in which the notable compositional differences between the more primitive magmas 229 of the SSVZ and NSVZ are the result of a MASH-like process. Villarrica volcano has a similar trace 230 element and isotopic composition to the other volcanoes of the SSVZ (Fig. 1, Fig. 3b), and has 231 erupted some of the most primitive magmas. For simplicity, an average composition of mafic 232 samples from Villarrica is used to represent the composition of SSVZ volcanics. 233

Crustal assimilation and mixing likely affect the evolved magmas of the Diamante Caldera (Sruoga
et al., 2005). Mixing between the primitive Don Casimiro-Maipo compositions and a set of sampled

236 crustal lithologies, for example, reproduces the chemical variability observed within the Diamante 237 Caldera (Fig. 4c-d). While it is plausible that the evolved Diamante Caldera lavas can be related to 238 the primitive Don Casimiro-Maipo lavas by a combination of assimilation and fractional 239 crystallization, mixing between these same crustal rocks and primitive Villarrica samples does not 240 reproduce the Don Casimiro-Maipo compositions (Fig. 4c-d). These assimilants produce rapidly 241 decreasing K/Rb ratios prior to producing adequate enrichment in ⁸⁷Sr/⁸⁶Sr, and none reach high 242 enough Rb/Y. For the MASH model to explain the chemical differences between the NSVZ and SSVZ 243 a lower-crustal assimilant distinct from that producing geochemical variability within the Diamante-244 Maipo caldera must be incorporated into Don Casimiro-Maipo primary magmas.

245

246 To test whether any plausible Andean crustal assimilant may be capable of bridging the 247 compositional gap between Don Casimiro-Maipo and the SSVZ, the compositions of 348 basement 248 outcrops and basement xenoliths from the SVZ and the surrounding area (22-46°S) were compiled 249 (Lucassen et al. 2001; Lucassen et al., 2004 and others; Appendix 5), and mixed with the average 250 composition of primitive Villarrica samples. Only 14% of potential assimilants produce mixed 251 compositions within ⁸⁷Sr/⁸⁶Sr± 0.0001 and ¹⁴³Nd/¹⁴⁴Nd± 0.00005 of the primitive Don Casimiro-252 Maipo average (Fig. 5a). Of these isotopically plausible mixed compositions, only 35% have Eu 253 anomalies within the range of the primitive Don Casimiro-Maipo samples (Eu/Eu*>0.9). None of 254 these mixed compositions have trace element signatures resembling Don Casimiro-Maipo (Fig. 5b). 255 Incorporating crystal fractionation alongside assimilation cannot account for these compositional 256 offsets, because the resulting compositions produce mismatches not only in overall elemental 257 abundances but also in a variety of incompatible element ratios, and substantial fractional 258 crystallization would rapidly drive down the magmatic Mg#.

260 It remains possible that the required MASH assimilant has evaded sampling within the Southern 261 Andes. Consequently, it is worth considering what compositional characteristics a hypothetical assimilant must possess to bridge the compositional gap between the southern and northern SVZ. 262 263 We assume a maximum of 20% assimilation, based on the relatively high Mg#s of primitive Don 264 Casimiro-Maipo lavas (as assimilation drives cooling, and thus crystal fractionation; DePaolo, 1981). 265 Given this, any suitable assimilant must have greater than ~450ppm Zr, ~1420ppm Sr, and 266 \sim 0.63wt% P₂O₅. It is improbable that a crustal rock will possess these characteristics, because these 267 elements typically become depleted, rather than enriched, during late stage crystal fractionation or 268 crustal melting (e.g. Turner and Langmuir, 2015a). To illustrate this point, a compilation of all 269 continental granites, diorites, syenites, and monzonites in the GEOROC database (N~3000 with 270 trace element data) was searched for suitable assimilants (considering only trace element 271 abundances). Only 15 samples possessed sufficient concentrations of Zr, Sr, and P_2O_5 . Mixtures of 272 these samples with the average primitive Villarrica composition have erratic trace element 273 patterns, testament to the complex petrological histories of these unusually enriched lithologies. 274 None recreate the observed trace element pattern of Don Casimiro-Maipo, particularly the 275 negative Nb-Ta anomaly (Fig. 5c). Thus, although crustal assimilation is near pervasive among the 276 samples of the NSVZ, and strongly influences the compositions of evolved samples in this region, 277 attributing the compositional offset between the NSVZ and SSVZ to MASH processing is inconsistent 278 with the available data from SVZ basement outcrops, as well as with fundamental considerations 279 regarding the composition of the required assimilant.

280

The geochemistry of the northern-most rear-arc samples further reinforces the conclusion that the isotopic offset between NSVZ and SSVZ samples is not a product of MASH. Rear-arc ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd data fall along the "mantle array" (Fig. 3a; Søager et al., 2013; 2015a-b; Søager and 284 Holm, 2013; Kay et al., 2013; Jacques et al., 2013; 2014; Holm et al., 2014; Turner et al. 2017), 285 which is difficult to recreate by assimilation of available basement (e.g. Fig. 5a). Additionally, rear-286 arc isotopic enrichment correlates with arc-front enrichment, despite substantial longitudinal 287 offsets (Fig. 6a-b). This is difficult to reconcile with assimilation, as accretion and eastward 288 migration of the arc-front since the Palaeozoic has produced a longitudinally fragmented basement, 289 with different basement domains striking parallel to the coast (Kay et al., 2005). Assimilation of 290 lithologies within these different basement domains would produce uncorrelated rear-arc and arc-291 front Sr and Nd isotope systematics and would be unlikely to generate the linear trend on Fig. 3a. 292 Finally, even the most primitive northern rear-arc samples, which bear olivines in equilibrium with 293 their whole-rock compositions and are nearly in equilibrium with the mantle (Fig. SB1), possess 294 enriched trace element and isotopic signatures. It is improbable that such primitive samples have 295 undergone substantial crustal overprinting of ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd.

296

297 4.2 Assessing the slab and mantle melting contributions to SVZ compositional variation 298 The goal of this section is to determine to what extent slab components and melting processes 299 contribute to the along-strike variability of SVZ volcanics. Volcanic trace-element abundances vary 300 as a function of the extent of mantle melting (F) and residual mantle mineralogy. Highly 301 incompatible elements, which partition preferentially into the melt phase, may have higher 302 concentrations in the NSVZ due to lower F, rather than different mantle source abundances 303 (Tormey et al., 1991). Lower F in the NSVZ may result from the thick northern lithosphere impinging 304 on the mantle wedge (Turner et al., 2016). This would be consistent with the finding of lower F 305 values in volcanic arcs with thicker crust globally (Plank and Langmuir, 1988; Turner and Langmuir, 306 2015b).

308 In addition to variability in F, the trace element compositions of primary arc magmas reflect the 309 transportation of slab materials into the mantle wedge. To assess whether the compositions of 310 both Villarrica and Don Casimiro-Maipo can be produced from a common ambient mantle 311 composition with variable slab components and F, it is useful to consider the maximum solution 312 space generated by these parameters alone. For these calculations, we assume slab melting, as the 313 slab top temperatures for Central Chile (784°C, D80 model of Syracuse et al., 2010) lie above the 314 wet pelite and AOC solidi (Hermann and Spandler, 2007; Carter et al. 2015). Furthermore, the 315 Syracuse et al. (2010) D80 values may represent minimum temperatures, given recent evidence for 316 shear heating and shallower depths of slab-mantle coupling (Penniston-Dorland et al., 2015; 317 England, 2018). Slab melt compositions were calculated by mixing the composition of subducted 318 sediment (ODP1232; Turner et al., 2017) and MORB (Gale et al., 2013) in various proportions, 319 following by melting using partition coefficients consistent with experimental values for slab 320 melting at temperatures between 800-900 °C (SIA4.1; Kessel et al., 2005; Hermann and Rubatto, 321 2009; Skora and Blundy, 2010). These slab melts were then mixed with an estimate of the depleted 322 MORB mantle composition (DMM; Workman and Hart, 2005). The composition of mantle melts 323 derived from this mixed composition were calculated using experimentally-derived hydrous 324 partition coefficients and melt reaction coefficients (model details in SIA4.1).

325

The resulting solution space is shown in Fig. 7a-b. The red area on Fig. 7a depicts the solutions made available by varying the proportions of slab components and melt extent. F=0.04 was chosen as the lowest plausible melt extent because F values <0.04 tend to produce alkali-basalt or silica undersaturated major element compositions (e.g. Baasner et al. 2016). Villarrica samples fall within this solution space (as do most SSVZ samples), and a model fit within this space (purple diamond) reproduces the entire suite of Villarrica incompatible trace elements (Fig. 8). While this model fit does not represent a unique solution for Villarrica, it does demonstrate that a feasible quantitative solution exists that is consistent with available experimental constraints. In contrast, there is no solution for Don Casimiro-Maipo. Increasing the amount of sediment and AOC contributions to the depleted mantle source can recreate the high La/Sm ratios, but only at Sr/Nd values that are far too high (Fig. 7a). While reducing F shifts the melt composition to higher La/Sm, even at F=0.04 there is no possible match to the Don Casimiro-Maipo volcanics.

338

The composition of the slab components also varies as a function of slab temperature (Hermann and Rubatto, 2009). There are systematic changes in the slab age and depth beneath the SVZ arc front, with corresponding variations in the slab thermal parameter, Φ (Φ =slab age x convergence rate x sin(dip angle)). Therefore, one might expect hotter slab temperatures in the NSVZ. However, while the magnitude of geochemical variation in the SVZ comprises much of the compositional range seen among arcs globally, differences in Φ between the SSVZ and NSVZ are relatively minor on the global scale (Turner et al., 2016).

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347 Certain aspects of the trace element differences between Don Casimiro-Maipo and Villarrica 348 suggest subtle slab temperature variations are indeed present. Compared to Villarrica, Don 349 Casimiro-Maipo has low Cs/Rb and high Hf/Sm (Fig. 1). These element pairs are not substantially 350 fractionated from each other by mantle melting, so likely reflect differences in the mantle source 351 composition. For these element ratios, it is plausible that observed mantle source variations are 352 caused by higher slab temperature in the NSVZ. Cs is highly mobile in low temperature aqueous 353 fluids, and may have been disproportionally lost during early slab dehydration (Savov et al., 2007; 354 Spandler et al., 2007). A hotter slab temperature can also destabilize zircon during slab melting, 355 producing the less pronounced Zr-Hf depletions at Don Casimiro-Maipo (e.g. Hirai et al., 2018). The blue field on Fig. 7a shows the expansion of the available solution space for slab temperatures
>900°C, as calculated using the maximum measured experimental mobility of light rare earth
elements (LREE) in slab melts. While the solution space shifts toward higher La/Sm at a given Sr/Nd,
the mafic NSVZ volcanics still plot well outside of the realm of possible solutions. Despite evidence
for differences in SVZ slab temperature, including a slab temperature parameter alongside variable
F and varying slab proportions only increases the available solution space to the Red+Blue fields on
fig. 7a, which clearly does not envelop the data from Don Casimiro-Maipo.

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The same conclusion can be drawn from ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd (Fig. 7b). The isotopic solution space is simplified by the fact that these isotope ratios are not fractionated by melting. The pink field on Fig. 7b shows the solution space produced by variation in slab temperature and the proportions of slab melts. Again, the Don Casimiro-Maipo data falls well outside of the available solution space, because slab contributions predominantly drive wedge compositions to higher ⁸⁷Sr/⁸⁶Sr at near constant or increasing ¹⁴³Nd/¹⁴⁴Nd.

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372

4.3 Is there geochemical evidence for subduction erosion?

373 SVZ rear-arc volcanics form a linear array between MORB-like compositions and EM1-type OIBs in ⁸⁷Sr/⁸⁶Sr vs. ¹⁴³Nd/¹⁴⁴Nd space (Kay et al., 2013; Søager et al., 2015a; Fig. 3a), and both rear-arc and 374 375 arc-front samples are characterized by similar along-strike variability in these isotopes (Fig. 6a-b). If 376 the isotopic offset between the mafic NSVZ and SSVZ samples is neither a product of crustal 377 assimilation nor variation in the slab component, some EM1-like contaminant must be introduced 378 to the mantle source of the SVZ. The origins of EM1-type OIBs are debated, with suggestions 379 including deep recycling of lower continental crust (LCC), pelagic sediment and oceanic crust, or 380 metasomatized subcontinental lithospheric mantle (Willbold and Stracke, 2010). The first

381 suggestion is particularly apt in the SVZ, as it has been proposed that the NSVZ endured extensive 382 subduction erosion (the removal of upper plate material via abrasion and plucking by the lower 383 plate) throughout the Cenozoic (Stern, 1989). While the composition of the South Andean LCC is 384 not well constrained, the possible compositions of this reservoir can be inferred from the ever-385 growing dataset of regional basement compositions (Appendix 5). Although these samples were 386 collected from surface outcrops, such outcrops may be representative of deep crustal lithologies 387 due to Miocene episodes of tectonic thickening and under-thrusting (Hildreth and Moorbath, 388 1991). Thus, this database likely represents lithologies within both the upper and lower crust, and 389 can be used not only to test models of lower crustal recycling, but also recycling of the entire 390 crustal column forming the outbound belt of the South American Plate vulnerable to subduction 391 erosion, including the upper crust (as suggested by Holm et al., 2014; 2016).

392

393 To assess the subduction erosion hypothesis, basement compositions (Appendix 5) were mixed 394 with the estimated composition of the Villarrica mantle source. Only 11% of the mixed compositions pass within ⁸⁷Sr/⁸⁶Sr±0.0001 and ¹⁴³Nd/¹⁴⁴Nd±0.00005 of the most primitive Don 395 396 Casimiro-Maipo samples (Fig. 9a). Only 15% of the isotopically plausible mixed compositions have 397 Eu anomalies within the range of the primitive Don Casimiro-Maipo samples (Eu/Eu*>0.9), none of 398 which recreate the concentrations of Y, Gd, Sm, Sr, Ba and ⁸⁷Sr/⁸⁶Sr ratios (Fig. 9b-c). To conclude, 399 no mixed compositions with adequate trace element data falls within the range of the Don Casimiro-Maipo for ⁸⁷Sr/⁸⁶Sr, ¹⁴³Nd/¹⁴⁴Nd, and trace element abundances. 400

401

While some studies in this region (e.g. Holm et al., 2014; 2016) have found that SVZ volcanic
compositions can be related by subduction erosion of sampled crustal lithologies, our preceding
analysis does not support this conclusion. This is possibly because the newly sampled Don Casimiro-

405 Maipo samples are more primitive, while those in previous studies were affected by plagioclase 406 fractionation, magma mixing, or crustal assimilation, which accounts for their lower Eu/Eu* values. 407 It is possible that limited basement exposure in the SVZ means that the appropriate mixing lithology 408 has avoided sampling, but regional considerations also indicate the LCC chemical model is 409 implausible. Willbold and Stracke (2010) provide a model by which EM1-like isotopic compositions 410 are produced by mixing LCC starting materials into normal MORB mantle. However, this model 411 assumes that the LCC domains in question have been extracted and isolated from the mantle for >4 412 Ga, while the Chilenia and Cuyania terranes comprising the lower crust of the NSVZ-TSVZ are only 413 of Grenvillian age (~1Ga; Ramos, 2010). Because of this relatively young age, the typical basement of the Southern Andes has ¹⁴³Nd/¹⁴⁴Nd values that are too high (relative to ⁸⁷Sr/⁸⁶Sr) to serve as an 414 415 appropriate end-member (see mixed compositions in Fig. 9a). When considering the full array of 416 available trace element abundances and isotope values, the subduction erosion hypothesis does 417 not appear to be consistent with the geochemical systematics, possibly indicating that the extent of 418 subduction erosion has been over-estimated for this region. For example, Stern (1989) estimated 419 erosion rates based on the assumption that the distance between the trench and the arc front 420 remained constant throughout the Miocene, while recent geophysical models find that arc-front 421 migration relative to trench location might be a common process (e.g. Karlstrom et al., 2014). 422 Alternatively, subduction erosion may have affected the Andean margin in the past, but the eroded 423 material has since been flushed from the mantle wedge by corner flow.

424

425 Rear-arc geochemical variability is also inconsistent with the subduction erosion hypothesis. Rear-426 arc isotopic enrichment persists at distances exceeding 600 km from the trench; it seems unlikely 427 that material eroded at the trench could spread hundreds of kilometres in the opposite direction of 428 corner flow. It may instead be carried down with the subducting slab and released alongside other

429	slab inputs, but in this case the enriched signature would co-vary with other slab components.
430	Instead, at 550 km from the trench rear-arc monogenetic samples have Ce/Pb values that range
431	from 5 (similar to the arc front) to >20 (similar to MORBs; Fig. 10b), while rear-arc 143 Nd/ 144 Nd
432	ratios are consistently enriched relative to DMM (Fig. 3a). Additionally, within a given latitude
433	bracket rear-arc ¹⁴³ Nd/ ¹⁴⁴ Nd ratios have limited variability, and do not correlate significantly with
434	Ce/Pb or other indices of slab addition. Finally, isotopic enrichment in both the rear arc and arc
435	front increases in a reasonably symmetrical, coherent fashion on either side of a pronounced
436	minima at 39° S (Fig. 6a-b; Turner et al., 2017). Although it has been suggested that subduction
437	erosion increases northwards (Stern, 1989), this phenomenon cannot account for the increase in
438	enrichment south of Villarrica. The spatial systematics of rear-arc compositions are therefore also
439	inconsistent with subduction erosion.
440	
441 442	4.4 EM1-type enrichment from a sub-continental lithospheric mantle source An alternative source for the enriched EM1-like mantle signature of the NSVZ is Metasomatised
443	Subcontinental Lithospheric Mantle (M-SCLM). The infiltration of low degree mantle melts
444	(F~0.005) into the base of the lithospheric mantle produces enriched incompatible element
445	concentrations, which rapidly evolve EM1-like isotopic compositions (McKenzie and O'Nions, 1995;
446	Turner et al., 2017; Fig. 11). Additionally, M-SCLM provides a more homogenous end-member
447	composition than crustally-derived materials (Rogers and Hawkesworth, 1989; Turner et al, 2017).
448	This material may be stored for long periods in the lithospheric mantle until it is returned to the

451 Field evidence supports the hypothesis that the SCLM surrounding the SVZ has EM1-like isotopic

452 affinities. Three geographically separated suites of isotopically enriched igneous rocks across South

453 America have been interpreted as direct melts of the enriched SCLM (see Fig. 2a):

Alkaline igneous rocks erupted near the edges of the Rio Apa-Luis Alves and Sao Franscisco
 Cratons (Gibson et al., 1995; 2005; Carlson et al., 1996).

456 2) Mesozoic metabasites from the Southern Rift (Lucassen et al., 2002).

457 3) Carboniferous granitoids from the Santo Domingo Complex of the Coastal Batholith (Parada
458 et al., 1999).

The ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd of samples from these regions plot as a linear extension of the SVZ 459 460 rear-arc array (Fig. 11) and cannot be accounted for by addition of variable slab components or 461 assimilation of available basement. The remarkable alignment of these lithologies in isotopic space 462 despite being geographically separated by hundreds to thousands of kilometres suggests that M-463 SCLM east and north of the SVZ has relatively homogeneous EM1 affinities. The isotopic trajectory 464 of these SCLM melts is consistent with the expected isotopic evolution of SCLM enriched by low 465 degree mantle melts (Turner et al., 2017; Fig. 11). In contrast, the field produced by addition of 466 sampled basement lithologies to the mantle has higher ⁸⁷Sr/⁸⁶Sr at a given ¹⁴³Nd/¹⁴⁴Nd than the 467 array of SCLM-melts and rear-arc volcanics. Additionally, the isotopic variability induced by 468 basement contamination is significantly broader than the narrow trajectory of rear-arc and M-SCLM 469 melts (grey field, Fig. 11).

470

The full trace element pattern of such low-degree, relatively high-pressure mantle melts is difficult
to assess, as no appropriate partitioning experiment has been carried out. Instead, a compositional
proxy can be calculated by inverting for the mantle source composition of Gough Island EM1-type
OIB volcanics (Turner et al., 2017), where enrichment has been linked to deep recycling of SCLM

475 (Gibson et al., 2005). Trace element and isotopic ratios of the inverted source composition were 476 subsequently adjusted within the limits of EM1-like ocean island basalts (Stracke et al. 2003; 477 Willbold and Stracke, 2010) to produce a generic EM1 source (SIA4.2). By including EM1-like mantle 478 heterogeneity as an additional parameter in the forward model, the solution spaces on Fig. 7a-b 479 expands to include the green fields, which finally overlap the primitive Don Casimiro-Maipo trace 480 element concentrations. Furthermore, a model fit was identified that is consistent with the full Don 481 Casimiro-Maipo trace element and isotopic composition (Fig. 7-8). As with the model fit for 482 Villarrica, the Don Casimiro-Maipo model does not represent a unique solution, but is a 483 demonstration of the model's quantitative viability under combined evidence from field and 484 experimental constraints.

485

486 The addition of M-SCLM to the mantle by erosion or delamination at the base of continental 487 cratons behind the volcanic arc also provides a more consistent explanation for the spatial 488 variability of both rear-arc and arc-front SVZ volcanic compositions. Turner et al. (2017) proposed 489 two explanations for the gradual decline in isotopic enrichment towards a distinct minimum at 39°S 490 (Fig. 6a-b). Firstly, they note the presence of a Permian terrane suture at 39° S (Rapalini et al., 491 2010). It is possible that M-SCLM was lost during rifting or subsequent collision between cratonic 492 blocks (Fig. 12a). Secondly, a slab tear may be present at 200km depth on the lower plate at ~39° S 493 (Pesicek et al., 2012). If M-SCLM is entrained equally throughout the SVZ, upwelling of depleted 494 Pacific mantle (which has not interacted with M-SCLM) through this slab window could dilute the 495 EM1 signature. The upwelling, depleted mantle would then spread northwards and southwards, 496 perhaps aided by toroidal flow around the slab tear (Zandt and Humphreys, 2008), resulting in a 497 gradual northward and southward increase in enrichment (Fig. 12b). This could also potentially 498 explain the magnetotellurically imaged "plume" in the rear-arc (Burd et al., 2014). Geochemical

- signatures associated with upwelling through a slab window in the Kula Volcanic Field (W. Anatolia,
- 500

) Turkey) are traced over >250km (Klaver et al., 2016), similar to the distances observed here.

501

502 4.5 The relationship between trace element heterogeneity and mantle mineralogy 503 If the geochemical differences between the NSVZ and SSVZ serve as an analog for the differences 504 between thin and thick crusted arcs, then ambient mantle enrichment may be a common feature of 505 continental arcs. Determining whether EM1-like signatures within the SVZ are characterized by a 506 distinct mineralogy (peridotite vs. pyroxenite) thus has potential implications for melting reactions 507 within these enriched mantle domains. It has been suggested that SVZ rear-arc EM1 signatures are 508 held within pyroxenitic domains, and that a southward decrease in rear-arc olivine Mn/Fe values 509 results from the presence of pyroxenite source lithologies beneath the southern rear-arc (Søager et al., 2015b; Brandt et al., 2017). However, ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd isotopes become *less* EM-1-like 510 511 as Mn/Fe ratios decrease, which is difficult to reconcile with this interpretation (Fig. 13). Moreover, 512 the samples presented in this study have some of the most enriched isotopic signatures in the rear 513 arc, yet have olivine compositions traditionally associated with a peridotite source (Sobolev et al., 514 2007; Fig. SB4). This suggests that EM1 isotopic signatures are unrelated, or even anti-correlated to 515 the amount of pyroxenite. Thus, although large pyroxenitic domains may be present in certain 516 areas of the rear arc, the distribution of this material is an unlikely candidate to explain isotopic and 517 trace element trends on the scale of the SVZ as a whole. Crucially, rear-arc isotopic ratios can be 518 reproduced with ~2% addition of the inferred SCLM melts (e.g. Gibson et al. 1995; 2005) to the 519 depleted mantle, which is unlikely to have a detectable influence on erupted olivine phenocryst 520 chemistry. It is also worth considering whether olivine compositional classifications, which have 521 been calibrated primarily for anhydrous ocean island basalts (e.g. Sobelov et al. 2007), can be 522 robustly applied to hydrous systems (Wang et al., 2016). The detailed relationship between erupted

- olivine compositions and mantle mineralogy in the SVZ, as well as the relationship between mantle
 mineralogy and general indices of mantle enrichment remains an exciting area of ongoing research.
- 526 4.6 Distribution of the slab components within the mantle

The geochemical characteristics of rear-arc centres not only reinforce the conclusions drawn at the arc-front regarding spatial trends in EM1-like enrichment, they also provide novel constraints on the spatial distribution of slab components beyond the arc front. Slab melts have high Th/Nb and Th/La, and low Ce/Pb, so these ratios serve as proxies for the extent to which slab components have infiltrated the mantle source of each monogenetic cone (Johnson and Plank, 1999; Plank, 2005). Remarkably, while slab-sensitive ratios generally decline with increasing trench distance, some cones retain elevated Th/La and Th/Nb and low Ce/Pb at nearly 600km from the trench (Fig.

534 10b).

535

536 Highly incompatible trace element ratios of rear-arc samples may also indicate the provenance of 537 rear-arc slab components. Thermal models predict that slab-surface temperatures at SVZ rear-arc 538 depths will be ~250° C higher than those under the arc front (Syracuse et al., 2010). Such high slab 539 temperatures should lead to a relative reduction of zircon stability in the slab, whereas the rear-arc 540 samples have Zr-Hf anomalies similar to those observed for the SSVZ arc-front (Hirai et al., 2018; 541 Fig. SB2). This indicates that the slab component reaching the rear-arc mantle source may have 542 been extracted from the slab at temperatures similar to the SSVZ arc-front slab component. 543 Significant fractionation of other slab-derived elements would also be expected if slab components 544 were extracted at temperatures ~250° C higher than at the arc-front. Th/La in slab melts may 545 increase by ~25%, and Th/Nb and U/Nb by nearly an order of magnitude (Hermann and Rubatto, 546 2009), which would result in rear-arc and arc-front lavas with distinctly different mixing trajectories for these trace elements. Instead, rear-arc and arc-front samples both lie on mixing lines between
the arc-front slab melt composition and a mantle with varying amounts of EM1-like enrichment
(Fig. 10a). These observations suggest that very little fractionation of these elements has occurred,
which is most simply explained by similar slab top temperatures at the source of arc-front and reararc slab inputs.

552

553 The observation that rear-arc mantle sources 600 km from the trench appear to have been 554 infiltrated by a slab component that is similar in composition to the arc-front slab component is 555 surprising. It is conceivable that the rear-arc slab signatures represent remnant metasomatism of 556 the mantle wedge from the period of Miocene shallow subduction. However, the samples of this 557 study were probably erupted between 0.01-0.7 Ma (Folguera et al., 2009), 4-5 million years after 558 the slab began to steepen. As the wedge will have "turned over" ~330km in this time, it is likely that 559 Miocene slab inputs were effectively flushed from the asthenospheric sources of the rear-arc cones 560 of this study. A more plausible explanation for both the abundance and composition of the slab 561 fluxes in the rear-arc is that the slab components added to the sources of subduction volcanics are 562 not derived from the slab directly beneath the sampled cone. Geophysical models demonstrate 563 that mantle flow and compaction pressure gradients can affect the flow paths of materials 564 separated from the slab (e.g. Cagnioncle et al., 2007; Wilson et al., 2014). Thus, the mantle source 565 at the arc front may be influenced by slab components that have been extracted from the slab 566 beyond the arc front, and the rear-arc mantle source may be supplied from small quantities of slab 567 components transported in the opposite direction in regions of low porosity (Cerpa et al., 2017; Fig. 568 14b).

570	It also is possible that slab materials could be transported non-vertically by "mélange diapirs".
571	However, recent experimental work (Cruz-Uribe et al., 2018) found that melts of such diapirs will
572	likely have >50 wt. % SiO ₂ , >18 wt. % Al ₂ O ₃ , and <8 wt. % MgO, while the most primitive SVZ rear-
573	arc basalts have >10 wt. % MgO, <48 wt. % SiO ₂ , and <15 wt. % Al ₂ O ₃ , as is typical of arc alkali
574	basalts generated by low degrees of melting from a lherzolite source (e.g. Baasner et al., 2016).
575	Additionally, the experimental mélange melts have positive Zr-Hf anomalies while the SVZ rear-arc
576	basalts have negative Zr-Hf anomalies (Fig. 1). We also find the possibility of a mélange diapir
577	ascending far above the slab surface without melting to be fundamentally implausible, given the
578	low wet solidus temperatures of sediments and altered ocean crust (Herman and Rubatto, 2009;
579	Carter et al., 2015). Thus, the geochemical systematics of SVZ rear-arc volcanics appear to be most
580	consistent with a strong role for slab melting followed by advective transport, compaction
581	channelling, down-dragging, and generally non-vertical transport of slab liquids through the mantle
582	wedge, consistent with recent numerical models of two-phase flow in the mantle wedge
583	(Cagnioncle et al., 2007; Wilson et al., 2014; Cerpa et al., 2017).
584	
584 585	4.6 Implications for crustal growth and the thermal structure of the arc crust
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585 586 587	The extent to which the compositions of continental arc volcanics are inherited from the mantle rather than overprinted in the crust has important implications for models of crustal growth.
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585 586 587 588 589	The extent to which the compositions of continental arc volcanics are inherited from the mantle rather than overprinted in the crust has important implications for models of crustal growth. Abundances of highly incompatible trace elements in oceanic arcs are much lower than those of the bulk continental crust (Fig. 1). If continental growth is driven primarily by the accretion of island
585 586 587 588 589 590	The extent to which the compositions of continental arc volcanics are inherited from the mantle rather than overprinted in the crust has important implications for models of crustal growth. Abundances of highly incompatible trace elements in oceanic arcs are much lower than those of the bulk continental crust (Fig. 1). If continental growth is driven primarily by the accretion of island arcs, which have incompatible element abundances that are often an order of magnitude lower

contributed a substantial portion of the bulk material to the continental crust (Cao et al., 2017), and
continental arc magmas have trace element patterns that more closely resemble the continents
(Fig. 1). If this trace element enrichment is inherited from the mantle, then a substantially smaller
amount of material must be returned to the mantle to account for the continental mass balance,
possibly closer to 50% (Sisson and Kelemen, 2018). The differences between these two
interpretations, in turn, have clear implications for the generation of large-scale mantle
heterogeneity.

601

602 Evaluating the role of parental magma enrichment versus crustal overprinting is also critical to 603 understanding the petrogenesis of differentiated arc magmas. If one assumes that an evolved arc 604 magma with an "enriched" isotopic composition originated from a "depleted" mantle source similar 605 to MORB, or that the mantle source has a uniform isotopic composition, they will reasonably infer 606 that the enriched magma is composed largely of recycled crustal material and that isotopic 607 variability among magmas is mostly driven by crustal recycling (e.g. Davidson et al., 1990; Luccasen 608 et al., 2006). If instead one assumes that the mantle source is enriched and heterogeneous, then 609 the inferred proportion of the crustal component in the erupted magma will be smaller. For 610 example if the mantle source is assumed to be isotopically similar to typical MORBs, a mixing 611 exercise such as that shown in Fig. 5 indicates that an average of 40% crustal assimilation is necessary to account for the erupted ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd values at Don Casimiro-Maipo. 612 613 Instead, by invoking an enriched mantle source, we have found that these samples are potentially 614 free of crustal contamination. The magnitude and variability of crustal assimilation and crustal 615 melting have clear implications for our understanding of the thermal structures of volcanic arcs 616 (e.g. Annen and Blundy, 2006).

618 5. Conclusions

619 The volcanic products of thick-crusted continental arcs have substantially higher abundances of 620 incompatible trace elements, and more "enriched" isotopic signatures than island arcs. The origin 621 of these enriched signatures has profound implications for the formation of the continental crust 622 and the petrogenesis of evolved magmas. The Andean Southern Volcanic Zone (SVZ) is an ideal 623 natural laboratory for investigating the role of crustal thickness in magma petrogenesis, because 624 along-strike trends in SVZ crustal thickness and magma chemistry are analogous to the salient 625 differences between island arcs and continental arcs. While primitive basaltic lavas are common in 626 the Southern SVZ, mafic magmas are scarce in the Northern SVZ, which has made it difficult to 627 distinguish between crustal and mantle signatures. The trace element and isotopic measurements 628 of new Northern SVZ mafic lava samples from Don Casimiro-Maipo presented in this study provide 629 valuable new constraints on these problems.

630

631 While it has been suggested that the geochemical offset between the northern and southern SVZ is 632 primarily driven by the assimilation of continental crust, the addition of known SVZ basement 633 lithologies to a melt composition characteristic of the Southern SVZ fails to recreate the enriched 634 isotopic and trace element signatures of these new Don Casimiro-Maipo samples. Most mixed 635 compositions have ⁸⁷Sr/⁸⁶Sr values that are too high at a given ¹⁴³Nd/¹⁴⁴Nd, and none have 636 appropriate trace element patterns. Moreover, no plutonic lithology sampled *globally* is sufficiently 637 enriched in the required suite of trace elements, due to the fractionation of accessory phases such 638 as zircon and apatite. Assimilation of these variably enriched lithologies produces erratic trace 639 element patterns, dissimilar to those observed at Don Casimiro-Maipo. Simulating the process of 640 subduction erosion by addition of crustal lithologies to a SSVZ-like mantle source also fails to

account for northern enrichment, as the resulting melt compositions display similarly unsuitable
 isotopic systematics and erratic trace element patterns.

643

Variable melt extents arising from the northwards impingement of the upper plate on the mantle wedge likely contribute to the observed trace element variability, but cannot account for the large isotopic differences. Similarly, although trace element and isotopic differences in the slab components of the SSVZ and NSVZ may arise due to differing slab temperatures or proportions of slab components, the solution space made available by these parameters, even in combination, does not encompass the trace element or isotopic compositions of mafic Don-Casimiro Maipo samples.

651

652 Thus, the compositions of primitive Don Casimiro-Maipo lavas appear to require not only the 653 addition of slab melts to the mantle wedge and low degrees of mantle melting, but also an enriched 654 northern SVZ mantle source prior to slab metasomatism. This is evident from the fact that no other 655 mechanism can successfully reproduce the compositions of the northern SVZ volcanics. 656 Additionally, rear-arc magmas filtered to only include samples with minimal slab contributions 657 extend along the "mantle array" towards compositions similar to EM1-type OIBs. Finally, the most 658 enriched rear-arc samples lie behind the northern SVZ, with a clear decrease in enrichment 659 southwards towards the SSVZ. A forward model based on an inversion for an EM1-like mantle 660 source can successfully reproduce the isotopic composition and trace element composition of Don-661 Casimiro Maipo, demonstrating that this interpretation is consistent with available experimental 662 constraints.

664 The isotopic trajectory of the rear-arc lavas is co-linear with other South American lavas that are 665 thought to be melts of metasomatized sub-continental lithospheric mantle (M-SCLM). Thus, a 666 plausible origin of the enriched mantle source within the SVZ is the addition of M-SCLM material to the mantle wedge by erosion or delamination from the cratonic lithosphere behind the arc. The 667 668 presence of significant isotopic enrichment >600km "upstream" of the trench is more consistent 669 with an M-SCLM origin than subduction erosion. While the systematic along and across-arc 670 geochemical variations are difficult to reconcile with assimilation or subduction erosion of 671 regionally variable basement lithologies, the composition of the M-SCLM appears to be relatively 672 homogenous across a large area of South America. Variable mantle enrichment within the SVZ may 673 result from upwelling of depleted mantle through a slab tear or the absence of M-SCLM domains 674 around a paleosuture zone.

675

Our new data from the northernmost rear-arc province show that in addition to chemical variability
induced by variable M-SCLM addition to the mantle source, some rear-arc cones have highly
variable slab inputs that are surprisingly similar in composition to the arc front. This supports recent
two-phase flow models indicating non-vertical transport of slab materials (Cagnioncle et al., 2007;
Wilson et al., 2014; Cerpa et al., 2017).

681

This results of this study imply that the enriched trace element and isotopic compositions of primitive lavas in the thick-crusted NSVZ are produced by low degree melting of an enriched ambient mantle that is metasomatized by slab melts, rather than fractionation and crustal assimilation. The similarities between the trace element enrichments of these primitive NSVZ and the bulk continental crust (Fig. 1) suggests that these mantle processes may play a major role in the origin of enriched continental compositions globally. An important avenue of future research is to

688	constrain the extent to which the processes leading to enrichment in the NSVZ are applicable to
689	thick crusted arcs elsewhere. This will require revaluation of enriched signatures in the most
690	primitive lavas of other thick crusted arcs (e.g. Cascades, Mexico, Guatemala, Colombia). If
691	similarities with the SVZ are found, they will provide valuable constraints on models of continental
692	crust generation and elemental cycling within the silicate earth. For example, if ambient mantle
693	heterogeneity contributes substantially to enriched continental compositions, continental mass
694	balances require smaller amounts of material to be returned to the mantle.
695	
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699	of the LA-ICP-MS, Jason Day for helping with analysis, and Patrick Sugden for assistance with
700	sample preparation for Sr and Nd isotopes in Leeds.
701	
702	
703	Figure Captions
704	
705	Fig. 1– Trace element Signatures. Trace elements at Don Casimiro-Maipo (this study) are highly
706	enriched compared with Villarrica and the majority of oceanic arcs, and show slight enrichment
707	over the Cascades and Central Chile (Turner et al., 2016). The trace element pattern is similar to the
708	bulk continental crust (Rudnick and Gao, 2003). Don Casimiro-Maipo shows extreme HREE
709	depletion, indicating abundant garnet in the source mantle. Villarrica shows a very similar trace

710 element signature to other SSVZ centers (Llaima, Puyehue and Osorno are shown here; Turner et al.

2016), justifying its use as the SSVZ end-member in the modelling of this study. Rear-arc samples
show distinctive arc-like signatures, and even greater enrichment than Don Casimiro-Maipo. Spider
diagrams normalized to Pyrolite, McDonough and Sun, (1995).

714

715 Fig. 2-Geographical context and sample locations. a) Map of the SVZ, with segment boundaries 716 defined by Dungan et al. (2001). The location of arc-front volcanoes with primitive samples, and the 717 rear-arc sample locations of this study and the GEOROC database are shown. The locations of 718 inferred M-SCLM melts in Fig. 11 are also shown. Basemap from GEOMAP APP. b) Sample locations 719 of Don Casimiro and Basal Maipo lavas within the Diamante-Maipo Caldera Complex. Imagery from 720 Google Earth. c) Moho depth versus projected latitude within the SVZ. Moho depth is calculated at 721 each volcanic centre south of Don Casimiro-Maipo in a) from Tassara and Echaurren (2012). 722 Projected latitude is calculated by projecting a GPS co-ordinate along the convergence vector of the 723 Nazca plate to the Chile trench (Turner et al., 2017). d) and e) Major and trace element 724 concentrations in primitive samples (Turner et al. 2016 and this study) increase northwards, 725 mirroring changes in crustal thickness. 726 727 Fig. 3- Isotopic and trace element variability within the Andean SVZ placed within a global context. 728 a) Literature SVZ rear-arc samples filtered to remove high slab inputs (Th/Nb<0.4) fall on a mantle 729 array between DMM and EM1-type ocean island basalts, greatly extending the variability observed 730 globally in rear-arc provinces (rear-arc averages from Turner et al., 2017). Fields are drawn 731 incorporating EM1 and EM2-type ocean island basalts from Stracke et al. (2003). Literature values 732 from Don Casimiro-Maipo lie at the enriched end of this field with respect to global rear-arc 733 variability. The isotopic end-members used for modelling are shown. The EM1 model isotopic 734 composition is taken from a sample from Pitcairn (57DS9; Stracke et al., 2003), which falls near the

735 enriched end of EM1-type ocean island basalts. The depleted end member, Baseline Andean 736 Mantle (BAM), was chosen to plot at the depleted end of the rear-arc array (Sample 126171 from 737 Søager et al., 2013). The Nazca Plate sediment composition is calculated from ODP1232 (Turner et 738 al., 2017). b) Don Casimiro and Maipo samples from this study show significantly more enriched 739 isotopic compositions than other SVZ arc-front centres (Turner et al., 2016). Villarrica lies at the 740 depleted end of the SVZ arc-front array. Within the Diamante-Maipo caldera, more evolved 741 samples from Don Casimiro and Maipo plot alongside literature data, following a shallower 742 trajectory than that separating Don Casimiro and Villarrica.

743

744 Fig. 4-Sample Characteristics. a) Don Casimiro-Maipo samples from this study are primitive basaltic 745 andesites. These samples significantly extend the number of primitive analyses available for this 746 centre. Circled samples are believed to have undergone olivine addition (Fig. SB1). Rear-arc samples 747 are primitive basalts-trachybasalts. b) Eu anomalies are negligible in the most primitive samples of 748 this study (>0.9), whereas the majority of literature samples show substantial anomalies. Eu/Eu* was calculated with $Eu^* = (Sm_N \times Gd_N)^{0.5}$ or with $Eu^{**} = (Sm^2 \times Tb)^{1/3}$ where no Gd data was reported. 749 750 In both cases, element concentrations are normalized to CI chondrite (McDonough and Sun, 1995). 751 c-d) Assimilation of eight SVZ basement sample into primitive Don Casimiro-Maipo samples 752 produce mixing lines that encompass the isotopic and trace element chemical diversity within the 753 Diamante Caldera (from Lucassen et al. 2001; 84-31-1, 84-31-2, 84-42-1, 84-42-11, and Lucassen et 754 al. 2004; 00-77, 00-55, 00-58 and 00-05; lithologies chosen as they best recreate the variability 755 within the Diamante-Maipo caldera). However, assimilation of the same 8 samples cannot recreate 756 the separation between Don Casimiro-Maipo and Villarrica (grey lines). Models originate from the 757 primitive average of Don Casimiro-Maipo and Villarrica samples. Error bars in c) show ±1 σ of the 758 variation among primitive samples.

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760	Fig. 5-Testing crustal assimilation models. a) Isotopic mixing lines produced by assimilation of 348
761	crustal lithologies thought to be representative of SVZ basement (compiled from GEOROC and
762	other sources) into the average composition of primitive Villarrica samples. Only a small proportion
763	pass near Don Casimiro-Maipo. b) Trace element patterns of the mixed compositions that produce
764	isotopic ratios close to Don Casimiro-Maipo (red lines in a), and have Eu/Eu*>0.9. c) Mixed
765	compositions for the small subset of the \sim 3000 plutonic rocks from GEOROC that have sufficient
766	concentrations of Zr, Sr and P_2O_5 to recreate the enrichment of these elements at Don Casimiro-
767	Maipo. Mixed compositions were calculated for the % mixing required to recreate Zr
768	concentrations. All produce erratic trace element patterns, with none recreating the distinctive Nb-
769	Ta enrichment at Don Casimiro-Maipo.
770	
770 771	Fig. 6 - Across and along-arc variation in rear-arc isotopes. a-b) Rear-arc isotopic ratios form
	Fig. 6 - Across and along-arc variation in rear-arc isotopes. a-b) Rear-arc isotopic ratios form latitudinally coherent trends, with statistically significant R ² values (p<0.005). A pronounced
771	
771 772	latitudinally coherent trends, with statistically significant R ² values (p<0.005). A pronounced
771 772 773	latitudinally coherent trends, with statistically significant R ² values (p<0.005). A pronounced minimum in 87 Sr/ 86 Sr, and a maximum in 143 Nd/ 144 Nd is observed at ~39° S, with a reasonably linear
771772773774	latitudinally coherent trends, with statistically significant R ² values (p<0.005). A pronounced minimum in 87 Sr/ 86 Sr, and a maximum in 143 Nd/ 144 Nd is observed at ~39° S, with a reasonably linear and symmetric decrease to the north and south. Rear-arc samples from this study fit within the
 771 772 773 774 775 	latitudinally coherent trends, with statistically significant R ² values (p<0.005). A pronounced minimum in ⁸⁷ Sr/ ⁸⁶ Sr, and a maximum in ¹⁴³ Nd/ ¹⁴⁴ Nd is observed at ~39° S, with a reasonably linear and symmetric decrease to the north and south. Rear-arc samples from this study fit within the regional trends. Arc-front centres lie close to the rear-arc best fit line, showing that these spatial
 771 772 773 774 775 776 	latitudinally coherent trends, with statistically significant R ² values (p<0.005). A pronounced minimum in ⁸⁷ Sr/ ⁸⁶ Sr, and a maximum in ¹⁴³ Nd/ ¹⁴⁴ Nd is observed at ~39° S, with a reasonably linear and symmetric decrease to the north and south. Rear-arc samples from this study fit within the regional trends. Arc-front centres lie close to the rear-arc best fit line, showing that these spatial trends in enrichment are also present at the arc-front. The deviation in ¹⁴³ Nd/ ¹⁴⁴ Nd between the
 771 772 773 774 775 776 777 	latitudinally coherent trends, with statistically significant R ² values (p<0.005). A pronounced minimum in ⁸⁷ Sr/ ⁸⁶ Sr, and a maximum in ¹⁴³ Nd/ ¹⁴⁴ Nd is observed at ~39° S, with a reasonably linear and symmetric decrease to the north and south. Rear-arc samples from this study fit within the regional trends. Arc-front centres lie close to the rear-arc best fit line, showing that these spatial trends in enrichment are also present at the arc-front. The deviation in ¹⁴³ Nd/ ¹⁴⁴ Nd between the rear-arc and the arc-front south of 39° S likely results from the sparse sampling of rear-arc centres,

Fig. 7 – Trace element and isotopic solution space produced by varying the composition of slab
melts, the extent of melting, and the composition of the ambient mantle. a) The red field

783 represents the maximum solution space available by the addition of varying slab melt compositions 784 (up to 2% sediment melt, 10% AOC melt) to a depleted mantle ("baseline ambient mantle" (BAM), 785 with a trace element composition of DMM, and isotopic composition of Sample 126171; Søager et 786 al., 2013) melted at F=0.04-0.3. The blue field represents the additional solution space made 787 available by an increase in slab temperature (with a change in the partition coefficients of La, Sm, 788 and Nd; see SIA4.1). The green field represents the additional solution space made available by 789 mixing in various proportions of an EM1-like mantle source to BAM. b) The pink field represents the 790 solution space made available by varying the proportion of AOC and sediment, and the total 791 amount of slab melt, as well as slab temperature. The latter has a much smaller effect than in a), 792 hence the red and blue fields are combined for simplicity. As in a), the green field shows the 793 additional solution space made available by increasing enrichment. In both figures, it is clear that 794 only increasing EM1-like enrichment can recreate the composition at Don Casimiro-Maipo. The 795 diamonds represent the best model fits for Villarrica (F=0.1, 7.5% slab addition consisting of 17% 796 sediment, with no enrichment) and Don Casimiro-Maipo (F=0.056, 7.5% slab addition consisting of 797 23% sediment, with 80% EM1 enrichment). The full trace element signatures of these models are 798 shown in Fig. 8.

799

Fig. 8 – Trace element modelling results. The trace element composition of Villarrica can be well matched at F=0.1 with addition of 7.5% slab melt consisting of 17% sediment. The extreme trace element enrichment at Don Casimiro-Maipo is best recreated by addition of 80% enriched source, and slight changes in the composition of the slab addition (23% AOC vs. 17% at Villarrica). The solid black lines in both figures represents the best model fit for each centre at the enrichment and proportions of AOC:SED mentioned above using low temperature slab partition coefficients. This provides the best match for Villarrica. The dashed grey lines represent the model fit for each centre at increased slab temperatures, assuming 70% loss of Cs in the forearc. This provides the best
model match for Don Casimiro-Maipo. Model envelopes are produced by adding and subtracting 1σ
of 33 measurements of ODP1232 (Turner et al., 2017). Melting conditions generated directly from
the physical modelling results of Turner et al. (2016).

811

812 Fig. 9– Modelling contamination of the mantle source by subduction erosion. a) Isotope systematics 813 of contamination of the Villarrica mantle source by subduction erosion. The isotopic composition of 814 the Villarrica mantle source was calculated from the Villarrica primitive average. Only a small 815 percentage of mixing lines pass near Don Casimiro-Maipo. b-c) Trace element and isotopic 816 signatures of the mixed compositions that produce isotopic ratios close to Don Casimiro-Maipo (red 817 lines in a), and have Eu/Eu*>0.9. The trace element composition of the Villarrica mantle source 818 was calculated from the best model fit in Fig. 8. None of these lithologies can recreate Sr, Ba and 819 ⁸⁷Sr/⁸⁶Sr systematics at Don Casimiro-Maipo. Error bars show 1σ of primitive samples at Don 820 Casimiro-Maipo and Villarrica based on 5 and 3 analyses respectively.

821

822 Fig. 10- Variation in rear-arc slab inputs. a) Mixing models of slab-sensitive ratios between a melt of 823 a rear-arc mantle source (F=0.02, melting at 3.3Gpa) that has not received slab inputs, and a melt of 824 a rear-arc mantle source at the same conditions that has received slab inputs (20% sediment, 80% 825 AOC). The orange wedge reflects progressive mixing of 10% slab (with ODP1232 \pm 1 σ) into a mantle 826 source with 0% EM1 enrichment, and the green wedge reflects mixing of 17% slab (with ODP1232± 827 1σ) into a mantle source with 80% EM1 enrichment. Rear-arc trace element ratios follow these 828 mixing lines remarkably closely, with the small amount of scatter likely explained by variable 829 conditions of melting along the rear-arc. b) Trace element ratios sensitive to slab additions versus 830 trench distance. MORB-OIB like values (red bar) start to appear at distances greater than 425km.

Remarkably high, arc-like ratios are seen up to 600km from the trench, with large scatter in ratios
at large trench distances, contrary to the narrow range of observed isotopes.

833

834 Fig. 11 – Isotopic compositions of inferred SCLM melts within South America (approximate locations 835 on Fig. 2a). These clearly extend the array that passes from DMM/DAM through the arc-front and 836 rear-arc, to Don Casimiro-Maipo. The field produced by contaminating the Villarrica mantle source 837 with crustal lithologies (Fig. 9a) is also overlain. This follows a much shallower trajectory than SCLM 838 melts, and is significantly broader than the SCLM field. Mobile belt M-SCLM melts represent 839 alkaline volcanics erupted on the edges of the Sao Fransisco craton and the Rio Apa-Luis Alves 840 craton on the east coast of South America (Gibson et al., 1995; 2005 and Carlson et al., 1996). 841 Southern Rift melts were erupted just north of the NSVZ (Lucassen et al., 2002). Carboniferous 842 granitoids are from the Santo Domingo Complex of the Coastal Batholith (Parada et al., 1999). All 843 are suggested to have sampled M-SCLM. The green and blue lines show a recycling model in which 844 low degree melts are extracted from the depleted mantle (Salters and Stracke, 2004) and then aged 845 for up to 2 Ga using the parameters from Stracke et al. (2003; c.f. Turner et al., 2017). A model with 846 F=0.003-0.008 encapsulates the range of isotopic compositions observed in SCLM melts.

847

Fig. 12- Schematic diagrams showing possible explanations for the isotopic enrichment minimum,
and its relationship to tectonic features. a) A Permian suture zone corresponds with an absence of
M-SCLM. b) A slab tear allows depleted Pacific MORB mantle to upwell, and dilute the EM1-type
enrichment above the tear. As the proportion of depleted Pacific MORB mantle declines
northwards and southwards, isotopic signatures become more enriched. Upper plate geometry
adapted from box model of Søager et al. (2015b), pp. 1514.

Fig. 13 – Assessing olivine chemistry alongside regional isotopic trends. Olivine Mn/Fe decreases
southwards. However, over the same interval, there is a decrease in isotopic enrichment
southwards. Literature values from Brandt et al. (2017), with rear-arc olivine compositions of this
study overlain.

859

860	Fig. 14 – Schematic diagrams of rear-arc slab supply. a) Traditional model of slab supply, with
861	buoyant rise of slab fluids to the rear-arc from a slab melting at higher pressure and temperature
862	than at the arc-front. The composition of these fluids should change with increasing trench
863	distance, as the depth to the slab increases. Additionally, at large trench distances, very little to no
864	slab supply would be expected, due to the exhaustion of hydrous phases. b) Proposed model of this
865	study, inspired by Cagniocle et al. (2007), Wilson et al. (2014) and Cerpa et al. (2017). Down
866	dragging of fluids released at arc-front depths explains the similar composition of arc-front and
867	rear-arc slab supply.

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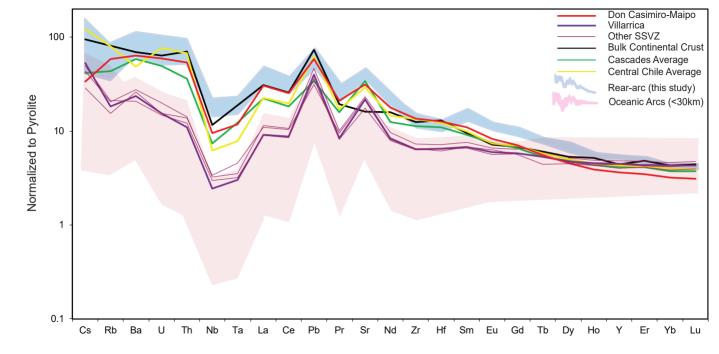
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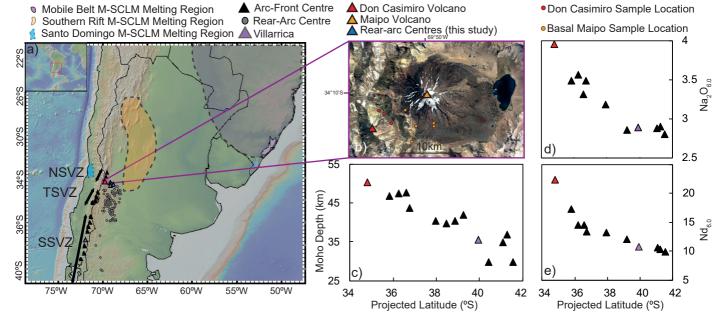
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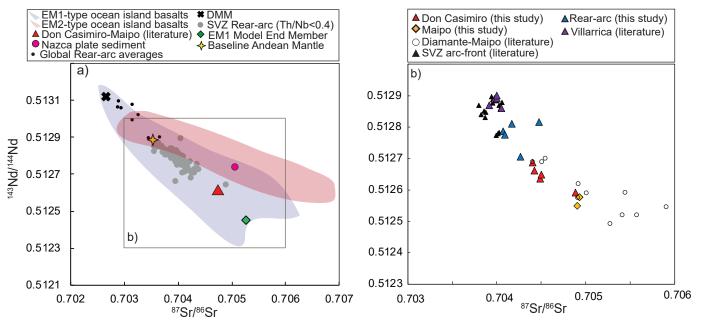
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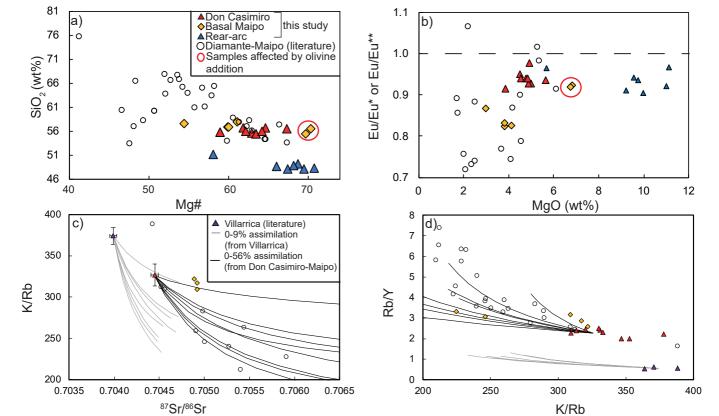
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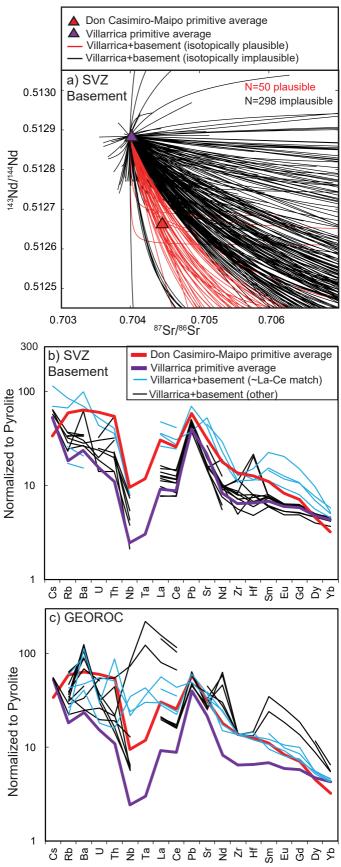
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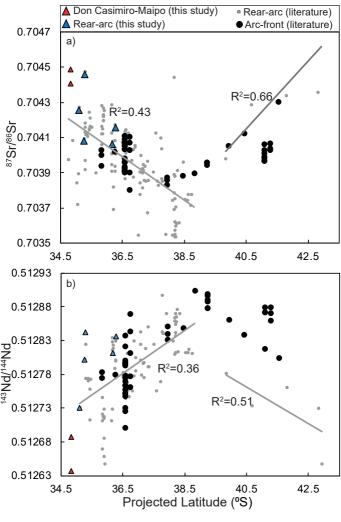


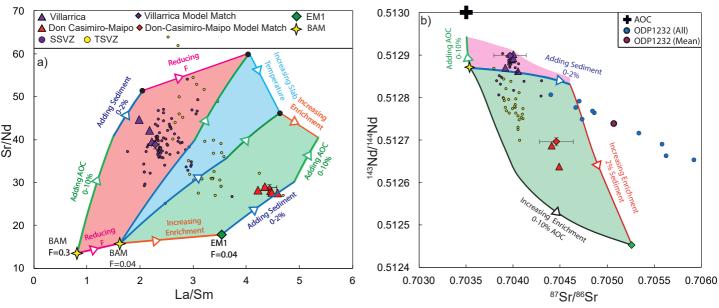


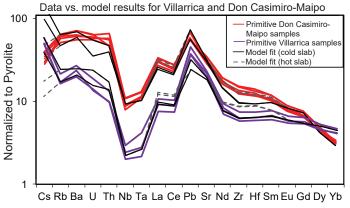


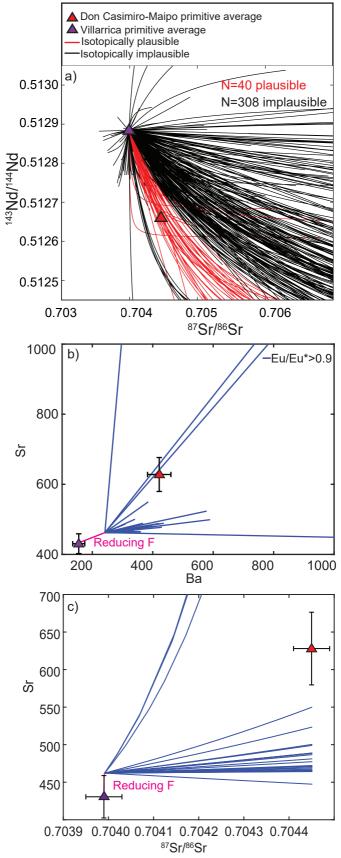


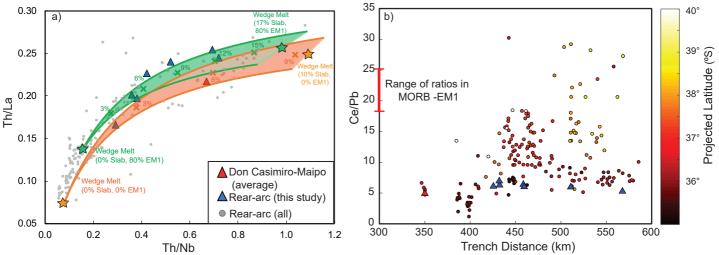


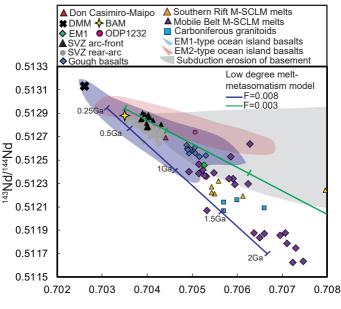




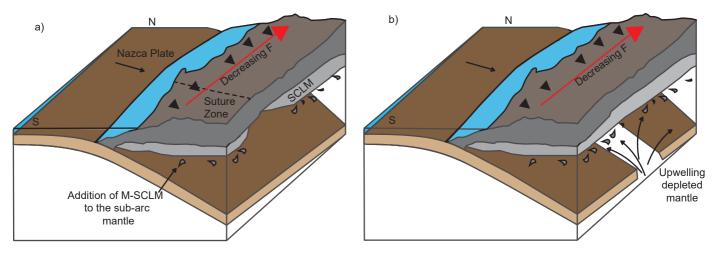


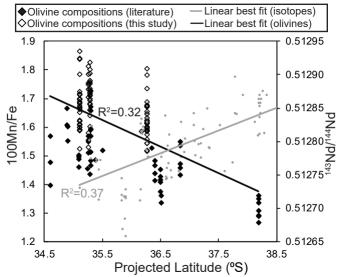


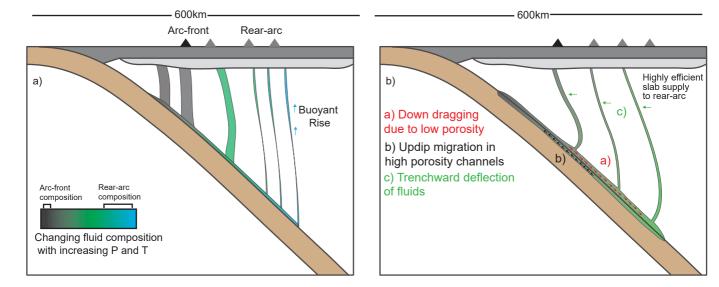




87Sr/86Sr







Appendix 1: Methods and Sample Details

SIA1 Data and Databases

Whole-rock and olivine analysis from this study is shown in Appendix 3. Compiled rear-arc

² and Diamante-Maipo literature data used in this study is in Appendix 4. The compilation

³ of SVZ basement compositions is in Appendix 5.

⁴ SIA2 Sample Characteristics

 $_{5}$ Six 100 μ m sections were made for Don Casimiro-Maipo samples spanning a range of MgO

6 contents. These were scanned using a 5MP slide scanner. Point counting was conducted in 7 JMicroVision by overlaying a 1000 point grid on these scans. Microscope observations were

^{*} made on any crystals that couldn't be identified with certainty in scans. Crystals smaller

 \sim than ~ 0.05 mm were classified as matrix. Amorphous or crystalline material sufficiently

¹⁰ distinct from the surrounding matrix that could not be identified as a mineral phase was classified as alteration.

Table SA1: Point counting results for Don Casimiro-Maipo samples (ordered by decreasing wt% MgO).

Sample	MgO (wt%)	Crystals (%)	Matrix (%)	Vesicles (%)	Alteration (%)
G0020315-1	6.82	11	89	0	0.1
G0020315-2	6.73	10	90	0	0.3
G0270215-1D	5.63	17	81	0	0.3
G0270215-1C	4.85	14	85	0	0.2
G0030315-4	3.87	11	72	16	0.0
G0030315-1	2.99	42	48	0	10

Table SA2: Percentage of each phenocryst phase in the crystal fraction. *The fine grained nature of these samples meant that phenocryst phases were harder to identify, so proportions are not as reliable as for other samples.

Sample	Olivine (%)	Clinopyroxene (%)	Plagioclase (%)	Oxide (%)	Orthopyroxene (%)
G0020315-1	66	34	0	0	0
G0020315-2	66	34	0	0	0
G0270215-1D*	6	15	46	33	0
$G0270215-1C^*$	8	20	32	41	0
G0030315-4	7	5	89	0	0
G0030315-1	3	13	82	0	3

Sample	Olivine	Plagioclase	Clinopyroxene	Matrix Textures	
G0020315-1	Predominantly subhedral-anhedral 2-0.5mm grains, and smaller, <0.5mm anhedral grains. Both types form clots, and show association with clinopyroxene. Most olivines contain oxide inclusions.	No phenocrysts.	Subhedral-anhedral 2-0.5mm grains, often forming clots. Smaller, <0.5mm anhedral grains within the matrix. Some show breakdown in their cores.	Dark, fine grained matrix with white streaks of amorphous material. Needle-shaped matrix plagioclase aligns with these streaks. A few 1-2mm patches of alteration are associated with reacted olivine cores.	
G0020315-2	Euhedral 1-3mm grains, subhedral 0.5-2mm clots associated with clinopyroxene, and smaller isolated <1mm anhedral grains.	No phenocrysts.	Subhedral 1-2mm grains, often associated with olivine clots. As above, some show broken down cores.	Dark, fine grained matrix, with white amorphous material in streaks. Small needle-shaped plagioclase shows alignment around large crystals. Abundant anhedral oxide grains.	
G0270215-1D	Anhedral <0.2mm grains with abraded edges. These are associated with patches of small red amorphous material.	Blocky to needle-shaped plagioclase. No alignment.	Sub-anhedral, 0.3-1mm grains. These are more commonly associated with the red material than olivines.	Dark, fine grained matrix, with poorly aligned, needle-shaped plagioclase, and dispersed anhedral oxides. Patches of white, amorphous material.	
G0270215-1C	Anhedral, <0.1mm grains.	Elongated needles, 20:1 aspect ratio. Aligned in matrix.	Sub-anhedral 0.3-1mm grains, associated with red amorphous material and oxide growth.	Dominated by small, aligned, needle-shaped plagioclase, with euhedral-anhedral opaque oxides.	
G0030315-4	Anhedral, <1mm grains, with highly abraded edges, likely these were originally clots.	Large, 2mm+, blocky plagioclase, showing sieve textured cores (indicating rapid growth/cooling/heating).	Euhedral, 2-4mm grains, and smaller, <1mm highly abraded anhedral grains.	Highly vesicular texture. Matrix dominated by glass.	
G0030315-1	A few subhedral, 1-2mm olivines. Some associated with large, 1-2mm cubic oxide phases.	Several populations of plagioclase. 1) blocky 2-4mm rectangular macrocrysts, with large fluid inclusions. 2) lath shaped 1-3mm grains, with inclusion-free cores. 3) needle shaped (<1mm) grains, with sieve textured cores.	Euhedral-subhedral, 1-2mm grains with oxide inclusions. Some are also associated with very large, 1-3mm oxide grains. Additionally, one 4mm clot of several grains and oxides is observed. In some places, clinopyroxene forms anhedral intergrowths with plagioclase, and orthopyroxene.	Extremely crystal-rich. Large areas of amorphous material within the slide are associated with large blocky plagioclase macrocrysts. Between phenocrysts, the matrix is dark and amorphous.	

 Table SA3:
 Description of Don Casimiro-Maipo samples in thin section.

¹² SIA3 Methodologies

¹³ SIA3.1 Whole Rock Chemistry

Sixteen samples from Don Casimiro-Maipo and seven rear-arc samples were prepared for whole-rock analysis
at the University of Oxford. Arc-front lava samples were cleaned in an ultrasonic bath until the water ran
clear. These were then split into 2-3cm chunks using a pneumatic rock splitter. Rear-arc samples were wet
sieved, and unaltered clasts from the coarsest fraction (>2mm) were hand picked for whole rock analysis.
Both sample types were crushed in a steel jaw crusher, and then powdered in an agate ball mill.

Major Element Analysis: Major element analyses was conducted by using a PANalytical Axios Advanced
 X-ray fluorescence spectrometer at the University of Leicester, UK, following the methods of Knott et al. (2016).

Trace Element Analysis: $\sim 100 \text{ mg}$ of each powder as well as reference materials BCR-2a, BHVO-2, 23 W-2a and AGV-2, were digested in \sim 4ml of 7.5M HNO₃ and \sim 1ml of 24M HF. The samples where then 24 evaporated and redissolved twice in ~ 4 ml 7.5M HNO₃ to eradicate fluoride precipitates. At this stage, a 25 sample split was made for isotopic analysis. Trace element analyses were conducted using a Perkin Elmer 26 NexION 350D quadrupole ICP-MS for the majority of elements. An additional 1200x dilution and addition 27 of an internal standard containing Rh, Re, and In was achieved using the ESI prepFAST system producing 28 a 6000x solution in which to measure lower concentration elements. All prior dilutions were performed 29 gravimetrically. Sample G0030315-4 was run every four samples, and the entire run was duplicated in reverse 30 order to monitor drift. Drift correction and data reduction were carried out following Turner et al. (2013). 31 Due to malfunctioning of the Prepfast system, a planned run at 60,000x dilution for higher concentration 32 elements using the same method failed. Thus, Sr and Ba concentrations were measured using a Thermo 33 34 Element 2 ICP-MS (also at the University of Oxford) at a 1,200,000x dilution (performed gravimetrically). The digestion procedure, and sample homogeneity can be assessed by calculating the % difference between 35 the measured element concentrations of two duplicates produced from the powders of this study, and two 36 from an additional set of samples digested alongside these. Most measurements lie within 5%, suggesting 37 that the digestion process, and samples, were reasonably homogeneous (table SA4). 38

39 Isotopic Analysis

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Strontium (Sr) and neodymium (Nd) isotope analyses were performed on a Thermo Scientific Triton-series 40 multicollector mass spectrometer at the School of Earth and Environment, University of Leeds. Eleven 41 whole-rock powders were digested alongside those for trace element analysis, followed by subsequent drying 42 and pick up in concentrated ultrapure HNO_3 and HCl acids. The final dried samples were diluted and 43 centrifuged for 15 minutes at 2000 rpm. Sr and Nd were extracted by using Sr-Spec and TRU-spec resins and 44 conventional ion-exchange chromatographic techniques. ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd ratios were normalized for 45 mass fractionation to ⁸⁶Sr/⁸⁸Sr=0.1194 and ¹⁴⁶Nd/¹⁴⁴Nd=0.7219. Samples for Sr were loaded onto previously 46 outgassed W filaments and samples for Nd were loaded onto previously outgassed Re filaments. Sr and Nd 47 were analysed while the current was maintained between 3-5 V (for 88 Sr) and 0.4-0.8 V (for 144 Nd). 48

The average ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ obtained from repeated measurements of NIST SRM-987 during the course of this study was 0.710271. Similarly, we measured ${}^{143}\text{Nd}/{}^{144}\text{Nd}=0.511837$ for the LaJolla standard. Two samples (G0020315-2 and G0270215-1C) were analyzed for ${}^{143}\text{Nd}/{}^{144}\text{Nd}$ in a separate run, in which we measured ${}^{143}\text{Nd}/{}^{144}\text{Nd}=0.511850$ for the LaJolla standard. We also successfully reproduced the published USGS standard BHVO-2 values for ${}^{87}\text{Sr}/{}^{86}\text{Sr}=0.703468$ and ${}^{143}\text{Nd}/{}^{144}\text{Nd}=0.512995$ (Weis et al., 2005). Details on the sample preparation and analytical protocols are described in Crummy et al. (2014).

⁵⁵ SIA3.2 Correction of Isotopic data of this study and Literature ⁵⁶ Data

57 Sr and Nd isotopes from the literature and this study were corrected to be in line with the analytical procedure 58 used for SVZ data presented in Turner et al. (2016). Various standards were corrected to be equivalent

to La Jolla=0.511858, NBS 987=0.710235 based on session averages.

Table SA4: % deviation of four duplicates ran during analysis on the "quad" (except starred elements, where % deviation is reported from the "element"). (4) represents a duplicate of G0030315-4, (1) represents a duplicate of MDZ1, 079a and B1 are samples measured in the same run, but for use in a study on the Main Ethiopian Rift. % deviation is defined as (Measurement 1-Measurement 2)/(Average of measurement 1 and 2) x 100.

	% difference (4)	% difference (1)	% difference (079)	% difference (B1)
Li	4.13	1.74	-0.97	0.35
Be	-4.52	60.2	-12.4	1.29
\mathbf{Sc}	5.89	-27.0	-20.5	-0.72
V	2.02	-1.95	-0.65	0.15
Cr	3.02	0.41	1.14	-0.99
Ni	3.53	0.74	0.86	1.47
Cu	4.71	-3.94	-0.26	1.76
Rb	3.64	0.28	-9.66	0.30
Sr^*	5.54	1.74	0.35	-5.67
Υ	2.99	-3.08	-6.18	-0.94
Zr	2.12	-0.29	-0.42	0.32
Nb	3.09	0.21	-0.30	1.16
\mathbf{Cs}	8.90	-5.49	-4.43	9.11
Ba^*	6.08	3.17	3.65	-2.89
La	3.52	0.58	1.10	0.82
Ce	3.31	-1.09	0.94	0.70
\Pr	4.57	0.71	2.05	0.80
Nd	3.29	3.63	-0.88	-0.53
Sm	0.00	2.17	1.35	2.23
Eu	2.21	2.20	1.55	-1.12
Gd	3.09	-2.92	-2.87	1.04
Tb	3.26	-2.17	2.29	0.91
Dy	3.83	-0.60	2.67	0.78
Ho	4.64	-2.73	-0.32	3.88
\mathbf{Er}	2.99	4.01	-2.53	3.70
Tm	2.81	-5.67	-3.75	0.51
Yb	2.67	1.49	-0.23	1.69
Lu	1.28	1.41	-3.58	1.15
$_{\mathrm{Hf}}$	3.34	0.93	-0.87	1.40
Ta	-2.98	-0.57	1.60	-0.94
\mathbf{Pb}	3.55	-2.14	-9.60	1.72
Th	3.48	-2.05	-17.6	-1.10
U	3.33	-0.09	0.56	-0.11

⁶⁰ SIA3.3 Olivine Chemistry

Olivines were picked from the 1000-250μm grain fractions, and mounted in epoxy rings. These
mounts were polished and carbon coated for EMPA analysis. Analysis was conducted with
an accelerating voltage of 15KeV, a current of 100nA and a 10μm spot size. St John's Island
and Fayalite were run as secondary standards, with % recoveries reported in table SA5.

LA-ICP-MS analysis was conducted at the University of Cambridge. Three instrument runs were performed over two days. The run conditions were 20Hz, 80µm, 9.5J/cm², 0.8L/min of gas flow for He and Ar. Where possible, three spots were placed in the centre of the olivine, with no overlap. For very small grains, only 2 spots were placed, as it is preferable to have fewer spots nearer the centre than more spots towards the grain edges, due to the reasonably high diffusivity of elements of interest to this study. To allow drift correction, 3 spots on each of

NISTSRM612, NISTSRM610, 06SGOL03 (inhouse olivine standard) and BD4074OL-7 (a San 71 Carlos olivine) were run every 30 analyses. ²⁹Si was used as an internal standard, with values 72 obtained from EMPA grain averages. Signals were selected using Glitter Processing software, 73 with normalization to NISTSRM612 via a quadratic drift correction. This was effective at 74 removing drift, as determined by observing the stability of NISTSRM610, 06SGOL03 and 75 BD4074OL-7 following this correction. Multiple secondary standards were run to assess the 76 accuracy of the measurement for elements of interest (NISTSRM610, NISTSRM614, BCR-2G). 77 Measurement of inhouse olivines O6SGO103 and BD407OL-7 provided an excellent assessment 78 of the success of the drift correction performed on glass standards. Elements of interest in this 79 study drifted by less than 7%. Although Mn and Ni recovery on glass standards were good, 80 these elements were recovered more poorly on BCR-2G. This, and the fact that previous 81 studies in this area have used EMPA Ni and Mn data, resulted in us choosing to use EMPA 82 data for consistency, as most of the peridotite-pyroxenite ratios involve normalization to Fe 83 (which is reported from EMPA). Detection limits, and 1 sigma uncertainties calculated from 84 Glitter for a representative sample (MDZ2) are shown in table SA6. 85

Table SA5: Peak count times, and calibration materials used. % recovery calculated below is defined as Measured Average/Published Average x 100. Where no published value exists for a standard, - is shown. Detection limits are reported from 9 spots on MDZ8. These are assumed to be representative of the analysis as a whole.

	Mn	Fe	Mg	Si	Ca	Ni
Peak count times (s)	20	30	20	20	70	70
Calibration material	Mn metal	Andradite	MgO	Albite	Wollastonite	Ni metal
% Rec Fayalite	97.7	94.2	-	99.6	97.0	-
% Rec St Johns Island Olivine	106.1	96.1	99.6	99.2	-	90.1
Detection limit (ppm)	173	197	173	98	74	137

Table SA6: Minimum (99% Confidence) detection limits (ppm), and 1 sigma uncertainties (ppm) for all spots on MDZ2, taken as representative for the analysis as a whole. The mass in brackets represents the isotope measured on the ICP-MS.

Element	MDL	1 σ uncertainty
$\operatorname{Li}(7)$	0.115	0.151
Na(23)	1.121	6.92
Al(27)	0.318	39.3
P(31)	13.76	47.3
Ca(43)	133	192
Sc(45)	0.062	0.412
Ti(47)	0.551	18.4
V(51)	0.043	0.504
$\operatorname{Cr}(53)$	0.578	56.0
Mn(55)	0.136	88.4
$\mathrm{Co}(59)$	0.049	7.91
Ni(60)	0.171	191
Cu(65)	0.351	0.416
$\operatorname{Zn}(66)$	0.373	10.1
Ga(69)	0.042	0.049
Ge(72)	0.370	0.208
Y(89)	0.037	0.025
$\operatorname{Zr}(90)$	0.033	0.052
Nb(93)	0.035	0.021

Table SA7: % recovery of NISTSRM610 (average of 3 spots). (x,y) refers to when this secondary standard was run, x referring to the instrument run (1-3), and y to the order of measurement within a given run. These values were used to check the success of the quadratic drift correction. Across the three instrument runs, most elements drift by less than 10%.

Element	(1,1)	(1,2)	(1,3)	(1,4)	(1,5)	(2,1)	(2,2)	(2,3)	(2,4)	(3,1)	(3,2)	(3,3)	(3,4)	AVERAGE
Li(7)	98.0	105.5	103.7	106.7	105.9	107.3	104.3	101.1	103.6	108.0	99.6	96.8	98.2	103.0
Na(23)	98.2	105.7	107.0	106.1	105.9	105.9	104.9	101.4	103.9	105.0	103.0	101.1	102.1	103.9
Al(27)	102.3	105.5	105.9	106.2	107.8	108.3	104.2	104.9	107.3	105.6	107.2	103.5	104.9	105.7
P(31)	107.8	105.2	99.7	130.9	194.5	100.5	103.9	101.0	100.5	125.0	113.1	117.6	133.9	118.0
Ca(43)	95.9	103.7	100.6	106.7	107.3	105.2	102.5	99.2	102.2	105.2	100.1	99.5	104.5	102.5
Sc(45)	98.7	109.1	108.0	109.8	108.8	110.2	110.9	105.6	108.9	108.5	106.7	108.0	109.6	107.9
Ti(47)	115.2	120.8	116.9	120.0	119.0	125.1	128.7	124.5	125.5	125.3	125.9	122.8	125.4	122.7
V(51)	95.7	101.9	100.1	105.2	105.8	104.8	102.6	100.1	105.1	105.9	101.7	99.9	103.7	102.5
Cr(53)	103.4	107.7	109.4	113.6	119.5	120.7	115.9	112.1	115.1	114.1	111.4	112.7	116.7	113.2
Mn(55)	94.6	100.0	100.0	103.7	102.4	105.1	100.2	99.0	103.5	104.9	97.1	96.1	97.7	100.3
Co(59)	95.5	103.0	101.7	103.7	100.8	104.2	102.0	98.9	104.3	103.6	101.0	100.3	103.4	101.7
Ni(60)	92.1	100.9	99.8	103.7	99.9	105.6	100.1	98.2	100.0	104.3	101.8	100.5	102.2	100.7
Cu(65)	89.9	98.8	97.3	100.1	97.5	100.5	98.4	94.9	99.0	100.1	96.0	95.9	97.6	97.4
$\operatorname{Zn}(66)$	98.5	104.1	102.3	108.8	110.2	111.1	104.4	100.1	106.6	109.9	106.4	104.1	102.9	105.3
Ga(69)	92.2	97.7	96.5	101.1	101.5	101.7	99.0	97.2	99.4	101.4	98.9	98.3	100.8	98.9
Ge(72)	81.5	86.7	85.7	89.1	88.8	92.4	89.0	85.6	86.6	92.0	87.5	87.8	89.8	87.9
Y(89)	93.1	102.1	99.8	102.8	102.1	104.0	102.9	100.5	100.3	102.5	101.4	101.8	103.8	101.3
$\operatorname{Zr}(90)$	89.8	95.9	94.6	97.9	98.1	98.6	98.5	94.6	95.4	97.5	96.8	96.0	98.7	96.3
Nb(93)	92.8	99.9	98.9	102.4	101.7	102.2	102.2	98.1	100.2	102.9	99.9	99.2	101.1	100.1

Element	(1,1)	(2,1)	(2,2)	(3,1)	(3,2)	BCR-2G Average	(2,1)	(3,1)	NISTSRM614 Average
$\operatorname{Li}(7)$	100.2	104.5	101.7	105.8	94.6	101.4	84.4	103.7	94.1
Na(23)	96.2	99.1	97.0	101.4	96.9	98.1	105.5	103.1	104.3
Al(27)	103.8	103.4	99.9	104.6	103.3	103.0	106.6	106.3	106.4
P(31)	101.3	84.9	86.8	108.4	120.1	100.3	268.2	260.0	264.1
Ca(43)	99.0	97.9	98.9	101.1	99.1	99.2	106.2	103.7	105.0
Sc(45)	97.5	102.1	96.4	100.8	99.9	99.3	217.1	185.9	201.5
Ti(47)	107.3	108.4	105.2	110.9	106.3	107.6	122.1	101.3	111.7
V(51)	97.6	100.1	101.1	102.8	99.6	100.2	109.7	99.9	104.8
Cr(53)	93.2	104.4	92.9	97.1	101.1	97.7	94.8	113.0	103.9
Mn(55)	92.4	96.6	94.9	98.5	90.0	94.5	105.6	89.7	97.7
Co(59)	99.0	97.8	97.0	97.9	96.2	97.6	92.6	95.0	93.8
Ni(60)	89.4	86.9	83.8	90.2	86.5	87.4	107.9	71.8	89.8
Cu(65)	71.0	72.5	67.5	71.1	69.6	70.3	88.8	109.0	98.9
$\operatorname{Zn}(66)$	121.5	134.0	127.4	134.9	120.7	127.7	91.4	89.2	90.3
Ga(69)	111.6	117.1	109.2	118.4	114.5	114.2	96.9	93.9	95.4
Ge(72)	99.6	112.9	95.6	101.1	102.4	102.3	86.3	110.8	98.5
Y(89)	87.3	90.0	81.9	90.6	89.1	87.8	102.0	99.4	100.7
Zr(90)	84.5	87.0	78.3	87.7	86.3	84.7	90.3	91.6	90.9
Nb(93)	88.6	91.6	86.6	91.0	90.2	89.6	93.7	99.3	96.5

Table SA8: % recovery of BCR-2G, using averages from 3 spots, and % recovery of NISTSRM614. The % recovery for NISTSRM614 is not shown for the first run. These were the first spots ran by the laser, and it has been shown that the system takes a few spots to warm up and settle after switching from solution mode to laser mode.

⁸⁶ SIA4 Mantle Melting Model Details

As discussed in the text, this study uses an updated version of the model of Turner and Langmuir (2015b). Below, the justification of various inputs to the model are discussed. The updated Matlab scripts ADD_SLAB.m, ADD_SLAB_HOTP.m and ModeMelt_Hydrous.m have been uploaded into the supplementary information.

91 SIA4.1 Model Partition Coefficients

Hydrous Partition Coefficients for Mantle Melting: This study utilizes hydrous
 partition coefficients and mantle modes from Turner and Langmuir (2015b; detail in their
 supplement). However, we slightly updated the partitioning behaviour of several elements:

⁹⁶ 1) The bulk partition coefficient of Eu is obtained by a log extrapolation from those of Sm and ⁹⁷ Gd:

$$\mathrm{Kd}_{Eu} = (\mathrm{Kd}_{Sm}^* \mathrm{Kd}_{Gd})^{0.5}$$

⁹⁹ correcting an error in Turner and Langmuir (2015b).

¹⁰⁰ 2) The partition coefficient of Hf is assumed to be equal to that of Sm.

¹⁰¹ 3) The partition coefficient of Zr is approximated as:

$$\mathrm{Kd}_{Zr} = (\mathrm{Kd}_{Nd}^* \mathrm{Kd}_{Sm})^{0.5}$$

Hydrous Partition Coefficients for slab melting: As above, this study uses slightly
adapted partition coefficients for slab melting to those in Turner and Langmuir (2015b).
These lie within the limits of experimental values (figure SA1). The partition coefficients for
the LREE (La, Ce, Nd, and Sm) were reduced by 40% to reflect the hotter slab underneath
Don Casimiro-Maipo (inline with experimental high temperature partitioning results; figure
SA1). Additionally, the partition coefficients of Zr and Hf were reduced, and 70% of Cs
was assumed to be lost to the forearc.

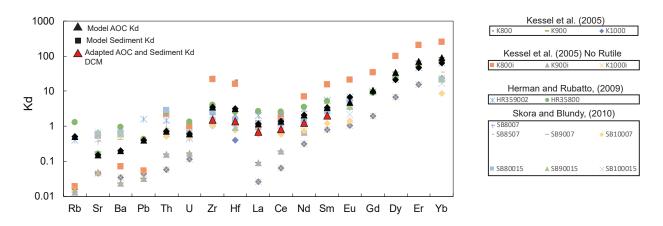


Figure SA1: Slab melting partition coefficients lie within experimental values for 800-1000°C.

110 SIA4.2 Model End-Members

"Ambient Andean Mantle" (Depleted Mantle End-Member): The isotopic composition
of the depleted mantle end member was set to ⁸⁷Sr/⁸⁶Sr=0.70355 and ¹⁴³Nd/¹⁴⁴Nd=0.512871,
the isotopic composition of sample 126171 of Soager et al. (2013) following the isotopic
correction discussed above. This sample lies at the depleted end of the rear-arc array,
and thus reflects the composition of the Andean mantle prior to the addition of EM1
enrichment. For trace element modelling, the depleted mantle end-member is DMM of
Workman and Hart (2005).

AOC: The subducting Nazca plate has Pacific affinities. Trace element concentrations were obtained from melting of N-MORB (Gale et al., 2013) at 876°C, F=0.3. The isotopic composition of AOC was set to ${}^{87}\text{Sr}/{}^{86}\text{Sr} = 0.7035$, ${}^{143}\text{Nd}/{}^{144}\text{Nd} = 0.5130$.

Sediment: The trace element composition of the sediment end-member was obtained from 33 samples of ODP1232 (c.f. Turner et al., 2017). To produce an envelope of model results for trace elements and isotopes, one standard deviation of measured samples were added and subtracted to the mean. Three outliers were discarded based on their anomalously high Sr isotope ratios.

Enriched EM1 end-member: Enrichment at Gough represents a mix of M-SCLM, and ambient mantle. The isotopic signature of recycled M-SCLM depends on the storage time (Turner et al., 2017). As several samples within this study have signatures more enriched than Gough, we use the isotopic composition of a sample from Pitcairn (57DS9; Stracke et al., 2003) as the EM1-like isotopic end member. This lies at the enriched end of the field enclosing EM1-type ocean island basalts. The trace element concentration was obtained from the inversion of Turner et al. (2017), with a few changes:

132 1) the bulk Eu partition coefficient was set at:

$$\mathrm{Kd}_{Eu} = (\mathrm{Kd}_{Sm}^*\mathrm{Kd}_{Gd})^{0.5}$$

¹³⁴ correcting an error in Turner et al. (2017) where it was set at 0.

¹³⁵ 2) The Zr concentration in Gough primary melts was adjusted to the standard EM1 value ¹³⁶ Zr/Hf = 45.

- ¹³⁷ 3) Zr partition coefficients were extrapolated from Salters and Longhi (1999).
- ¹³⁸ 4) Nb and Ta partition coefficients were taken as $Kd_{La}/3$, based on ratios from Workman and ¹³⁹ Hart (2005).

Table SA9: Gough mantle source composition used in this study compared to that of Turner et al. (2017; T2017).

	Eu	Zr	Nb	Ta
This study	0.130	6.70	0.403	0.024
T2017	0.0234	11.47	0.472	0.028

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Appendix 2

SIB1 Evidence for Olivine Addition

EMPA analysis indicates that Don Casimiro-Maipo olivines lie significantly below the olivine-whole rock equilibrium line (Figure SB1a), indicating excess olivine accumulation. The two most primitive samples, which are from the basal section of Maipo volcano, can be related to the otherwise coherent liquid line of descent among the Diamante Caldera samples by the addition of 7 wt. % Fo 81 olivine (figure SB1b-d.). All samples have phenocrysts of olivine and clinopyroxene, so this trend is not consistent with fractionation of these two phases together. Olivine may have been accumulated from a mush pile produced by fractionation before clinopyroxene appears on the liquidus. This serves as an excellent example of the importance of utilizing individual mineral analyses in conjunction with whole-rock data when interpreting subtle geochemical variations within individual volcanic groups. While it is tempting to represent the parental composition of a volcanic group using the most olivine-phyric samples (6-7vol% olivine, in this case), such samples can often have more complicated petrogenetic histories.

SIB2 Changes in Slab Conditions Inferred from Zr-Hf Anomalies

Using Hf/Sm as a proxy, we find that rear-arc and most arc-front volcanics have a similar range of Zr-Hf values, but Don Casimiro-Maipo has a less pronounced Zr-Hf anomaly (figure SB2). The smaller Hf/Sm (and thus Zr-Hf anomaly) at Don Casimiro-Maipo may reflect the fact that the slab is slightly hotter, which causes residual slab zircon to become more soluble in the slab melt (Hirai et al., 2018).

SIB3 Evidence for AFC processing

As mentioned briefly in the main text, when samples from the entire Diamante-Maipo caldera complex are considered, the correlations between ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ and ${}^{143}\text{Nd}/{}^{144}\text{Nd}$ and indices for fractionation and assimilation (MgO and Eu/Eu*) provide strong evidence for AFC (figure SB3)

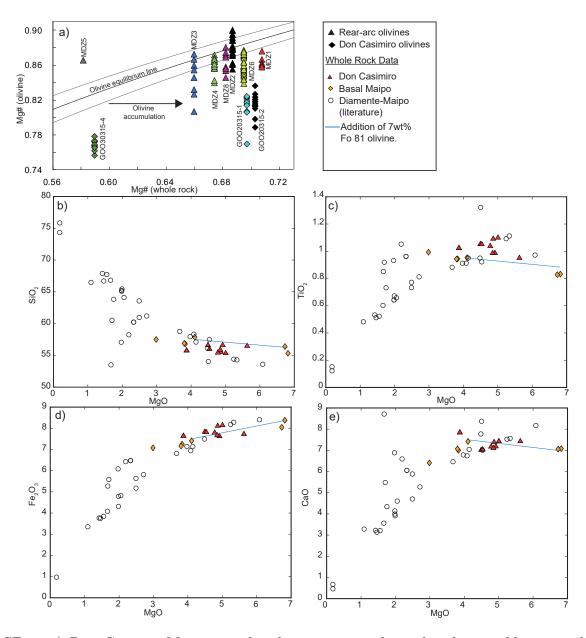


Figure SB1: a) Don Casimiro-Maipo samples show strong evidence for olivine addition, with olivine compositions plotting below an equilibrium line calculated using Roeder and Emslie (1970), Kd= 0.3 ± 0.03 , assuming whole-rock Fe³⁺/Fe_T=0.3 (Holm et al., 2016). Most rear-arc olivines plot near to or below the equilibrium line, suggesting minor olivine addition (assuming whole-rock Fe³⁺/Fe_T=0.15, Søager et al., 2015b). b-d) Addition of 7 wt% Fo 81 olivine from more evolved Maipo samples produces a good match to the most primitive samples, showing that their high MgO contents are spurious. Diamante-Maipo whole-rock data literature data used in this study is from Hickey et al. (1986), Futa and Stern, (1988), Sruoga et al. (2005), Holm et al. (2014) and Hickey et al. (2016).

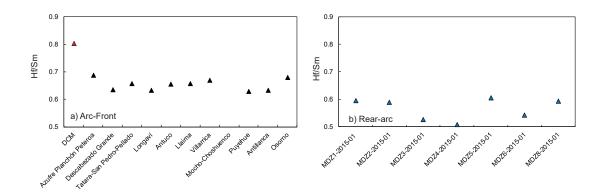


Figure SB2: a) Don Casimiro-Maipo has a significantly different Hf/Sm signature to the rest of the SVZ (data for other centers from Turner et al., 2016). b) Rear-arc centers of this study have similar Hf/Sm ratios to the SVZ arc-front as a whole.

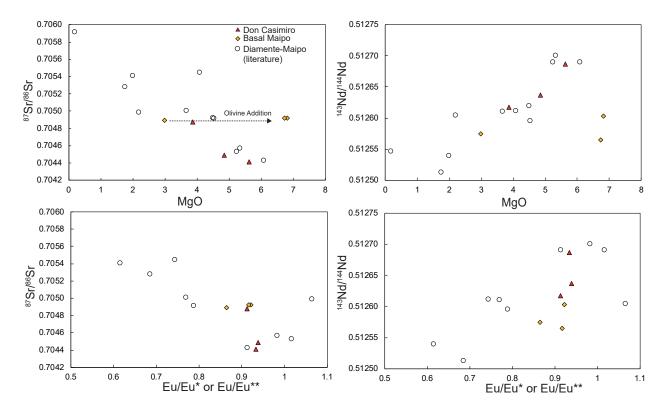
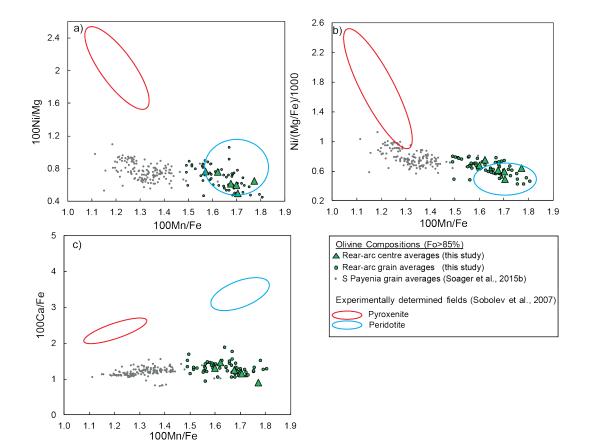


Figure SB3: Within the Diamante-Maipo Caldera, isotopic enrichment increases with assimilation and fractionation. The signal of olivine addition for the two most primitive Maipo samples is also clear.



SIB4 Olivine Chemistry

Figure SB4: Olivine analysis of this study lie within, or close to the peridotite field from Sobolev et al. (2007). Additionally, they clearly lie at more peridotic values than the results of Soager et al. (2015b), despite showing significantly higher isotopic enrichment. While in b), the samples of Soager et al. (2015b) trend towards the pyroxenite field of Sobolev et al. (2007), in a) and c) they do not.

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