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### Version of attached file:

Accepted Version

### Peer-review status of attached file:

Peer-reviewed

### Citation for published item:

Chamberlain, K.J. and Barclay, J. and Preece, K.J. and Brown, R.J. and Davidson, J.P. (2019) 'Lower crustal heterogeneity and fractional crystallization control evolution of small-volume magma batches at ocean island volcanoes (Ascension Island, South Atlantic).', *Journal of petrology.*, 60 (8). pp. 1489-1522.

### Further information on publisher's website:

<https://doi.org/10.1093/petrology/egz037>

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This is a pre-copyedited, author-produced PDF of an article accepted for publication in *Journal of Petrology* following peer review. The version of record Chamberlain, K.J., Barclay, J., Preece, K.J., Brown, R.J. Davidson, J.P. (2019). Lower Crustal Heterogeneity and Fractional Crystallization Control Evolution of Small-volume Magma Batches at Ocean Island Volcanoes (Ascension Island, South Atlantic). *Journal of Petrology* 60(8): 1489-1522 is available online at: <https://doi.org/10.1093/petrology/egz037>

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1 Lower Crustal Heterogeneity and Fractional Crystallisation Control  
2 Evolution of Small Volume Magma Batches at Ocean Island  
3 Volcanoes (Ascension Island, South Atlantic)

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31 Manuscript for: *Journal of Petrology*

32 Running title: Evolution of Ascension Island magmatism

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34 Keywords: Ascension Island, ocean island volcanism, fractional crystallisation, magmatic  
35 processes, oceanic crust

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40

41 **ABSTRACT**

42 Ocean island volcanoes erupt a wide range of magmatic compositions via a diverse range of  
43 eruptive styles. Understanding where and how these melts evolve is thus an essential  
44 component in the anticipation of future volcanic activity. Here we examine the role of crustal  
45 structure and magmatic flux in controlling the location, evolution and ultimately composition  
46 of melts at Ascension Island. Ascension Island, in the south Atlantic, is an ocean island  
47 volcano which has produced a continuum of eruptive compositions from basalt to rhyolite in  
48 its 1-million-year subaerial eruptive history. Volcanic rocks broadly follow a silica-  
49 undersaturated subalkaline evolutionary trend and new data presented here show a continuous  
50 compositional trend from basalt through trachyte to rhyolite. Detailed petrographic  
51 observations are combined with *in-situ* geochemical analyses of crystals and glass, and new  
52 whole rock major and trace element data from mafic and felsic pyroclastic and effusive  
53 deposits that span the entire range in eruptive ages and compositions found on Ascension  
54 Island. These data show that extensive fractional crystallisation is the main driver for the  
55 production of felsic melt for Ascension Island; a volcano built on thin, young, oceanic crust.  
56 Strong spatial variations in the compositions of erupted magmas reveals the role of a  
57 heterogeneous lower crust: differing degrees of interaction with a zone of plutonic rocks are  
58 responsible for the range in mafic lava composition, and for the formation of the central and  
59 eastern felsic complexes. A central core of nested small-scale plutonic, or mush-like, bodies  
60 inhibits the ascent of mafic magmas, allowing sequential fractional crystallisation within the  
61 lower crust, and generating felsic magmas in the core of the island. There is no evidence for  
62 magma mixing preserved in any of the studied eruptions, suggesting that magma storage  
63 regions are transient, and material is not recycled between eruptions.

64

65 **INTRODUCTION**

66 Ocean island volcanoes remain enigmatic in terms of their origin (Niu et al., 2011), evolution  
67 and ability to produce a range of magmatic compositions and eruptive activity. Many ocean  
68 islands such as Iceland, Socorro, and the archipelagos of Hawaii, the Galapagos and the  
69 Canaries have been studied in detail (e.g. Sparks & Sigurdsson, 1987; Geist et al., 1988;  
70 Ablay et al., 1998; Geist et al., 1995; Bohrson et al., 1996; Harpp & White, 2001; Koppers &  
71 Staudigel, 2005; Carracedo et al., 2007; Carley et al., 2011; Mancini et al., 2015), but some  
72 ocean island volcanoes remain relatively poorly understood (e.g. the Azores, St Helena), and  
73 yet still pose a significant hazard to populations often living proximal to volcanic vents.  
74 Ascension Island, in the south Atlantic, is an example of the latter. It is small (subaerial  
75 dimensions of 8 km by 12 km), has no associated hotspot trace (cf. Hawaii, the Canaries; e.g.  
76 Zhao, 2004; Montelli et al., 2004) and has erupted magmas with a wide range of  
77 compositions (Daly, 1925; Coombs, 1963; Weaver et al., 1996; Kar et al., 1998; Jicha et al.,  
78 2013). This compositional diversity is matched by a diversity in eruption styles and range of  
79 volcanic deposits (Weaver et al., 1996; Kar et al., 1998; Hobson, 2001; Preece et al., 2016).

80 Evolved, more felsic, melts have the potential to generate significant hazards at ocean  
81 island volcanoes, and thus understanding where and how they evolve in the crust is  
82 imperative to forecasting future styles of eruption. The various means by which felsic  
83 magmas evolve have the potential to produce a range of compositions, which affects  
84 magmatic viscosity (Papale et al., 1998), and the amount of dissolved volatiles, all of which  
85 contribute to the mode of evacuation of magma from crustal storage regions (e.g.  
86 Eichelberger, 1995; Giordano et al., 2004). At ocean islands these silicic melts are  
87 considered to be generated via: anatexis of crustal material by hotter mafic melts (e.g.  
88 Sverrisdottir, 2007; Carley et al., 2011; Kuritani et al., 2011); extensive crystal fractionation  
89 from a more primitive mafic magma (e.g. Geist et al., 1995; Mungall & Martin, 1995; Larrea  
90 et al., 2014; Jeffrey et al., 2016); from direct derivation from mantle partial melting (Ashwal

91 et al., 2016) or through a combination of these processes (e.g. Bohrson & Reid, 1997;  
92 Wiesmaier et al., 2013; Sliwinski et al., 2015). Understanding the relative importance of these  
93 processes in any one setting has significant implications for understanding the relationships  
94 between the timescales of magma genesis, magmatic heat flux and potential triggering  
95 mechanisms of eruptions.

96 Ascension Island has produced more than 70 explosive eruptions of felsic magma in  
97 its ~ 1 Myr subaerial history (Preece et al., 2016), in addition to numerous eruptions that  
98 produced scoria cones, mafic lava flows, and felsic lava flows and domes. The erupted rocks  
99 have largely been used to investigate the origins of Ascension Island magmatism (e.g. Harris  
100 et al., 1982; Weaver et al., 1987, 1996; Weis et al., 1987; Kar, 1997; Kar et al., 1998; Paulick  
101 et al., 2010). As yet, little is understood about the magmatic plumbing system on Ascension  
102 Island and the control it exerts on magmatic composition and styles of eruptive activity. Thus,  
103 we focus here on the relationship between the mafic and felsic magmatism on Ascension,  
104 utilising whole rock major and trace element data, and a comprehensive suite of *in situ* crystal  
105 major and trace element data by EPMA (electron probe microanalyses) and LA-ICPMS (laser  
106 ablation inductively coupled plasma mass spectrometry). Samples studied here represent the  
107 products of 22 eruptions representing the full range in composition and eruptive styles  
108 presented by subaerial volcanism on Ascension Island. By combining these data with  
109 previous isotopic work and work on a compositionally-zoned fall deposit (Chamberlain et al.,  
110 2016), we present a model for the magmatic plumbing system of Ascension Island where  
111 felsic magmas evolve and stall in the lower crust, and highlight the role of crustal structure in  
112 the evolution of felsic melts on ocean islands with a low magmatic flux. Other potentially low  
113 magmatic flux ocean islands include the archipelagos of the Azores and Cape Verde islands,  
114 and thus the results of this study could be tested at other ocean islands around the world.

115

## 116 **ASCENSION ISLAND**

117 Ascension Island (7° 56' S; 14° 22' W) is located in the southern Atlantic Ocean, 90 km west  
118 of the Mid-Atlantic Ridge (MAR) and 50 km south of the Ascension Fracture Zone (AFZ;  
119 Fig. 1). Volcanism began on the sea bed 5 – 6 Myr ago, and subaerial volcanism has occurred  
120 from ~1 Ma to present (Kar et al., 1998; Minshull et al., 2010; Paulick et al., 2010; Jicha et  
121 al., 2013; Preece et al., 2016). The most recent eruption at Ascension Island has been dated  
122 using  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  dating to  $0.51 \pm 0.18$  ka (Preece et al., 2018) - with no evidence for pauses in  
123 eruptive activity of greater than 130 kyr throughout the 1 million years of subaerial volcanism  
124 (Jicha et al., 2013).

125

### 126 **Crustal structure of Ascension Island**

127 Ascension Island is built on 5 – 7 Myr old oceanic crust (Klingelhöfer et al., 2001; Paulick et  
128 al., 2010) on, or close to, the MAR. Due to the OIB-like trace element affinities of Ascension  
129 mafic lavas (Harris, 1983; Weaver et al., 1996), it has been suggested that magmatism at  
130 Ascension Island is the product of a shallow mantle plume, rising at the MAR then diverted  
131 along the Ascension Fracture Zone (AFZ, Fig. 1; Burke & Wilson, 1976; Montelli et al.,  
132 2006). However, seismic surveys have revealed a crustal structure that cannot be reconciled  
133 with a classic intraplate ocean island (i.e. a lack of lithospheric flexure, cf. Klingelhöfer et al.,  
134 2001), and instead suggest that significant growth of the Ascension Island edifice occurred on  
135 the MAR-axis. The seismic surveys show that the crust is 12 – 13 km thick under Ascension,  
136 with over-thickening of layer 3, to 7 km thick (Klingelhöfer et al., 2001). Active-source  
137 seismic tomographic studies failed to find evidence for magmatic underplating beneath the  
138 island, suggesting that the island's origins are not related to a hot spot (Evangelidis et al.,  
139 2004). Additionally, Evangelidis et al. (2004) located areas of anomalously high velocity

140 within the middle crust, which were inferred to be the crystallised remains of a relict magma  
141 chamber.

142

### 143 **Geochemical insights into the source of Ascension Island**

144 OIB-like trace element patterns have been measured from magmas erupted on Ascension  
145 Island, and spurred extensive research into the origins of magmatism, in particular its  
146 relationship to an undefined mantle anomaly and the proximal MAR. Sr and Nd isotopic data  
147 show little difference between the subaerial and the (volumetrically dominant) submarine  
148 products of Ascension:  $^{143}\text{Nd}/^{144}\text{Nd}$  varies between 0.51292 and 0.51310, and significant  
149 variation in  $^{87}\text{Sr}/^{86}\text{Sr}$  has been measured- between 0.70276 and 0.70656 (Fig. 2a; Weaver et  
150 al., 1996; Kar et al., 1998; Paulick et al., 2010). Submarine products have distinctly different  
151 Hf isotopic characteristics to the subaerial products studied here, and this has lead previous  
152 workers to suggest that the mantle source tapped by the submarine stage is no longer present  
153 (Fig. 2c; Paulick et al., 2010). There is little variation in Sr, Nd or Pb isotopic characteristics  
154 with time in the subaerial edifice (Kar, 1997; Paulick et al., 2010; Jicha et al., 2013), nor is  
155 the significant variation in  $^{87}\text{Sr}/^{86}\text{Sr}$  coupled to variations in  $^{143}\text{Nd}/^{144}\text{Nd}$  (Fig. 2a). The large  
156 variation in  $^{87}\text{Sr}/^{86}\text{Sr}$  has been suggested to reflect post-emplacement alteration of samples  
157 (especially evolved rocks) by seawater-derived groundwater fluids as the samples were not  
158 acid-leached prior to analysis (Kar et al., 1998; cf. Davidson et al., 1997). Due to the low Sr-  
159 contents of the Ascension magmas, they are particularly susceptible to alteration either  
160 through post-emplacement alteration, or by small degrees of assimilation of seawater-altered  
161 lithologies (Kar et al., 1998).

162 Previous studies utilised the trace element variation in basaltic magmas erupted on  
163 Ascension Island to infer source composition and to define three main magmatic groups,  
164 principally based on Zr/Nb ratios (Weaver et al., 1996; Kar, 1997). The oldest and most

165 voluminous magma type in submarine and subaerial Ascension is represented by the high  
166 Zr/Nb ( $Zr/Nb > 5.7$ ) mafic lavas (principally exposed on the south coast, Fig. 1; Weaver et  
167 al., 1996; Kar, 1997). Low Zr/Nb ( $Zr/Nb < 4.3$ ) lavas are relatively spatially restricted,  
168 outcropping only in the southwest (Fig. 1). Intermediate Zr/Nb lavas ( $Zr/Nb 4.3 - 5.7$ ) are the  
169 most common lavas in the subaerial history (but have erupted coevally with high and low  
170 Zr/Nb lavas), and dominate the northern and western regions (Fig. 1; Weaver et al., 1996;  
171 Kar, 1997; Jicha et al., 2013). The origins of these mafic lavas, and the process responsible  
172 for their variation has been related to varying degrees of partial melting of a consistent  
173 source, or melting of different mantle regions with differing mineralogy (Weaver et al., 1996;  
174 Kar, 1997; Jicha et al., 2013). Isotopic and trace element data on the mafic lavas types  
175 (Weaver et al., 1996; Kar et al., 1998; Paulick et al., 2010; Jicha et al., 2013), show that  
176 fractional crystallisation alone cannot reproduce the variability in Ti, Ta and Nb (Fig. 2c),  
177 and that variations in degree of partial melting, or source composition are more likely causes  
178 of this variability (Jicha et al., 2013). As the Zr/Nb ratios are more characteristic of  
179 variations during the initial production of magma (source lithology or degree of partial  
180 melting), and will not be used here to investigate the nature of the magmatic plumbing  
181 system.

182

### 183 **Evolution and distribution of volcanism at Ascension Island**

184 The magmatism on Ascension Island defines a transitional to mildly-alkaline, silica under-  
185 saturated array from olivine basalt - hawaiite - mugearite - benmoreite - trachyte - rhyolite  
186 (Daly, 1925; Weaver et al., 1996; Fig. 3). Mafic volcanic products are erupted all across the  
187 island (Fig. 1), while felsic products are limited to central and eastern areas (Fig. 1). Previous  
188 authors have divided the silicic eruptive products into two main centres: the older (Kar et al.,  
189 1998; Hobson, 2001; Jicha et al., 2013) central felsic region, which contains the oldest dated



190 exposed lava on the island (at 1094 ka, Jicha et al., 2013), and the younger eastern complex  
191 (youngest published Ar-Ar date of  $52 \pm 3$  ka; Jicha et al., 2013; Fig. 1). Felsic magmas are  
192 inferred to be the product of high degrees of fractional crystallisation (Fig. 2c; Weis et al.,  
193 1987; Kar et al., 1998; Webster & Rebbert, 2001; Jicha et al., 2013) originating chiefly from  
194 similar mafic melts to those erupted around the peripheries of the felsic complexes (both high  
195 and intermediate Zr/Nb basalts have been suggested as parental melts; Figs. 1, 2c; Weaver et  
196 al., 1996; Kar et al., 1998; Jicha et al., 2013).

197         The nature of the magmatic plumbing system on Ascension Island has not been  
198 established, yet fractional crystallisation is suggested as the dominant process for the  
199 formation of felsic melts (Kar et al., 1998; Jicha et al., 2013). There is only limited  
200 geochemical evidence for interaction between evolved magma batches to date (Kar et al.,  
201 1998; Chamberlain et al., 2016). Melt inclusion compositions and whole rock isotopic ratios  
202 of plutonic lithic clasts have been used to infer a genetic association between the plutonic  
203 lithics and the spectrum of volcanic rocks (Roedder & Coombs, 1967, Harris et al., 1982,  
204 Weis et al., 1987, Webster & Rebbert, 2001). These studies have suggested that the formation  
205 of the granitic plutonic bodies occurred at temperatures of 710 – 865 °C, and pressures of 200  
206 – 300 MPa (Webster & Rebbert, 2001, and references therein). Recent work on the crystal  
207 cargo and melt inclusions from a compositionally-zoned fall deposit on the island  
208 (Chamberlain et al., 2016) supports the hypothesis that felsic magma evolved through closed-  
209 system evolution on Ascension (as suggested by Harris, 1986, Weaver et al., 1987), with no  
210 petrological or textural evidence for magma mixing or multiple phases of stalling. We test the  
211 relevance of this model for the generation of all felsic magmas on Ascension Island, and use  
212 these data to improve our understanding of the temporal and spatial relationships between  
213 felsic and mafic volcanism.

214

## 215 **SAMPLING & METHODOLOGY**

216 The volcanic rocks on Ascension Island are extremely diverse and present evidence for  
217 Hawaiian, Strombolian and as well as more explosive (Subplinian to Plinian) eruptions, as  
218 well as phreatomagmatic activity. These products include lava flows and domes, pumice,  
219 scoria and ash fall deposits and pyroclastic density current deposits (Preece et al., 2016). The  
220 products of 22 representative eruptions that cover the full range in magmatic composition,  
221 eruptive style (Fig. 3) and the temporal and spatial extent of subaerial volcanism were  
222 analysed in detail to capture this range (Fig. 1; Table 1). Mafic lavas outcrop widely across  
223 the island and dominate its submarine history (see Nielson & Sibbett, 1996; Minshull et al.,  
224 2010). Mafic lava samples were selected to represent the main fields of mafic lava flows  
225 found on Ascension Island: the north coast (Sisters; samples AI14-411, AI14-471); the south-  
226 west region (Wideawakes; samples AI14-445, AI14-449); the south-east (Letterbox, AI14-  
227 423, AI14-429); the south coast (South Coast, AI14-514, AI14-522). In this way, the spatial  
228 and temporal variability (old vs. young lavas from the same eruptive centres) of mafic lavas  
229 on Ascension Island can be examined (Fig. 1; Table 1). Felsic samples come from effusive  
230 and explosive products, including samples from the older central felsic complex (AI-94, AI-  
231 103, AI15-621, AI14-459, AI14-488, AI14-493) and the younger eastern felsic complex  
232 (AI14-511, AI14-419, AI14-618, AI14-428, AI14-435, AI14-438). These samples were  
233 chosen to ensure a wide spatial and temporal sampling of the felsic volcanism (for individual  
234 sample names see Table 1).

235         Scoria and pumice samples were sieved to > 16 mm (or 8 mm if juvenile clasts were  
236 less than 16 mm) and all lithic clasts were removed by hand. These samples were then  
237 thoroughly cleaned by removing any adhering matrix or oxidised rind followed by soaking in  
238 (frequently changed) milli-RO water for a minimum of one week. Lava was treated by  
239 removing external, altered material then washed to remove any cutting fluid. All samples

240 were then dried thoroughly at 60 °C prior to crushing. An aliquot of each sample was selected  
241 to mill for X-ray fluorescence (XRF) analysis at the University of East Anglia (UEA) using a  
242 Bruker-AXS S4 Pioneer. For major elements (>0.5 wt.%) analyses of multiple international  
243 standards yielded uncertainties  $\leq \pm 0.5$  wt.% ( $2\sigma$ ), except for SiO<sub>2</sub> which yielded  
244 uncertainties of  $\pm 1.06$  wt.% ( $2\sigma$ ). Values of the standards compared with published values  
245 yielded accuracies within 2% for all major elements, except for MnO, CaO and P<sub>2</sub>O<sub>5</sub> where  
246 values were within 9% of published values; for full details of standards used, and their  
247 precision see Electronic Appendix 1. Trace element analyses of selected standards gave  
248 uncertainties < 5% ( $2\sigma$ ) for V, Cr, Ni, Cu, Zn, Rb, Sr, Y, Zr, Nb, Mo and Ba, with  
249 uncertainties <10% for As, Ce and U. For full details see Electronic Appendix 1.

250         The remaining material was prepared for *in-situ* analyses of major and trace elements  
251 in crystals and glass. Thin sections were made of lava samples, whilst pyroclastic samples  
252 were crushed, sieved into various size fractions (< 2 mm), before crystal and glass separates  
253 (from the 0.5 – 1 mm size fraction), were hand-picked and mounted into low-activity epoxy  
254 discs, and polished.

255         Prior to *in-situ* analyses, back-scattered electron (BSE) images were taken of all thin  
256 sections and epoxy blocks to identify crystal phases present, any zonation preserved in  
257 crystals and to locate suitable analytical spots. These images were obtained on a JEOL JSM  
258 5900LV scanning electron microscope (SEM) at UEA. Percentages of phases were calculated  
259 using ImageJ® software of transmitted light photomicrographs and BSE imagery, by filtering  
260 images based on colour or greyscale characteristics.

261         *In-situ* major element analyses were obtained by EPMA using a JEOL JXA 8230  
262 system at Victoria University of Wellington (VUW), or using a CAMECA SX100 at  
263 Edinburgh University, both using wavelength-dispersive spectrometry. Operating conditions  
264 varied depending on the phases and elements analysed, but precision of standard analyses of

265 major elements (>1 wt.% concentration) were always within 2 relative % (2 s.d.); with  
266 slightly higher uncertainties for minor elements (see Electronic Appendix 1 for further details  
267 on accuracy and precision of secondary standards). Due to their hydrous nature, only glass  
268 analyses with totals of <93 wt.% were set aside; values for the remaining analyses were then  
269 normalised to 100 %.

270 Trace element analyses of crystal phases and matrix glass were carried out at the  
271 University of Durham using a New Wave deep UV laser (193 nm solid state) coupled to an  
272 X-series 2 ICPMS (inductively-coupled plasma mass spectrometer). Analyses were run using  
273 a 35  $\mu\text{m}$  spot (for glass) or 50  $\mu\text{m}$  spot (for crystals). The LA-ICPMS data were internally  
274 normalized to  $^{29}\text{Si}$  or  $^{43}\text{Ca}$  from EPMA analyses. Abundances of single trace elements were  
275 calculated relative to a bracketing standard (NIST 612) which was analysed throughout the  
276 run under identical conditions. Precision and accuracies varied depending on the analytical  
277 conditions but generally have <10% (2 s.d.) uncertainties (see Electronic Appendix 1 for full  
278 details of precision and accuracy).

279

## 280 **RESULTS**

### 281 **Whole rock data**

282 The whole rock data confirm that the samples typify the full range in magmatic compositions  
283 exposed on Ascension Island, from the least-evolved Green Mountain scoria sample (AI14-  
284 552; 47.7 wt.%  $\text{SiO}_2$ , 3.01 wt.% Total Alkalis [TA]) through to the most-evolved sample of  
285 1094 kyr-old (AI-94; Jicha et al., 2013) felsic lava (72.3 wt.%  $\text{SiO}_2$ , 10.3 wt.% TA; Fig. 3a,  
286 Table 2). When selected whole rock trace element data are normalised to primitive mantle  
287 (Palme & O'Neill, 2003) felsic lava and pumice samples are clearly depleted in Sr, Ti and Ba  
288 relative to mafic lava and scoria samples (Fig. 3b). Felsic pumice and lavas are generally  
289 more enriched in incompatible trace elements than the mafic lavas and scorias (Fig. 3b).

290           The whole rock data show increasing total alkalis, and decreasing CaO and MgO,  
291 with increasing SiO<sub>2</sub> (Fig. 4). Mafic lavas are typically more-enriched in MgO, CaO and  
292 TiO<sub>2</sub> (Fig. 4a, c), and depleted in FeO, Na<sub>2</sub>O, K<sub>2</sub>O, Zr and Ba (Fig. 4b, d, f, Table 3), relative  
293 to felsic lavas. While having the textural characteristics of basaltic volcanism, samples of  
294 Letterbox ‘mafic’ lavas are in fact intermediate, with 55.8 – 57.6 wt.% SiO<sub>2</sub>, and 2.10 – 2.29  
295 wt. % MgO. Pumice and felsic lava samples have similar major and trace element  
296 concentrations (Fig. 3, 4), as do mafic lavas and scorias (Fig. 3, 4); thus there is no  
297 appreciable difference in whole rock compositions between magmas erupted effusively or  
298 explosively (Fig. 3, 4). Whilst there appear to be gaps within the MgO content (Fig. 4), these  
299 solely reflect the samples selected for study, when compared with a large published data set  
300 no gaps in composition are observed for Ascension Island volcanics (Fig.2c; Fig. 4 grey  
301 fields).

302

### 303 **Petrology of Ascension lavas**

304 Mafic and intermediate lavas studied are generally crystal poor, with less than 5%  
305 phenocrysts, apart from the samples from the South Coast (high Zr/Nb) lava with 18% and  
306 40% phenocrysts in the two samples studied here (see Table 3, Fig. 5a, b). In the mafic and  
307 intermediate lava flows, phenocrysts (> 500 µm) are predominantly plagioclase feldspar (Fig.  
308 5c, d), with minor olivine in some samples (Table 3, Fig. 5). Phenocryst phases in the crystal  
309 rich lavas (from the South Coast) are larger, up to 5mm, modal size 3mm, when compared  
310 with all other mafic and intermediate phases, where phenocrysts are generally < 1mm in  
311 diameter. The groundmass of all mafic and intermediate lavas studied here is  
312 microcrystalline, with no glass present. The groundmass consists of plagioclase feldspar,  
313 clinopyroxene, olivine and Fe-Ti oxides (Fig. 5a-d, Fig. 6) in decreasing order of abundance.

314 Felsic lava samples come from the (older) central felsic complex (AI14-103, AI15-  
315 621, AI-94; Fig. 1) and the (younger) eastern felsic complex (AI14-511, AI14-485, AI14-  
316 419, AI14-428). All felsic lavas are crystal poor, with <6% phenocrysts visible in hand  
317 sample (Table 3). Phenocrysts phases are dominantly ternary feldspars (plagioclase to alkali-  
318 feldspars, Fig. 7), with minor Fe-Ti oxides and aegirine-augite present as phenocrysts phases  
319 in two samples (Table 3). Phenocrysts are always <3 mm diameter. In thin section, crystals  
320 are largely euhedral, and feldspar crystals often present as clots of 2-5 crystals (Fig. 5e-h; Fig.  
321 6d, e). The groundmass is usually microcrystalline, consisting of ternary feldspar, cristobalite,  
322 interstitial aegirine-augite and Fe-Ti oxides in decreasing order of abundance (Table 3; Fig.  
323 6d, e, f). A single felsic lava sample has a glassy matrix (AI14-419, the Letterbox felsic lava  
324 sample), which has SiO<sub>2</sub> concentrations between 71.2 and 74.0 wt.% (Table 3; Fig. 11).

325

### 326 **Mineral compositions of the mafic and intermediate lavas**

327 Plagioclase feldspar is a dominant mineral phase in all mafic and intermediate lavas with  
328 compositions varying between An<sub>37</sub>Ab<sub>61</sub>Or<sub>2</sub> and An<sub>82</sub>Ab<sub>17</sub>Or<sub>1</sub> (Fig. 7). BSE images of  
329 feldspar crystals typically show faint oscillatory zoning (Fig. 6c) with the South Coast (high  
330 Zr/Nb) lavas having better developed zonation and resorbed cores (Fig. 6a, b). The lack of  
331 well-developed BSE image zonation patterns in the Sisters, Wideawakes and Letterbox  
332 feldspars (mafic lava fields, see Fig. 1 for name origins) is reflected in their major and trace  
333 element compositions which show limited variations (Fig. 7, 8). The South Coast lavas  
334 exhibit some compositional variation between core and rims in their feldspar population (Fig.  
335 8c, 9a), with cores typically being less-evolved than feldspar rims. Olivine compositions in  
336 the groundmass (or as phenocrysts in South Coast and Wideawakes samples) vary between  
337 Fo<sub>49</sub> – Fo<sub>89</sub>. Where groundmass olivine can be identified separately to phenocrysts,  
338 groundmass olivines have higher CaO and lower MnO concentrations at any given value of

339 Fo number. Olivine crystals are faintly normally-zoned, if they are zoned at all, with no  
340 evidence for resorbed cores in any samples studied (Fig. 6a, b, 9b, 10).

341 No variation is seen in phenocryst compositions between the stratigraphically older  
342 and younger samples from the same geographic regions. However, some variation in mineral  
343 compositions exists between different geographic regions, with differences in major and trace  
344 elements between geographic regions (Fig. 8a, c). Letterbox samples have more restricted  
345 feldspar and olivine compositions, representing the more-evolved end of the range in mineral  
346 compositions displayed by all mafic lavas (Fo 56 – 69; An 36 – 47; Fig. 8a; Figs. 9, 10a,  
347 10c), consistent with their whole rock compositions (see Table 2). South Coast, Sisters and  
348 Wideawakes lavas generally contain more variable An-plagioclase (An 32 – 82; see Table 5,  
349 Fig. 1, 8, 9), which has lower Sr concentrations (and extends to higher anorthite contents; Fig.  
350 8a, c), and more variable olivine (that extends to less-evolved compositions) than Letterbox  
351 (Fo49 – Fo89; Table 5, Fig. 1, 9, 10), consistent with their less evolved whole rock  
352 compositions.

353

#### 354 **Mineral compositions of the felsic lavas**

355 Ternary feldspar is the dominant mineral phase in all felsic lavas with compositions varying  
356 between  $An_{20}Ab_{75}Or_5$  and  $Ab_{70}Or_{30}$  (Fig. 7). BSE images of feldspar crystals from felsic lava  
357 samples are unzoned (Fig. 6d, e). However, some lava flows show core-rim-groundmass  
358 variations in feldspar compositions (see Fig. 8c and Electronic Appendix 2), which is  
359 unresolvable in BSE images alone. In particular, sample AI14-485 (from the eastern felsic  
360 complex; Fig. 1) has systematically more-evolved rims than cores, and more-evolved  
361 groundmass than rims. Feldspar compositions of felsic lavas range from andesine  
362 ( $An_{20}Ab_{75}Or_5$ ) through to anorthoclase ( $Ab_{70}Or_{30}$ ; Fig. 7, Table 4), with the most-evolved  
363 compositions ( $Ab_{70}Or_{30}$ ) present in both the older and younger felsic samples (see Fig. 8b;

364 Electronic Appendix 2). Most felsic feldspars (that are ternary) define a separate trend to the  
365 mafic and intermediate lava samples (with plagioclase feldspar only), having higher  
366 concentrations of Eu and Ba at any given Sr concentration. (Fig. 8d). Aegirine-augite is  
367 present as a groundmass phase in all felsic lava samples studied here, with compositions  
368 between 0.5 – 12.1 wt.% Na<sub>2</sub>O and 0.1 – 6.2 wt.% MgO. Aegirine-augite, typically < 500 μm  
369 (Fig. 11), commonly contains Fe-Ti oxide inclusions, and has higher Mn and lower Sr  
370 concentrations than the intermediate lavas (Fig. 11c), whilst the minor olivine (< 1%), present  
371 only in the Letterbox felsic lava, is Fo<sub>12.5</sub> (see Electronic Appendix 2).

372         The crystal compositions between samples of felsic lavas and samples of mafic to  
373 intermediate lavas do not overlap in their major elements. The samples from the younger  
374 eastern felsic complex (Fig. 1) have the greatest range in both feldspar (An<sub>20</sub>Ab<sub>75</sub>Or<sub>5</sub> –  
375 Ab<sub>70</sub>Or<sub>30</sub>) and clinopyroxene compositions (0.5 – 12.2 wt.% Na<sub>2</sub>O; Fig. 8b, 11a, b, Table 5,  
376 7), while crystals within samples from the central felsic complex are typically more-evolved;  
377 containing anorthoclase feldspar (An<sub>7</sub>Ab<sub>73</sub>Or<sub>20</sub> – Ab<sub>70</sub>Or<sub>30</sub>) and more aegirine-rich  
378 clinopyroxene (2.9 – 9.0 wt.% Na<sub>2</sub>O; Fig. 11a, b; Table 7).

379

### 380 **Petrological variation in mafic pyroclastic deposits**

381 Pyroclastic deposits are exposed all across the island and have compositions ranging from  
382 basalt to rhyolite, with no obvious Daly Gap when all published samples are considered  
383 (Daly, 1925, Fig. 3, 4). The scoriaceous deposits studied here are crystal poor, with  
384 phenocrysts of plagioclase feldspar and minor olivine, clinopyroxene and Fe-Ti oxides. The  
385 Green Mountain scoria (AI14-552) is a relatively voluminous eruption, with widespread  
386 deposits originating from within the central felsic complex. Its key identifying feature is the  
387 presence of abundant white-cream plutonic lithic clasts. In thin section, juvenile scoria  
388 typically contains plagioclase feldspar + olivine ± clinopyroxene ± Fe-Ti oxides. The



389 groundmasses of the scoria clasts from these deposits have varying proportions of microlites  
390 (feldspar + olivine).

391 In BSE imagery, crystal phases are not zoned, and preserve euhedral crystal habits.  
392 Feldspar compositions overlap those of both the mafic and felsic lavas, ranging between  
393  $An_{14}Ab_{72}Or_{14}$  and  $An_{84}Ab_{15}Or_1$ . The Green Mountain scoria has the largest variation in  
394 feldspar compositions (AI14-552; Table 5), but there are no systematic differences in core  
395 and rim compositions of individual crystals (Fig. 8c, Table 5). Olivine, the other major  
396 phenocrystic phase, overlaps compositionally with olivine in the mafic lavas, with Fo74 to  
397 Fo87 (Fig. 9b, 10b; Table 6) and also has no systematic difference in core and rim  
398 compositions from individual olivines (Fig. 10c). Matrix glass compositions are typical of  
399 mafic melts, with low  $SiO_2$  ( $< 50$  wt.%) and high CaO ( $> 6$  wt.%), (Fig. 12, Table 8); the bulk  
400 scoria composition is slightly less-evolved (45.8 – 49.3 wt.%  $SiO_2$ ) than the NE Bay scoria  
401 (AI14-438, 49.0 – 52.4 wt.%  $SiO_2$ ).

402

### 403 **Petrological variation in felsic pyroclastic deposits**

404 Pumice clasts are typically crystal poor, with  $< 5\%$  crystals. Ternary feldspar is the dominant  
405 crystal phase (oligoclase to anorthoclase composition  $\pm$  sanidine) with one ferromagnesian  
406 phase of either amphibole or fayalitic olivine, with minor magnetite  $\pm$  ilmenite  $\pm$  apatite, and  
407 rarely augitic clinopyroxene is present. One exception to this is the intermediate sample  
408 AI14-459 which has  $\sim 15\%$  crystals, with large amphibole (up to 3 mm) phenocrysts. Crystal  
409 phases are not zoned in BSE, and preserve euhedral crystal habits. Groundmass is typically  
410 glassy, with varying amount of feldspar microlites.

411 Feldspar compositions from these pumice fall deposits intersect and straddle those  
412 from the evolved effusive eruptions (Fig. 7, Table 4): with compositions ranging between  
413  $An_{51}Ab_{47}Or_2$  and  $An_{0.5}Ab_{58.5}Or_{41}$ . Some eruptions have relatively restricted feldspar

414 compositions (e.g. AI14-488:  $An_{0.5}Ab_{65.5}Or_{34} - An_1Ab_{69}Or_{30}$ ) whereas other extend over wide  
415 ranges (e.g. AI14-435:  $An_2Ab_{73}Or_{25} - An_{18}Ab_{76}Or_6$ ). In trace elements, the felsic pyroclastic  
416 feldspars plot within the higher- and lower-Eu trend (Fig. 8d). No clear core-rim relationships  
417 were found in feldspar compositions: only one pumice fall deposit in Middleton's Valley  
418 (AI14-459) has slightly less-evolved cores relative to rim compositions (cores:  $An_{51}Ab_{47}Or_2 -$   
419  $An_{20}Ab_{74}Or_6$ ; rims  $An_{43}Ab_{54}Or_2 - An_{20}Ab_{74}Or_6$ ). This comparatively crystal-rich sample (see  
420 Table 3) also bridges the divide between our felsic and mafic lava feldspar populations.  
421 Fosterite content in olivines (Fo1 – 57) from felsic pyroclastic samples mirrors the  
422 relationships between pyroclastic and effusive lava feldspars- intersecting with and extending  
423 the range in compositions of mafic lava sourced olivines (Fig. 10b, Table 6). Again, no  
424 differences between core and rim compositions are observed in olivine from all pyroclastic  
425 samples (Fig. 10c).

426         Glass compositions of felsic pyroclastic deposits range from ~ 66 wt.% to 75 wt.%  
427  $SiO_2$  (Fig. 12; Table 8). All sampled pyroclastic deposits have relatively homogenous glass  
428 compositions, with the one exception being the glass from the compositionally-zoned fall  
429 deposit, previously described in Chamberlain et al. (2016; Fig. 12a). In detail, individual  
430 sample variation in trace elements shows up to a four-fold variation in elements compatible in  
431 feldspar (Ba, Sr; Fig. 12b). However, limited variations in source-related trace elements (such  
432 as Th/U ratio; Fig. 12c) are evident between samples, where variation within a single sample  
433 is of the same magnitude or greater than variations between samples.

434

### 435 **Intensive variables**

436 Where available, two co-existing oxides (magnetite and ilmenite) were analysed and Fe-Ti  
437 oxide thermometry of Ghiorso & Evans (2008) was applied (if the pairs passed the  
438 equilibrium test of Bacon & Hirschman, 1988). Iron-titanium oxide derived temperatures

439 range from 932 °C to 1037 °C for the mafic lavas (Table 8). Oxygen fugacity varies between  
440 -0.45 log units relative to the Nickel-Nickel Oxide ( $\Delta$  NNO) buffer to +0.05 log units  $\Delta$   
441 NNO. Felsic lavas and pyroclastic samples yield Fe-Ti oxide temperatures ranging from 850  
442 °C to 960 °C, and  $fO_2$  ranging from -0.5 log units  $\Delta$  NNO to -2.3 log units  $\Delta$  NNO (Table 8).

443 Plagioclase-melt thermometry (Putirka, 2008) and alkali feldspar-melt thermometry  
444 (Putirka, 2008) was applied to all samples where equilibrium between feldspars and melt  
445 could be established (Putirka, 2008). Pressures of 330 MPa were assumed for mafic samples,  
446 as this is consistent with equilibration at the base of the crust, (Klingelhöfer et al., 2001) and  
447 pressures of 250 MPa were used for felsic samples, based on melt inclusion entrapment  
448 pressures modelled from samples of a zoned fall deposit on Ascension Island (Chamberlain et  
449 al., 2016). This modelling yielded temperatures within a similar range (but often higher) to  
450 that of the Fe-Ti oxide thermometry (Table 8) with modelled temperatures ranging between  
451 772 °C and 1034 °C for felsic samples, and between 1093°C and 1174 °C for mafic samples  
452 (Table 8). Although a specific pressure was assumed, testing demonstrated that the pressure  
453 effect is minimal, with less than 10 °C variation in estimated temperatures with > 200 MPa  
454 variation in assumed pressures.

455 Using measured alkali feldspar-melt compositions and temperatures modelled from  
456 plagioclase-melt thermometry, the alkali feldspar-melt hygrometer of Mollo et al. (2015) was  
457 also applied (see Table 8 for summarised results, full results in Electronic Appendix 2).  
458 Calculated water concentrations are high (average concentrations for the felsic units between  
459 4.66 wt.% and 8.12 wt.%, Table 8). Modelled water contents associated with the felsic lava  
460 and dome samples have consistently lower water concentrations than the explosively erupted  
461 samples. These high concentrations of water in explosive felsic samples are similar to those  
462 measured in Ascension Island melt inclusions (Chamberlain et al., 2016) and with the high

463 loss on ignition from felsic pyroclastic samples measured during XRF analyses (see  
464 Electronic Appendix 2 for full XRF results).

465

## 466 **DISCUSSION**

### 467 **Origin of compositional variations in Ascension Island magmas**

468 A range in whole rock and crystal compositions are evident in Ascension Island samples,  
469 with no clear differences observed between effusively erupted lava samples and explosively  
470 erupted pumice and scoria samples (Fig. 4, 7a). When considered alone, the variations in  
471 whole rock chemistry presented here (Fig. 4) do not give reason to disagree with previous  
472 whole rock studies of Ascension Island, which have suggested that fractional crystallisation is  
473 the dominant mechanism for producing evolved melts (e.g. Harris, 1983; Kar et al., 1998;  
474 Jicha et al., 2013; Chamberlain et al., 2016), evidenced by the continuous trend in major and  
475 trace element concentrations (Figs. 3, 4; Weaver et al., 1996; Kar et al., 1998; Jicha et al.,  
476 2013), and the lack of whole rock radiogenic isotopic ratio variation with major element  
477 concentrations (Fig. 2d; Weaver et al., 1996; Kar et al., 1998; Paulick et al., 2010).

478         Partial melting of mafic material is another process through which felsic melts have  
479 been proposed to be generated at ocean island volcanoes (e.g. Borhson & Reid, 1997;  
480 Sverrisdottir, 2007; Carley et al., 2011; Kuritani et al., 2011). Unlike other ocean islands,  
481 there is no evidence for divergence in Nd isotopic ratios between mafic and felsic melts (Kar  
482 et al., 1998), and major and trace elements vary co-linearly (Fig. 2b; Kar et al., 1998; Jicha et  
483 al., 2013) which previous studies suggest is not supportive of a partial melting origin for  
484 evolved melts at Ascension. Some minor assimilation and contamination has been inferred to  
485 be partly responsible for the radiogenic Sr isotopic compositions of more felsic melts (Kar et  
486 al., 1998), but this cannot be quantified based on the new data presented here, and it is noted  
487 that none of the earlier isotopic data was acid leached prior to analysis (cf. Davidson et al.,

488 1997). At other volcanic systems where partial melting has been shown to be a significant  
489 driver for the production of evolved magmas, magmatic compositions can be bimodal, with  
490 few erupted magmas of intermediate composition (e.g. Charlier et al., 2013, Meade et al.,  
491 2014), which is not observed at Ascension (Fig. 3, 4). However, small degree direct partial  
492 melting of a seawater-altered basaltic/gabbroic parent cannot be ruled out with our new data  
493 set for Ascension (cf. Kar et al., 1998), and may be responsible for some of the variation in  
494  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios (Fig. 2a).

495 Magma mixing between mafic and felsic magmas to produce the intermediate rocks  
496 could be responsible for the continuum in whole rock compositions at Ascension (Fig. 3), yet  
497 non-linear variations in major and trace elements in both whole rock compositions (especially  
498 in  $\text{TiO}_2$ ,  $\text{K}_2\text{O}$  and Zr, Fig. 4) and crystal phases (Fig. 8d, 10d) suggest that this is unlikely.  
499 Similarly, crystal compositions form coherent trends within individual samples, with no  
500 evidence for the distinct populations (cf. Geist et al., 1995; Troll & Schminke, 2002; Fig. 8 –  
501 11) that could represent growth in different magmas. Petrographic data shows no evidence of  
502 magma mixing, therefore fractional crystallisation is hypothesised to be the main mechanism  
503 for generation of felsic melts at Ascension Island, in agreement with previous authors (cf. Kar  
504 et al., 1998; Jicha et al., 2013).

505 To test the fractional crystallisation hypothesis further, MELTS modelling of isobaric  
506 liquid lines of descent (Gualda & Ghiorso, 2015) from the least evolved sample (AI14-438,  
507 see Table 1) has been undertaken at a range of pressures, from 330 MPa (the base of the  
508 crust, Klingelhöfer et al., 2001) to 90 MPa (~ 3.5 km depth), as well as an isothermal  
509 decompression model from 90 MPa to 1MPa (Fig. 13, 14). An initial starting composition  
510 from AI14-438 was used, with an assumed water concentration of 0.5 wt.%  $\text{H}_2\text{O}$ , and  $f\text{O}_2$  at  
511 the NNO-buffer based on Fe-Ti oxide thermometry (Table 8) and analyses of nearby MAR  
512 glasses (Almeev et al., 2008) which tap a mixed enriched Ascension Island-type source and a

513 depleted high  $\epsilon_{\text{Hf}}$  mantle (Paulick et al., 2010). It has been shown that oxygen fugacity is not  
514 solely fractionation dependent at Ascension Island (Chamberlain et al., 2016) and this one-  
515 step MELTS modelling does not properly reflect the fractionating phases at the more-evolved  
516 end. Thus, at  $T < 950$  °C the composition of AI15-621 was used, the least-evolved of the  
517 felsic lava samples, with an assumed water concentration of (5.1 wt.%  $\text{H}_2\text{O}$ ) from feldspar-  
518 melt hygrometry, and  $f_{\text{O}_2} -1.5 \Delta\text{NNO}$  from Fe-Ti oxide thermometry (Table 8).

519         The modelled liquid lines of descent of this two-step MELTS modelling reproduce the  
520 observed variations in whole rock composition (Fig. 13), with the models run at higher  
521 pressure (250 MPa or greater) having a better fit for MgO and CaO concentrations (Fig. 13b,  
522 c), suggesting fractionation at depths of more than 7 km. At the more evolved compositions  
523 (>68 wt.%  $\text{SiO}_2$ ), fractionation at 170 MPa better reproduces the more evolved compositions  
524 of Ascension Island magmas (Fig. 13a). Whilst isobaric fractionation is unlikely to occur at  
525 Ascension Island, the MELTS modelling shows that simple fractional crystallisation can  
526 replicate the observed variations in whole rock composition, when oxygen fugacity can be  
527 controlled to represent that measured in Ascension Island magmas. Similarly, modelled  
528 increases in  $\text{H}_2\text{O}$  with increasing  $\text{SiO}_2$  support the modelled high melt  $\text{H}_2\text{O}$  concentrations  
529 from feldspar-melt hygrometry (Table 8), especially at higher pressures of evolution (Fig.  
530 13d).

531

### 532 *Fractional crystallisation in mafic and intermediate magmas*

533 Fractional crystallisation, as modelled by MELTS, has demonstrated that the observed whole  
534 rock compositional range within the Ascension Island magmatic suite can be reproduced in  
535 this process. For the mafic and intermediate magmas, the crystal compositions measured  
536 overlap well with the modelled crystallising phases from MELTS (Fig. 14), and up to 70%  
537 total fractionation has occurred to produce the most evolved intermediate magmas (Fig. 13).

538 Whilst MELTS details proportions of phases fractionating, previous studies have  
539 demonstrated some limitations on the model's ability to accurately predict crystallisation of  
540 water-bearing phases (Gualda et al., 2012), clinopyroxene (Fowler & Spera, 2010) and apatite  
541 (Rooney et al., 2012). Therefore, to yield further insights into proportions of phases  
542 crystallising least-squares modelling of the major elements has been undertaken, constrained  
543 by using only phases observed in samples of Ascension Island volcanics.

544 Major element least-squares modelling was carried out using Petrograph (Petrelli et  
545 al., 2005, built on the least-squares modelling of Stormer & Nicholls, 1978), and to include  
546 entrainment where appropriate (cf. Kar et al., 1998, Electronic Appendix 3). A comparison of  
547 phases crystallising at each modelled stage between MELTS and Petrograph can be seen in  
548 Electronic Appendix 3. Two stages of evolution are considered initially, to attempt to  
549 reproduce the variations in mafic to intermediate whole rock compositions (Fig. 3): Stage 1)  
550 from the NE Bay Scoria (the most primitive basalt; AI14-438) to the Wideawakes (an  
551 intermediate Zr/Nb basalt marking the inflection in MgO vs TiO<sub>2</sub>, Fig 4c; AI-445); Stage 2)  
552 Wideawakes (AI14-445) to Letterbox (the intermediate lava; AI14-423). In both stages,  
553 compositions of plagioclase feldspar, olivine, clinopyroxene, ilmenite and magnetite  
554 measured in the parent sample were used. Apatite was also used as an accessory phase, given  
555 its presence as inclusions in mineral phases and the variation in P<sub>2</sub>O<sub>5</sub> evident in whole rock  
556 data (Table 2; full details of modelling conditions see Electronic Appendix 3). Whilst this  
557 modelling is limited by the assumption of uniform compositions of the fractionating phases,  
558 and has no pressure or temperature dependence, it provides a first order constraint on modal  
559 fractionating assemblage provided the sum of the squared residuals is < 2 (Stormer &  
560 Nicholls, 1978). Stage 1 fractionation modelling (sum of squared residuals [SSR] 0.25)  
561 suggests that ~ 8% fractionation of an assemblage dominated by plagioclase feldspar with  
562 subordinate olivine and apatite, coupled with the minor entrainment (4%) of clinopyroxene

563 and two oxides could produce the compositions of AI-445. The second stage of modelling  
564 (SSR 0.05), to produce the intermediate Letterbox magmas, suggests a further 56%  
565 fractionation (0.4 melt fraction remaining) of an assemblage dominated by feldspar and  
566 clinopyroxene, with minor olivine, magnetite, ilmenite and apatite (See Electronic Appendix  
567 3 for graphic representation; and comparison with modelled MELTS fractionating phases).

568         These modelled fractionating assemblages have been further tested using published  
569 distribution coefficients for Rb, Sr, Y, Zr, Nb and Ba in the fractionating phases (Fig. 15, full  
570 details and references for the distribution coefficients used is given in Electronic Appendix  
571 3). Generally fractional crystallisation (excluding any accumulation) can reproduce observed  
572 variations in Rb, Nb, Ba and Zr, with a poorer agreement with Sr and Y data (Fig. 15 and  
573 Electronic Appendix 3) for the variation observed in mafic magmas.

574

#### 575 *Felsic magma evolution*

576 Felsic magma evolution through closed system fractional crystallisation, modelled by  
577 MELTS, is shown to reproduce well the major elements of the whole rock data for Ascension  
578 in most elements (Fig. 13), and pressure of fractionation has a significant effect on total  
579 alkalis (Fig. 13a) reflecting the pressure-sensitivity of feldspar crystallisation (Fig. 14a).  
580 Fractionating assemblages again reproduce well the measured compositions of feldspar,  
581 olivine and clinopyroxene. In the more-evolved lavas, growth of low-pressure phases  
582 (evidenced by feldspars with ~4 – 6 wt.% K<sub>2</sub>O and Na-rich clinopyroxenes, Fig. 14a, c)  
583 reflect growth at a range of pressures (cf. isothermal decompression crystal compositions  
584 from MELTS modelling; Fig. 14) which are not observed in equivalent explosive deposits.

585         Again, to supplement the MELTS modelling, major element least-squares modelling  
586 was carried out using Petrograph (Petrelli et al., 2005; from Stormer & Nicholls, 1978). Two  
587 further stages of evolution are considered, to attempt to reproduce the trends observed in



588 whole rock concentrations (Fig. 3): Stage 3) Letterbox (AI14-423) to Devils Riding School  
589 (representative of the trachytic lavas on Ascension, Fig 3; AI15-621) and Stage 4) Devils  
590 Riding School (AI15-621) to Middleton's Ridge (most-evolved rhyolite, AI-94). In both  
591 stages, compositions of feldspars (plagioclase and ternary feldspar), olivine, clinopyroxene,  
592 ilmenite and magnetite measured in the parent sample were used. Apatite was also used as an  
593 accessory phase, given its presence as inclusions in mineral phases and the variation in  $P_2O_5$   
594 evident in whole rock data (Table 2; full details of modelling conditions see Electronic  
595 Appendix 3).

596         Stage 3 (SSR 1.23) requires a further 60% fractionation (0.16 melt fraction remaining  
597 from initial mafic starting sample) of an assemblage again dominated by plagioclase feldspar,  
598 with subordinate clinopyroxene, apatite, ilmenite, and minor entrainment of magnetite (1%).  
599 Contrastingly, MELTS modelling suggests both biotite and orthopyroxene should begin to  
600 fractionate (neither of which have been observed on Ascension Island, Electronic Appendix  
601 3). Stage 4 (SSR 0.08) shows a clear change in fractionating assemblage with ternary feldspar  
602 dominating the fractionating assemblage with clinopyroxene and magnetite, and requiring a  
603 further 57% fractionation (0.07 melt fraction remaining, in agreement with MELTS  
604 modelling of liquid lines of descent: 0.06 melt fraction remaining, Fig. 13). Minor  
605 accumulation (4%) of fayalite, ilmenite and apatite contributes to the evolution of the felsic  
606 magmas (see Electronic Appendix 3). These inferred fractionating phases are in good  
607 agreement with the observed crystal phases present in Ascension samples with feldspar being  
608 the dominant crystal phase present, with minor fayalitic olivine or clinopyroxene present,  
609 unlike MELTS modelling which again fails to accurately reproduce the observed mineral  
610 phases, with the suggested presence of leucite and orthopyroxene (Table 3, Electronic  
611 Appendix 3).

612 Trace element modelling of the proposed fractionating assemblages was undertaken  
613 for stages 3 and 4, again excluding any accumulation of phases. The results of this modelling  
614 show that Ba is successfully modelled by this fractionating assemblage, with moderate but  
615 less consistent results for modelled vs. measured Sr and Nb (Fig. 15, Electronic Appendix 3).  
616 There is poor agreement between modelled and measured Zr, Rb and Y (Fig. 15, Electronic  
617 Appendix 3). Zircon is observed in the most evolved samples, but fractionation of this phase  
618 has not been modelled as the proportions are challenging to quantify using major element  
619 modelling, although it could be the cause of the variations between observed and modelled  
620 concentrations of Zr and Y in the felsic magmas of Ascension. We note that accumulation or  
621 fractionation of minor phases, whilst not significant in terms of the major element evolution  
622 of the felsic melts, can significantly affect the trace element compositions of melts. With a  $K_d$   
623 of  $\sim 40$  in rhyolitic apatites (Pearce & Norry, 1979), Y concentrations will be significantly  
624 affected by even minor amounts of fractionation of this phase.

625

#### 626 *The role of ascent rate and oxygen fugacity*

627 Use of MELTS modelling to yield crystal compositions has highlighted the role of crystal  
628 growth in the upper crust during ascent in the presence or absence of phases in the felsic  
629 magmas. Moderate 4 – 6 wt.%  $K_2O$  feldspar and aegirine-augite are only found in felsic  
630 lavas; in felsic pyroclastic samples no feldspar has 4 – 6 wt.%  $K_2O$  and fayalite is the  
631 common ‘mafic’ phase (Fig. 10b)- rare clinopyroxene is augitic in composition, and reflects  
632 fractionation at pressures  $\geq 90$  MPa (Fig. 14c). Pyroclastic samples have high modelled water  
633 concentrations (Table 8) perhaps reflective of higher pressure storage (Fig. 13d, cf. Di Matteo  
634 et al., 2004; Brenna et al., 2014). Combining the lack of chemical zonation (Figs 9c, 10c), the  
635 euhedral nature of the phenocryst phases (Fig. 6), and no low-P phases forming (cf. Fig. 14c),

636 ascent rates for pyroclastic magmas, fractionating at high pressures ( $\geq 250$  MPa, c.f. Fig. 13)  
637 is likely to be rapid and warrants further comparison with their effusive counterparts.

638           In other alkaline systems it has been shown that variations in  $fO_2$  at constant  
639 temperatures can result in variable phase assemblages (White et al., 2009; Markl et al., 2010).  
640 Given the large variation in calculated  $fO_2$  within Ascension Island magmas (Table 8), some  
641 of the small variation around liquid lines of descent, and mismatch between the MELTS-  
642 modelled vs. observed phases, may stem from variable phase crystallisation at differing  $fO_2$   
643 conditions, as well as variations in ascent rate and depth to storage regions within the lower  
644 crust.

645

## 646 **Spatial and temporal variations in magmatism at Ascension Island**

### 647 *Temporal and spatial variation of mafic magmas*

648 Whole rock major and trace element data and crystal compositions show little variation  
649 between samples of older and younger lavas (Table 2, Fig. 4) from the same eruptive centres,  
650 suggesting that the mode of mafic magma generation and ascent has been relatively constant  
651 for the subaerial (and exposed) history of Ascension Island. This is in agreement with  
652 previous whole rock isotopic data, which shows no clear variation in the subaerial edifice of  
653 Sr, Nd or Pb isotopic ratios over time, indicative of source characteristics (Weaver et al.,  
654 1996; Kar, 1997; Paulick et al 2010; Jicha et al., 2013). While there is no temporal variation  
655 in the composition of mafic subaerial magmas erupted on Ascension, there are significant  
656 spatial differences in the phenocryst percentages, whole rock compositions, and crystal  
657 compositions, depending on the location of the eruptive centre (detailed in Table 1; Table 3,  
658 Figs. 4, 8, 10). South Coast lavas have the highest crystal contents (>17 %) and the least-  
659 evolved crystal compositions (down to An<sub>82</sub>, and Fo<sub>87</sub>; Fig 8, 10), whereas the Letterbox

660 samples from the SE of the island are intermediate in composition, contain clinopyroxene as  
661 a minor component (instead of olivine; Table 3), and more restricted feldspar compositions.

662

663 *Origin of the crystal cargo*

664 Given the marked differences in crystallinity between the South Coast lavas (AI14-522 and  
665 AI14-514; Table 1) and the other mafic to intermediate lavas (Table 3), modelling of  
666 plagioclase and olivine equilibrium compositions, based on the whole rock compositions at  
667 an assumed temperature and pressure, following the method of Price et al. (2012), was  
668 undertaken (Fig. 9). The range in olivine Mg# from different eruptive centres (Fig. 9) shows  
669 that not all of the olivine crystal cargo is modelled to be in equilibrium with the melt in which  
670 it is erupted (Fig. 9b). Rim analyses are just as likely to be in disequilibrium with the whole  
671 rock compositions as core analyses. This range in olivine Mg# shows that some crystals are  
672 not phenocrystic, despite the lack of significant overgrowths and the crystals mostly having a  
673 euhedral to subhedral habit (Fig. 6).

674 Similarly, plagioclase feldspar compositions are not in equilibrium with their whole  
675 rock compositions (Fig. 9a) and yet only feldspars from the South Coast lavas display  
676 reaction rims and anhedral cores (Fig. 6b). Whilst the calculation of equilibrium plagioclase  
677 compositions (following the method of Panjasawatwong et al., 1995) is only calibrated for  
678 plagioclase feldspar compositions (and not in alkaline systems such as Ascension), and thus  
679 could be a potential reason for why feldspars are in apparent disequilibrium, single samples  
680 preserve a wide variation in An content in the feldspars, showing that irrespective of the  
681 modelled equilibrium conditions, significant amounts of feldspar crystals will not be in  
682 equilibrium with their host rock composition.

683 Mafic lavas, excluding those from the South Coast group, are clearly in chemical  
684 disequilibrium with their feldspars, with less variation from the calculated equilibrium

685 composition for olivines (Fig. 9). Densities of the mafic magmas were calculated following  
686 the method of Bottinga & Weill (1970), at pressure of 330 MPa (the base of the crust  
687 (Klingelhöfer et al., 2001) using measured whole rock compositions and a water  
688 concentration of 0.5 wt.%. Densities of mafic magmas are on the order of  $2.7 \text{ g/cm}^3$ ,  
689 remarkably similar to that of plagioclase feldspar ( $2.6 - 2.7 \text{ g/cm}^3$ , Scoates, 2000; Ghiorso &  
690 Gualda, 2015), yet less dense than olivine and clinopyroxene, ( $\sim 3.6 \text{ g/cm}^3$  and  $\sim 3.2 \text{ g/cm}^3$   
691 respectively, Scoates, 2000; Ghiorso & Gualda, 2015). Thus, fractionating feldspar in a zone  
692 of magma storage will not sink, and instead may form lateral mushy cumulates as the magma  
693 evolves, whereas olivine and clinopyroxene have bigger density differences with the mafic  
694 magmas, and thus can sink more effectively. As the mafic magma reaches eruptible  
695 conditions, either due to concentration of volatiles within the magma (e.g. Stock et al., 2016),  
696 tectonic destabilisation of the system (e.g. Allan et al., 2012) or through gas injection (e.g.  
697 Caricchi et al., 2018), the mafic magma rapidly incorporates these lateral mushy feldspar  
698 antecrysts, producing the wide range in feldspar compositions found within single eruptions,  
699 that are in disequilibrium with their whole rock composition, yet native to the magmatic  
700 system (Fig. 9a, 14a). The timescale of incorporation of these antecrystic crystals prior to  
701 eruption must be short, as no zonation is observed (see Figs. 8 – 10), and feldspars generally  
702 retain their euhedral appearance (Figs. 5, 6). Similarly, the mushy storage regions in which  
703 the feldspars and mafic phases are forming are transient; no evidence is preserved for long-  
704 lived melt-dominant magma storage regions for mafic to intermediate magmas, with no  
705 evidence for incorporated crystals seeing more than one ‘triggering’ event (cf. Kahl et al.,  
706 2013).

707 *Role of crystal entrainment in mafic to intermediate magma genesis:* Excluding the  
708 South Coast group lavas, all other mafic and intermediate lavas have crystallinities less than  
709 5% (Table 3), with limited evidence for chemical zonation (Fig. 10c) and euhedral crystal

710 habits (Fig. 4, 6), thus incorporation of mushy antecrysts (which are subsequently resorbed)  
711 appears unlikely to constitute more than 5% (total rock crystallinity) of the rock volume. The  
712 similarities in antecrystic and phenocrystic crystal cargo in Ascension Island lavas make  
713 assessing the proportions of assimilated material challenging, as incorporation of these  
714 antecrysts may merely shift the whole rock compositions along the modelled liquid line of  
715 descent. This has implications for modelling the total amount of fractional crystallisation  
716 responsible for individual mafic and intermediate magmas, thus these estimates of degree of  
717 fractional crystallisation from both MELTS and the least-squares modelling must be treated  
718 with caution.

719         Lavas in the South Coast group have relatively high crystallinity compared with other  
720 mafic and intermediate lavas (Table 3), with crystals that show the most zoning in BSE  
721 imagery and major and trace element analyses (Fig. 6, 8), and feldspars that lie furthest from  
722 the modelled equilibrium compositions (Fig. 9). These South Coast lavas are also observed to  
723 contain significant proportions of plutonic lithic clasts (ranging from gabbros to syenites,  
724 Roedder & Coombs, 1967; Harris et al., 1982; Harris, 1983; Webster & Rebbert, 2001).  
725 These observations suggest that the range in compositions of feldspar and olivines found in  
726 South Coast lavas is the result of incorporation of large amounts crystals from other sources,  
727 as antecrysts or xenocrysts (Charlier et al., 2005). Potential sources of contaminant crystals  
728 are oceanic crust (thus crystals are xenocrystic), plutonic bodies related to Ascension Island  
729 magmatism (thus antecrysts), or from a mushy, not yet solid fractionated crystal residue (also  
730 antecrysts, as in the other mafic to intermediate lavas, above). Few analyses of crystals from  
731 the abundant lithic clasts exist, however feldspar compositions from gabbros overlap those  
732 found in the mafic lavas (Harris, 1983) and isotopic data suggests a cogenetic origin for  
733 gabbros and mafic lavas (Weis et al., 1987). Contrastingly, ocean crust feldspars define a  
734 lower  $K_2O$  trend than alkaline-magmatism-sourced feldspars when plotted against An content

735 (Davis et al., 2007). No difference is seen in the feldspar and olivine compositions of the  
736 South Coast lavas (Fig. 7 – 10), suggesting that incorporated crystals are antecrystic (and not  
737 xenocrystic, see representative compositions of Mid-Atlantic Ridge feldspar crystals on Fig.  
738 8b) in origin. No evidence is seen in crystal compositions or textures for mixing with a  
739 chemically distinct magma, thus mechanical incorporation of antecrystic plutonic material  
740 (e.g. Humphreys et al., 2009) is our favoured method by which these materials are included  
741 within the South Coast lavas, on top of the ‘background’ accumulation of mushy antecrystic  
742 material similar to the other mafic and intermediate lavas.

743 South coast lavas are the only samples studied here which show overgrowths on  
744 feldspars (Fig. 6b), glomerocrystic textures (Fig. 5b) and increased crystallinity, suggestive of  
745 higher degrees of antecryst incorporation. The relative importance of antecrysts in south coast  
746 lavas could be due to these south coast magmas having a greater ability to erode these deep  
747 mafic plutonic bodies upon ascent, or that that these deep mafic plutonic bodies are only  
748 present in the source and ascent region of the crust through which the south coast lavas travel  
749 prior to eruption. A magma’s ability to mechanically disaggregate any lithic fragment is  
750 dependent upon its enthalpy and composition (Glazner, 2007). The variations in whole rock  
751 compositions between south coast lavas and other mafic lavas is not significant (Fig. 4), thus  
752 only variations in temperature could change the magmas ability to erode and disaggregate the  
753 plutonic lithic fragment. Modelled feldspar-melt temperatures are similar across the suite of  
754 mafic lavas (Table 8) and so it seems unlikely that increased antecryst incorporation is due to  
755 an increased ability of south coast magmas to erode plutonic lithic fragments. Instead, the  
756 increased proportion of antecrysts in south coast lavas is suggested to reflect an increased  
757 presence of mafic plutonic rocks at depth in this region. These plutonic rocks are anteliths,  
758 related to Ascension Island generation and not oceanic crust plutonic rocks (Fig. 8b), and

759 highlight the lateral variability in plutonic or mushy regions within the Ascension Island  
760 crust.

761

762 *Crustal control on the location of felsic eruptive centres*

763 Eruptions of felsic magma occurred from two main centres: the older central felsic complex,  
764 and the younger eastern felsic complex with mafic magmas erupted around the peripheries of  
765 these centres (Fig. 1; Jicha et al., 2013). In these regions, felsic magmas - erupted as lavas  
766 and pyroclasts - are the dominant magma type. Spatial separation of mafic and felsic lavas  
767 has been seen at other ocean islands (both active and extinct), and has been suggested to be a  
768 result of crustal structure inhibiting the ascent of more dense mafic magmas in the felsic  
769 complexes (e.g. Mahood & Hildreth, 1983; Druitt et al., 1995; Carracedo et al., 2007; Brenna  
770 et al., 2015).

771 Active-source tomography reveals an area of elevated seismic velocities in the core of  
772 the island, at 6.5 km depth beneath the felsic complexes (Evangelidis et al., 2004). This was  
773 interpreted to represent a single crystallised magma body, supported by the presence of  
774 plutonic lithic clasts in the erupted products (e.g. Roedder & Coombs, 1967; Harris, 1986;  
775 Weis et al., 1987; Hobson, 2001; Webster & Rebbert, 2001). This central core of nested  
776 plutonic rocks (of gabbroic through to granitic compositions – Roedder & Coombs, 1967)  
777 under the felsic complexes could inhibit the ascent of almost all mafic magmas in these  
778 regions.

779 The presence of plutonic rocks in ocean island crust has long been known to affect the  
780 evolution of magmas prior to eruption—the volcanic islands of Terceira (Mungall & Martin,  
781 1995) and Oki-Dōzen (Brenna et al., 2015), among others, have evidence for central felsic  
782 complexes (both as plutonic rocks, and as storage regions for felsic volcanic eruptions) which  
783 inhibit the eruption of mafic magmas. Ascension Island has a modelled growth rate of 0.4



784 km/Myr (Minshull et al., 2010), which is significantly lower even than post-shield Hawaii  
785 (0.9 km/Myr, Minshull et al., 2010 and references therein) and dramatically lower than shield  
786 stage Mauna Kea (8.6km/Myr, Minshull et al., 2010 and references therein). Therefore we  
787 suggest that this ‘filtering’ of mafic magmas and spatial segregation of mafic and felsic  
788 eruptive centres could be a result of low rates of magmatic flux. If magmatic flux is higher,  
789 this could destabilise the central felsic systems, and mafic and felsic magmas would no  
790 longer show such clear spatial separation, and mixing textures would be much more  
791 dominant, such as those observed in Tenerife (Sliwinski et al., 2015) or Iceland (Carley et al.,  
792 2011), where mafic and felsic magmas are still produced, but with less clear spatial  
793 separation and increased role of partial melting of crustal material than that inferred at  
794 Ascension.

795         We suggest that multiple plutonic bodies representative of multiple ephemeral magma  
796 reservoirs, rather than a single magma body, are necessary to generate the range in  
797 compositions seen on Ascension Island as there is no evidence from crystal compositions for  
798 repeated use of a single magma storage region which is repeatedly rejuvenated (cf. Kahl et  
799 al., 2013). This observation suggests the ‘plutonic body’ identified seismically (Evangelidis  
800 et al., 2004) beneath Ascension is a series of smaller-volume, nested plutonic bodies which  
801 cannot be resolved seismically from a single large body in the same dimensions (Bauer et al.,  
802 2003).

803         The presence of plutonic lithic clasts in pyroclastic fall deposits on Ascension Island  
804 (including some of the oldest fall units on the island, Hobson, 2001), combined with the  
805 rhyolitic nature of the oldest-dated subaerial lava (Jicha et al., 2013), implies that the plutonic  
806 complex was established well before the beginning of the subaerial phase of Ascension  
807 Island’s volcanic history. While the felsic plutonic and volcanic rocks share an apparently  
808 common origin (with similar isotopic characteristics, Weis et al., 1987 and following the

809 same liquid line of descent, Harris et al., 1982) the timing of plutonic formation must pre-date  
810 the earliest subaerial eruptions on the island (1094 ka; Jicha et al., 2013), and could relate to  
811 the more voluminous submarine volcanism that began ~5 – 6 Myr ago (Minshull et al., 2010;  
812 Paulick et al., 2010).

813

#### 814 **The magmatic plumbing system**

815 Here we combine our new results with all previously published data to present a model for  
816 the magmatic plumbing system underlying Ascension Island (Fig. 16) during the subaerial  
817 phase of activity from ~1 Ma to present. The key features of the model are as follows:

- 818 1) A zone of melt extraction from which all Ascension Island volcanism is sourced, at  
819 varying degrees of partial melt (Fig. 16; Paulick et al., 2010; Jicha et al., 2013). This  
820 melt extraction zone is geochemically distinct from the mantle melting responsible for  
821 the significantly larger volume submarine volcanism of Ascension which was active  
822 from ~5-6 Ma to ~3 Ma (Minshull et al., 2010; Paulick et al., 2010).
- 823 2) Heterogeneity in the lower crust affects the ascent and incorporation of antecrysts in  
824 basaltic melts. The variably over-thickened layer 3 (lower crust) presently underlying  
825 Ascension Island (Klingelhöfer et al., 2001) likely formed during the large volume,  
826 on-axis volcanism which built the submarine edifice (Minshull et al., 2010). We  
827 suggest that the over-thickening of layer 3 (represented mainly by mafic plutonic  
828 lithic clasts, not the nested felsic plutonic lithic clasts) is most significant underneath  
829 the vents for the South Coast lavas (AI14-514, AI14-522). South Coast lavas contain  
830 many crystals that are not in equilibrium with their whole rock compositions (Fig. 9),  
831 suggesting that these magmas have mechanically incorporated large numbers of  
832 antecrystic feldspar and olivine, shortly prior to eruption. This spatial heterogeneity in

833 deep mafic plutonic rocks is reflected in the proportions of antecrysts incorporated  
834 during ascent of mafic magmas.

835 3) A central nested region of small discrete plutonic bodies of varying composition (of a  
836 higher proportion of felsic bodies), which forces the majority of ascending mafic  
837 magmas to stall and fractionate until they reach a level where their buoyancy has  
838 increased enough to allow them to ascend further. This is supported by the felsic  
839 plutonic lithic clasts within the Green Mountain scoria, erupted in the central felsic  
840 complex, and by mechanically incorporated plutonic lithic fragments in many felsic  
841 pyroclastic deposits (Fig. 16).

842 4) Felsic magma evolution occurs only in the lower crust at pressures greater than 170  
843 MPa (Fig. 13). No evidence is seen for magmatic stalling (and subsequent eruption)  
844 of felsic melts stored at pressures less than 170 MPa. The only modelled entrapment  
845 pressures from melt inclusions from a zoned fall deposit on Ascension Island showed  
846 that crystals grew in a storage region at 250 MPa (Chamberlain et al., 2016), with  
847 melt inclusions having up to 4 wt.% H<sub>2</sub>O. This is in agreement with modelled  
848 entrapment pressures from felsic plutonic bodies being between 200 and 300 MPa (or  
849 6.8 – 10.2 km, assuming a crustal density of 3000 kgm<sup>-3</sup>; Webster & Rebbert, 2001).  
850 The modelled high water concentrations of melts from which feldspars have grown  
851 (Table 8) could support this storage and evolution within the lower crust (or Layer 3  
852 of Klingelhöfer et al., 2001; Fig. 13, 16). Lower crustal storage means that any unrest  
853 signals will be harder to resolve, and has implications for the methods implemented to  
854 monitor future volcanic unrest on Ascension Island.

855 5) Magmas are erupted as discrete batches, with no evidence for long-lived storage, or  
856 for crystal recycling between eruptions in a mushy or melt-dominant magma storage  
857 region (cf. Kahl et al., 2013). Magma mixing is therefore not an eruptive trigger.

858            Instead, triggers could be internal (due to volatile over-pressure), or from external  
859            tectonic changes, which again presents challenges for monitoring potential future  
860            volcanic unrest at Ascension Island.

861            6) The magmas that fed explosive and effusive felsic eruptions exhibit differences in the  
862            presence or absence of low pressure phases (Fig. 14) perhaps suggestive of variation  
863            in the location of their storage regions and ascent rates, and warrants further  
864            investigation. However, fractional crystallisation (with minor crystal entrainment) of  
865            predominantly feldspars induced by stalling (at varying depths) in the nested (more  
866            felsic) plutonic region, remains the dominant evolutionary process for all felsic  
867            magmas, irrespective of the eruptive style.

868

#### 869    **Implications for the generation of felsic magmas on ocean islands**

870    The petrogenetic processes responsible for the generation of felsic magmas in ocean island  
871    volcanoes built on thin oceanic crust have long been debated. Many authors suggested that  
872    felsic magma production at ocean island volcanoes could be a proxy for the initiation of  
873    continental crust formation in the Archean (e.g., Gazel et al., 2014; Mancini et al., 2015).  
874    While the low growth rates of Ascension Island (Minshull et al., 2010) negate its use in  
875    understanding Archean felsic magma genesis, the dominance of fractional crystallisation in  
876    the generation of felsic magmas, with little evidence for magma mixing, is unusual when  
877    compared with other ocean island volcanoes (e.g., the Canary Islands and Iceland: e.g.  
878    Borhson & Reid 1997; Caroff et al., 1999; Sverrisdottir, 2007; Carley et al., 2011; Longpré et  
879    al., 2014).

880

881    *Open vs. closed system evolution*

882 The new data presented here suggests evolution of Ascension Island magmas is dominated by  
883 simple crystal fractionation in a relatively closed system of short-lived discrete storage  
884 regions, and supports conclusions of previous workers (Harris et al., 1982; Kar et al., 1998;  
885 Webster & Rebbert, 2001; Jicha et al., 2013; Chamberlain et al., 2016). This is contrary to  
886 many other ocean island volcanoes where open system processes such as magma mixing and  
887 significant partial melting of oceanic crust contribute to the formation of evolved magmas,  
888 and where there is less evidence for closed system fractional crystallisation (Bohrson & Reid  
889 1995, 1997; Caroff et al., 1999; Carley et al., 2011; Weismaier et al., 2013; Longpré et al.,  
890 2014; Sliwinski et al., 2015). The main factor controlling whether open or closed system  
891 behaviour dominates could be the magmatic flux: it has been shown that increased magma  
892 fluxes correspond to increased degrees of crustal assimilation and crystal entrainment at mid  
893 ocean ridges (Michael & Cornell, 1998). Ascension Island has an order of magnitude slower  
894 growth rate when compared with Hawaii (Sharp & Renne, 2005; Minshull et al., 2010), and  
895 thus we infer a significantly lower magmatic flux, which led to the development of small-  
896 scale, short-lived magma storage regions, and closed system evolution of felsic magmas.  
897 Multiple volcanic centres also display evidence for relatively closed system evolution of  
898 mafic magmas by extensive fractional crystallisation to produce alkaline felsic magmas (e.g.  
899 Volcán Alcedo in the Galápagos [Geist et al., 1995], Terceira, São Miguel and Graciosa in  
900 the Azores [Mungall & Martin, 1995; Larrea et al., 2014; Jeffrey et al., 2016], and the extinct  
901 Oki-Dōzen volcano, Japan [Brenna et al., 2015]). These islands are all likely the result of low  
902 magmatic production rates, and are likely not related to a deep-seated mantle hotspots  
903 (Hildenbrand et al., 2014; Métrich et al., 2014). Fractional crystallisation always requires the  
904 formation of a large volume of plutonic rocks as a by-product of extensive fractional  
905 crystallisation. Erupted lithic fragments or surface exposures provide evidence for these

906 plutonic rocks (Mungall & Martin, 1995; Larrea et al., 2014; Brenna et al., 2015; Jeffrey et  
907 al., 2016).

908

909 *Fractional crystallisation: the space problem*

910 Extensive fractional crystallisation of > 80% mafic melt to produce the felsic magmas seen  
911 on small ocean islands such as Ascension Island and the Azores implies that significant  
912 volumes of plutonic material remain in the crust. While eruptive volumes are unconstrained  
913 at Ascension Island due to high erosion rates and dispersal over the ocean, the high degrees  
914 of fractionation responsible for a single body of felsic magma evolution suggest that  
915 significant volumes of plutonic rocks remain in the crust. Taking the evolution from NE Bay  
916 scoria to Middleton's Ridge rhyolite as an example, in total the rhyolite represents only ~9%  
917 of the original mass of basalt. If we assume a modest eruptive volume of 0.2 km<sup>3</sup> (compared  
918 with the AD 1630 eruption from São Miguel which evacuated ~0.85 km<sup>3</sup> of magma [DRE,  
919 Cole et al., 1995]), then ~2 km<sup>3</sup> of fractionated crystals, preserved as plutonic rocks, remain  
920 in the crust. Erupted volumes and degree of fractionation vary between eruptions, yet as over  
921 70 eruptions of felsic magma have occurred over the last 1 million years on Ascension  
922 (Preece et al., 2016), then a minimum of ~140 km<sup>3</sup> of plutonic rocks may remain in the crust,  
923 with no evidence for their rejuvenation preserved in any volcanic products on Ascension  
924 Island. This could be viewed as a minimum volume, given the lack of constraints on eruptive  
925 volumes, and the unknown number of magmatic (not necessarily eruptive) events. Evidence  
926 for a central nested plutonic core (Evangelidis et al., 2004) and over-thickening of layer 3  
927 (where magmatic evolution is interpreted to occur at Ascension Island, see above) has been  
928 observed in seismic reflection surveys (Klingelhöfer et al., 2001). Evangelidis et al. (2004)  
929 suggest that the central high velocity region, inferred to consist of plutonic rocks (of  
930 unknown compositions), could have a volume approaching 7000 km<sup>3</sup>. This appears large

931 compared to our minimum estimates (above), however small discrete bodies of plutonics of  
932 varying composition would not be resolvable from the seismic reflection surveys, and  
933 therefore this volume represents an area affected by plutonism, not necessarily the volumes of  
934 individual plutonic bodies. Aside from over-thickening of layer 3 (Klingelhöfer et al., 2001),  
935 it remains unclear how such large potential volumes of plutonic rocks are accommodated in  
936 thin oceanic crust, yet their presence seems vital for the formation of evolved melts in low  
937 magma flux ocean island volcanoes.

938

## 939 **CONCLUSIONS**

- 940 • There are no differences in whole rock or crystal compositions from the lavas erupted  
941 in the same spatial regions of the island, suggesting that in the last 1 Myr the  
942 processes controlling mafic melt production and eruption have remained constant,  
943 even if source region composition and degree of partial melting have changed (cf.  
944 Jicha et al., 2013).
- 945 • Spatial variation in crystallinity and in the composition and origin of crystals between  
946 mafic eruptive centres highlights heterogeneity in the lower crust under Ascension  
947 Island. The mafic magmas which erupted along the South Coast region, as well as the  
948 Green Mountain scoria sample in the felsic complexes incorporated higher volumes of  
949 antecrysts during ascent, consistent with the observation of increased numbers of  
950 coherent plutonic lithic fragments in these lava flows.
- 951 • Felsic melt evolution at Ascension Island is dominantly controlled by crystal  
952 fractionation of ternary feldspar with minor fayalite or sodic clinopyroxene,  
953 dependent upon ascent rate and oxidation state of the magma, with isotopic evidence  
954 of minor crustal assimilation of seawater-altered crustal material (Kar et al., 1998).  
955 There is no evidence for magma mixing in any of the eruptive deposits studied here,

956 indicating that while a wide range of magma compositions are erupted across a ~12  
957 km diameter island, they do not encounter other melts during their transport through  
958 the crust. Magmatic evolution occurs in small, short-lived, isolated magma storage  
959 regions in the lower crust. Antecrysts do not show reaction rims, and therefore their  
960 incorporation is purely mechanical and occurred late (during ascent).

961 • Alkali feldspar-melt hygrometry reveals high water concentrations of up to 8 wt.%  
962 H<sub>2</sub>O in the more-evolved magmas. These high H<sub>2</sub>O concentrations, combined with  
963 the lack of evidence for an external trigger in the crystal chemistry suggests that  
964 internal over-pressure from high degrees of fractional crystallisation concentrating  
965 H<sub>2</sub>O in the liquid phase is a likely eruptive trigger for explosive eruptions on  
966 Ascension Island.

967 • Felsic magmas are almost entirely erupted in the central and eastern areas of  
968 Ascension Island implying a spatial control on the evolution of felsic melts. We infer  
969 that the presence of significant volumes of plutonic bodies at depth inhibits magma  
970 ascent in these regions, and allow magmas to evolve and incorporate antecrysts.  
971 While the origin and nature of these plutonic bodies is not yet known, they must pre-  
972 date the subaerial stage of Ascension Island volcanism. The presence of plutonic  
973 rocks as abundant lithic clasts in the central and eastern regions highlights the  
974 importance of heterogeneous crustal structure in the evolution of magmas in thin  
975 young oceanic crust at Ascension Island.

976

## 977 **ACKNOWLEDGEMENTS**

978 In memory of Jon Davidson (1959–2016). We thank Marc Holland, the Ascension Island  
979 Government, the Heritage Society, the Conservation Department and island residents (in  
980 particular Drew Avery) for their logistical support during field seasons. George Cooper, Chris



981 Hayward, Ian Schipper, Bertrand Lezé and Chris Ottley are thanked for their laboratory and  
982 technical assistance during the course of analyses for this project. Vibrant discussions with  
983 Ben Cohen, Julia Crummy, Bridie Davies, Sue Loughlin, Darren Mark, Jane Scarrow, Fin  
984 Stuart, Charlotte Vye-Brown and Barry Weaver helped in formulating our ideas. Constructive  
985 and considered reviews by Marco Brenna, Felix Genske, Karen Harpp and an anonymous  
986 reviewer, and the expert editorial handling of Wendy Bohrson have led to significant  
987 improvements to this manuscript.

988

### 989 **FUNDING**

990 This project was funded by a Leverhulme Trust Research Project Grant (RPG-2013-042),  
991 with the second field season supported by a Gloyne Outdoor Geological Research award  
992 from the Geological Society of London.

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1317 **FIGURE CAPTIONS**

1318 Figure 1: Geological map of Ascension Island, located 90 km from the Mid Atlantic Ridge  
1319 and between the Ascension Fracture Zone (AFZ) and Boca Verde Fracture Zone  
1320 (BVFZ; inset), adapted from Weaver et al. (1995), Paulick et al. (2010) & Chamberlain  
1321 et al. (2016). Sample locations indicated by triangles, diamonds and pentagons;  
1322 eruptive centres identified in grey font. Surface expressions of known faults outlined in  
1323 red. Major lava flow boundaries are indicated with black lines in the mafic lava flows.

1324 Figure 2: (a) Radiogenic isotope variations in Ascension Island lavas (subaerial unless  
1325 otherwise stated); (b) Trace HFSE variation with major element variations; (c) Adapted  
1326 from Jicha et al., 2013, trace element variation in Ascension lavas, showing the  
1327 variation due to source heterogeneity, differentiation, and Fe-Ti oxide controlled  
1328 fractionation paths; (d) variation in  $^{143}\text{Nd}/^{144}\text{Nd}$  with degree of evolution (MgO). All  
1329 data presented is a combination of Weaver et al. (1995, grey diamonds), Kar et al.  
1330 (1998, orange diamonds), Paulick et al. (2010, blue squares) and Jicha et al. (2013,  
1331 crosses).

1332 Figure 3: (a) Whole rock compositions of selected samples compared with all known  
1333 Ascension Island whole rock data in grey field- (data from Weaver et al., 1996, Kar et  
1334 al., 1998, Jicha et al., 2013). Diagram adapted from LeMaitre et al. (1989). (b)  
1335 Primitive mantle (Palme & O'Neill, 2003) normalised trace element ranges for mafic-  
1336 intermediate lavas (purple), scoria samples (orange), pumice samples (yellow) and  
1337 felsic lava samples (blue).

1338 Figure 4: Whole rock variation in major and trace elements for all studied samples of  
1339 Ascension Island mafic-intermediate lavas (purple triangles), felsic lavas (blue  
1340 pentagons) and pyroclastics both pumiceous (yellow diamonds) and scoriaceous

1341 (orange diamonds). Grey fields represent the full range of compositions found at  
1342 Ascension from Weaver et al. (1995). For full data set, see Electronic Appendix 2.

1343 Figure 5: Photomicrographs of Ascension lava samples. (a, b): Young South Coast lava  
1344 sample showing a glomerocryst of feldspar and olivine; (c, d): Young Sisters lava  
1345 sample showing euhedral plagioclase feldspar in a microcrystalline matrix; (e, f):  
1346 Cricket Valley lava with sodic feldspars and minor aegirine-augite in a microcrystalline  
1347 groundmass; (g): Letterbox felsic lava with large ternary feldspar phenocrysts in a  
1348 microcrystalline groundmass; (h): Ariane lava flow with ternary feldspars and aegirine-  
1349 augite in a microcrystalline groundmass. Images (a, c, e, g) in plane-polarised light;  
1350 images (b, d, f, h) in cross-polarised light. Sample numbers relate to those listed in  
1351 Table 1 and numbers in square brackets show whole rock SiO<sub>2</sub> concentrations.

1352 Figure 6: Back Scattered Electron (BSE) images of Ascension Island lavas. Sample numbers  
1353 relate to those listed in Table 1 and numbers in square brackets show whole rock SiO<sub>2</sub>  
1354 concentrations. Features labelled in red refer to A: Reverse-zoned feldspar; B:  
1355 Normally-zoned olivine; C: Faint oscillatory-zoned plagioclase. D: Mafic lava  
1356 groundmass, typically consisting of feldspar + olivine + magnetite ± clinopyroxene ±  
1357 ilmenite; E: Unzoned feldspar typical of felsic products; F: Unzoned fayalitic olivine;  
1358 G: Groundmass of felsic lavas flows consists dominantly of feldspar with minor  
1359 interstitial aegirine and fayalite; H: Dark in BSE cristobalite with characteristic fish-  
1360 scale textures replacing groundmass and some feldspar crystals.

1361 Figure 7: Ternary feldspar diagrams showing all feldspars differentiated by composition and  
1362 eruptive style. For full data set see Electronic Appendix 2.

1363 Figure 8: Compositions of feldspars from (a) mafic to intermediate lavas; (b) felsic lavas and  
1364 (c, d) all Ascension Island samples. Sample areas relate to those used in Table 1. For  
1365 full data set see Electronic Appendix 2. The purple field on panel (b) shows the



1366 compositions of feldspars from mafic to intermediate lava samples. The pink field on  
1367 panel (b) shows the Mid Atlantic Ridge ocean crust feldspars from the Kane Fracture  
1368 Zone (Coogan et al., 2000). Core-rim differences within individual samples shown by  
1369 filled (core) and open (rim) symbols in (c); sample colours and shapes refer to those  
1370 listed in the key on (c).

1371 Figure 9: Variation in plagioclase (a) and olivine (b) compositions with whole rock SiO<sub>2</sub> (a)  
1372 or Mg# (b), and the calculated equilibrium compositions of the respective crystal  
1373 phases based on whole rock XRF data (Table 1, and Electronic Appendix 2). In (a)  
1374 equilibrium compositions of feldspar are calculated at 1150 °C and 330 MPa (black)  
1375 and 1050 °C and 330 MPa (grey) respectively, based on feldspar-melt and Fe-Ti oxide  
1376 thermometry (see Table 8), and the pressure at the base of the crust at Ascension Island  
1377 (Klingelhöfer, 2001).

1378 Figure 10: Compositions of olivines from (a) mafic to intermediate lavas; (b) all pyroclastic  
1379 samples. Variations between core and rim compositions shown in panel (c), where open  
1380 symbols refer to rim analyses, and filled symbols core analyses. Sample areas relate to  
1381 those used in Table 1. For full data set see Electronic Appendix 2. The purple field on  
1382 (b) shows the compositions of olivines from mafic to intermediate lava samples.

1383 Figure 11: Compositions of pyroxenes from all Ascension Island samples. Triangles refer to  
1384 samples of mafic to intermediate lava samples; squares and circles are felsic lava  
1385 samples. Sample areas relate to those used in Table 1. For full data set see Electronic  
1386 Appendix 2.

1387 Figure 12: Matrix glass compositions from all Ascension Island samples. Sample areas relate  
1388 to those used in Table 1. For full data set see Electronic Appendix 2.

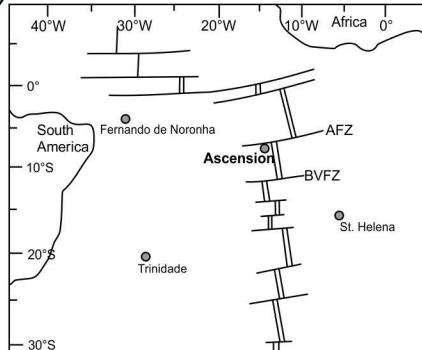
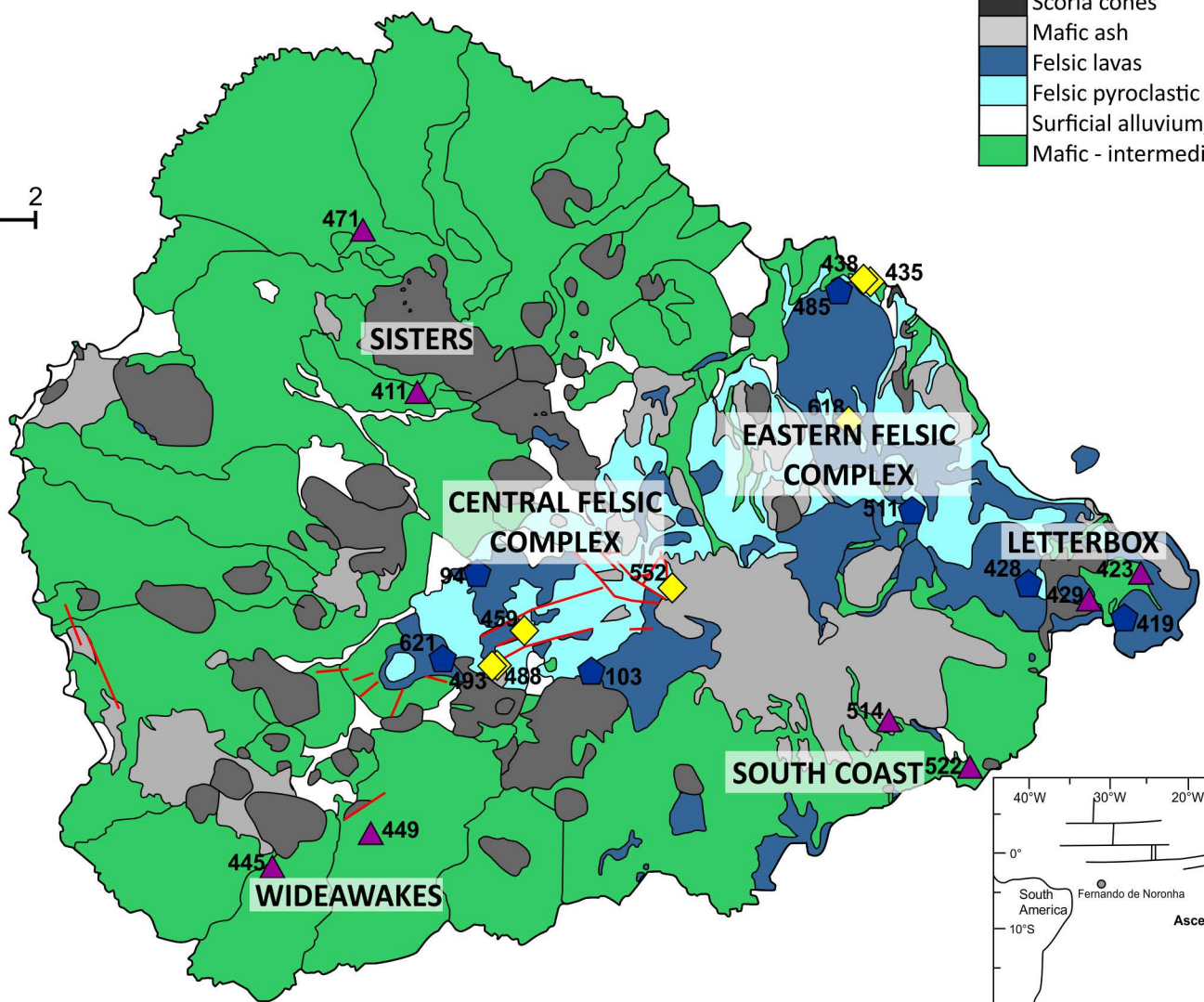
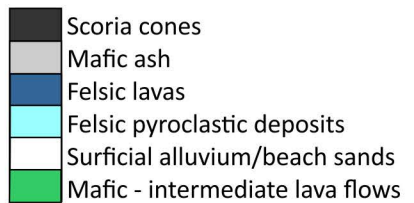
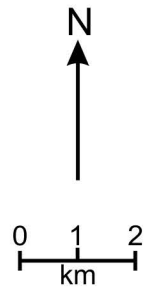
1389 Figure 13: Combined liquid lines of descent modelled from AI14-438 (to 950 °C, end marked  
1390 by dashed line) and AI-621 (< 950 °C, start from dashed line) using rhyolite-MELTS

1391 (Gualda & Ghiorso, 2015) at 330 MPa, 250 MPa, 170 MPa and 90 MPa. Purple ticks  
1392 mark 20% increments of melt fraction remaining; grey ticks mark 10% melt fraction  
1393 remaining, black ticks show liquid composition at 5% melt fraction remaining. Initial  
1394 water concentrations for AI-438 were 0.5 wt.% H<sub>2</sub>O, from MAR glasses showing a  
1395 component of the fertile mantle end member suggested to be the source of magmatism  
1396 at Ascension Island (Almeev et al., 2008; Paulick et al., 2010). \*XRF data used is a  
1397 combination of data presented here and Weaver et al. (1996) and Kar (1997).

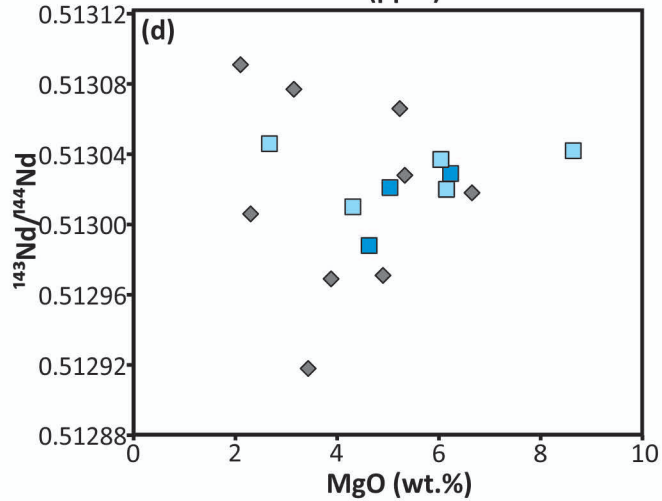
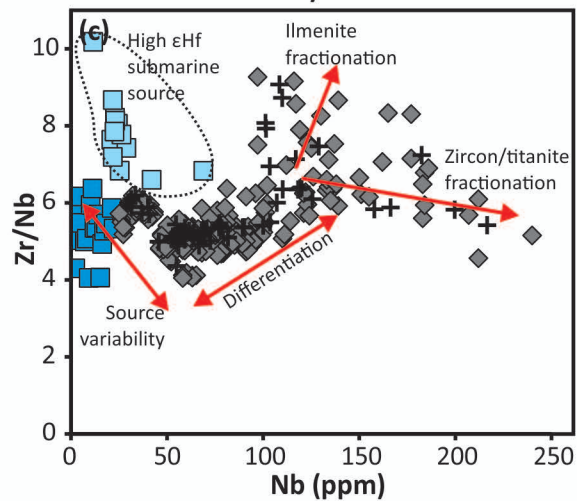
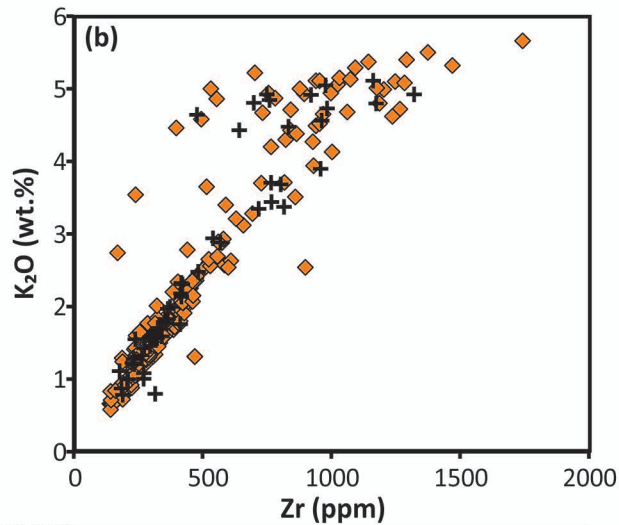
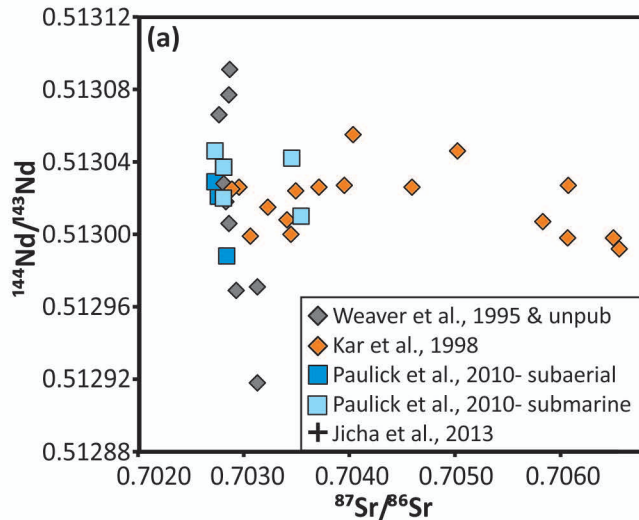
1398 Figure 14: Modelled fractionating crystal compositions from MELTS (purple symbols  
1399 representing different pressures of crystallisation, Gualda & Ghiorso, 2015) compared  
1400 with those measured in this study (grey symbols). The decompression trend (black  
1401 crosses) represents isothermal decompression at 800 °C from 90 to 1 MPa.

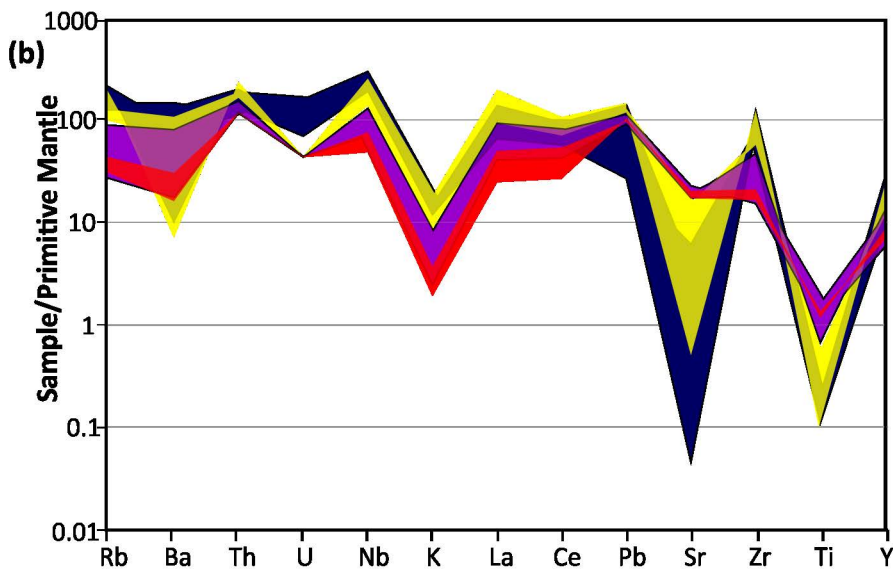
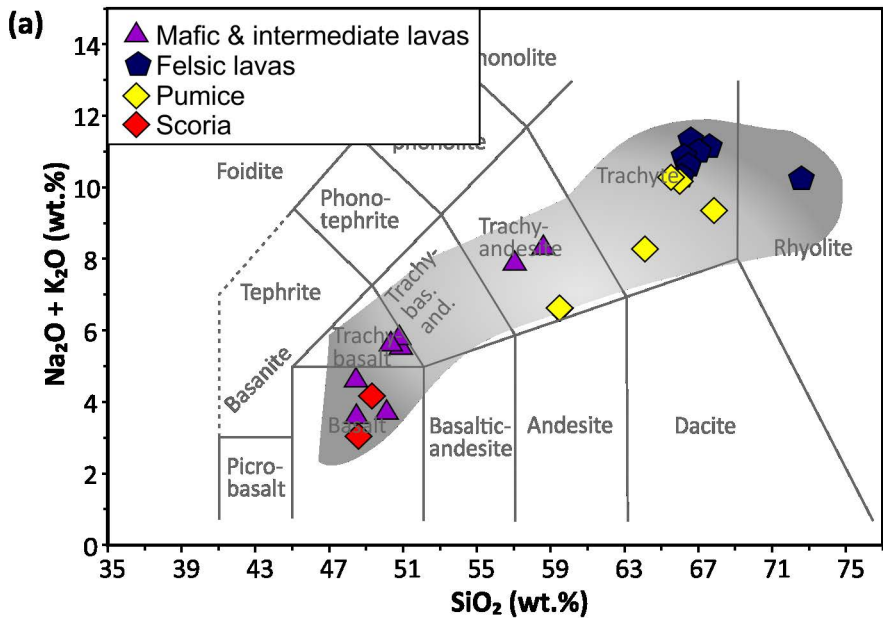
1402 Figure 15: Trace element fractional crystallisation modelling following the stages modelling  
1403 using Petrograph (Petrelli et al., 2005). XRF data used for the modelling shown in  
1404 diamonds, the resultant modelled liquid as diamonds. Each stage (1 – 4, connected by  
1405 dashed lines) represents 0.92, 0.40, 0.16, 0.07 total melt fraction remaining. \*Total  
1406 XRF data field in grey is a combination of data presented here, and Weaver et al., 1996  
1407 and Kar, 1997. <sup>1, 2</sup>: For stages and full modelled compositions and references for  
1408 partition coefficients used, see Electronic Appendix 3. The dashed field in (a)  
1409 represents a subset of samples which plot towards syenitic samples of Ascension (not  
1410 sampled in this study) and thus not modelled here.

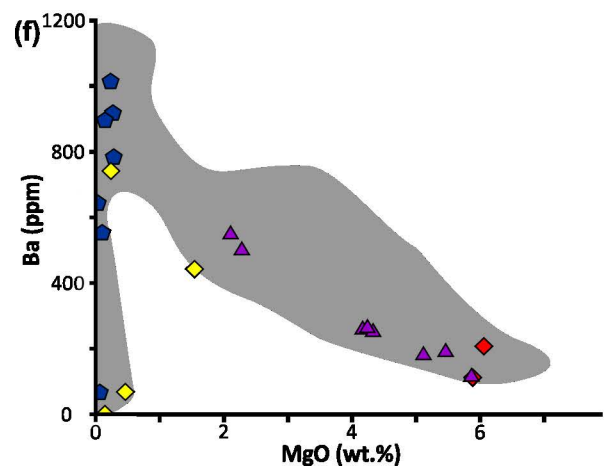
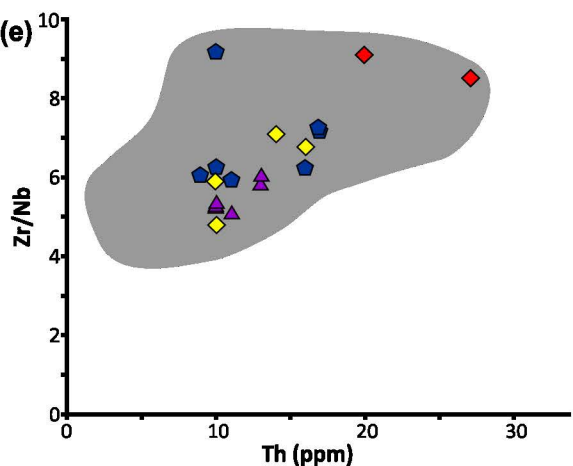
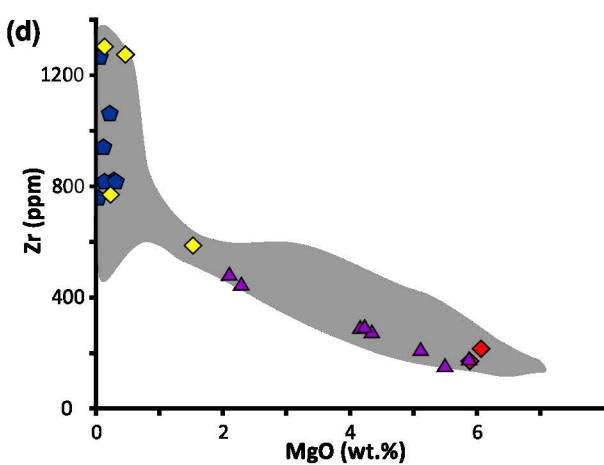
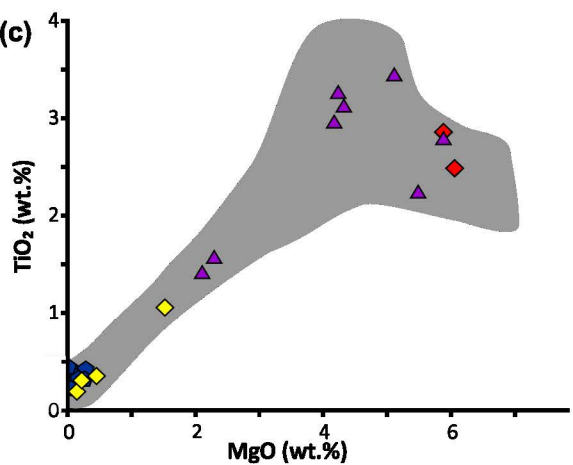
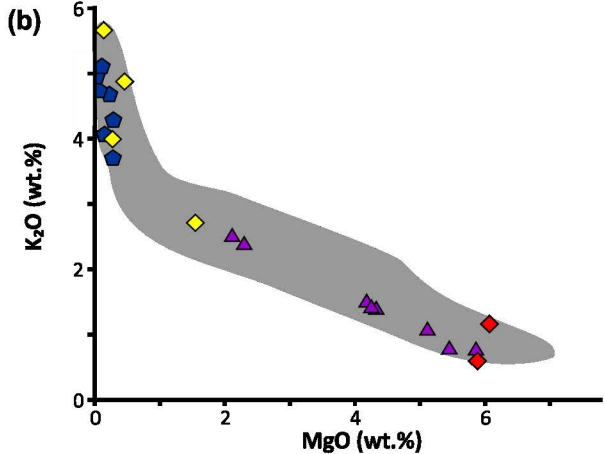
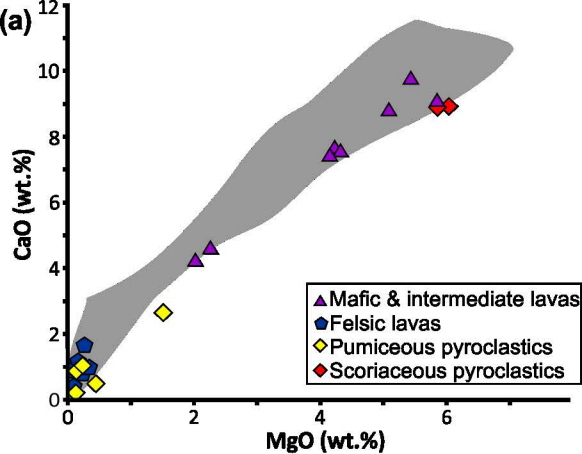
1411 Figure 16: Magmatic plumbing system model for Ascension Island- magma storage regions  
1412 are not to scale. Note the geographic variation in the distribution of plutonic bodies  
1413 (mafic plutonics in blue, zone of nested felsic plutonics hashed, with examples in  
1414 purple) within the crust. Ascent paths shown in red (mafic) and purple-blue (felsics).  
1415 Crustal structure derived from Klingelhöfer et al. (2001).



▲ Mafic & intermediate lava sample; 
 ◆ Pyroclastic sample; 
 ▣ Felsic lava sample; 
 / Fault



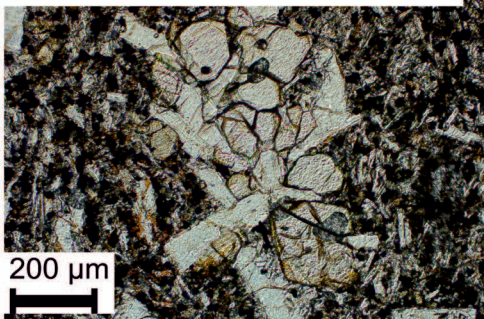






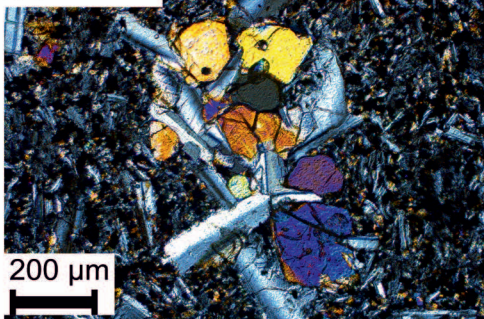
# PPL

(a) Al14-522: South Coast [48.9 wt.%]

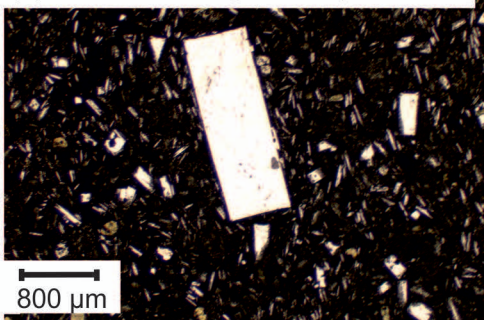


# XPL

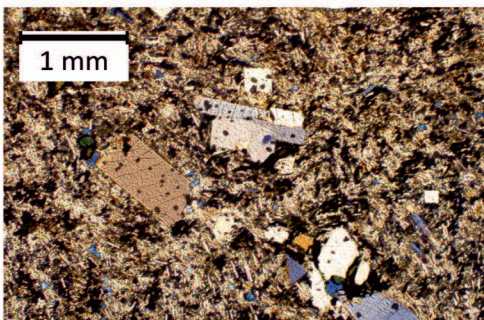
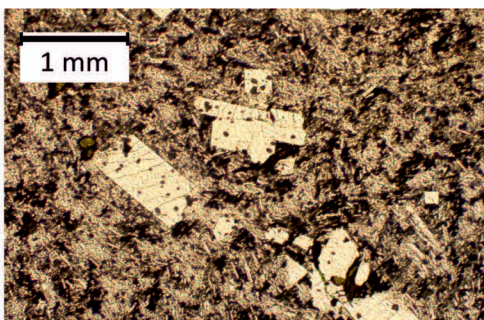
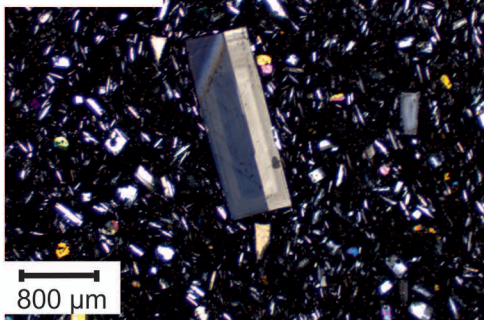
(b) Al14-522



(c) Al14-411: Young Sisters [49.5 wt.%]

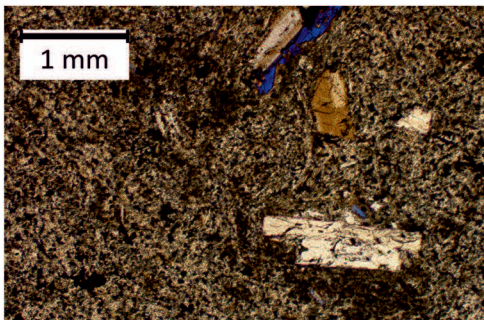
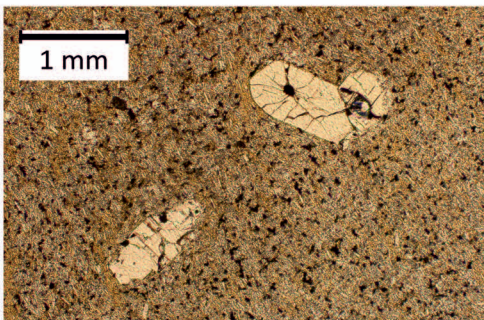


(d) Al14-411



(e) Al14-511: Cricket Valley [67.3 wt.%]

(f) Al14-511

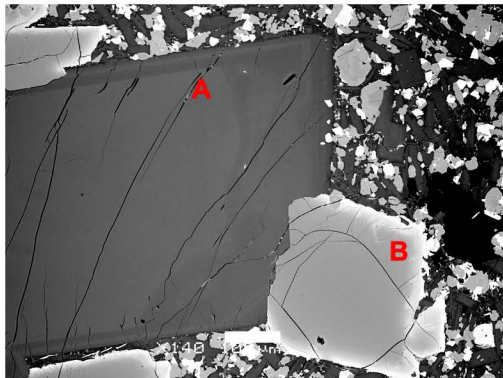


(g) Al14-428: White Horse [66.8 wt.%]

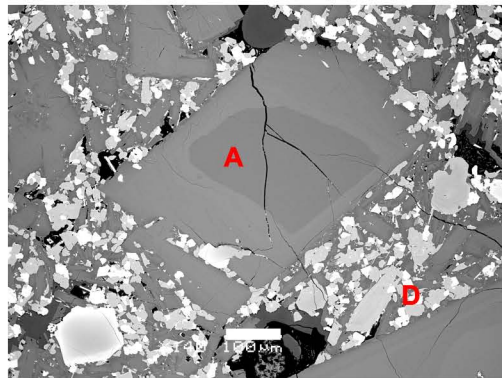
(h) Al14-485



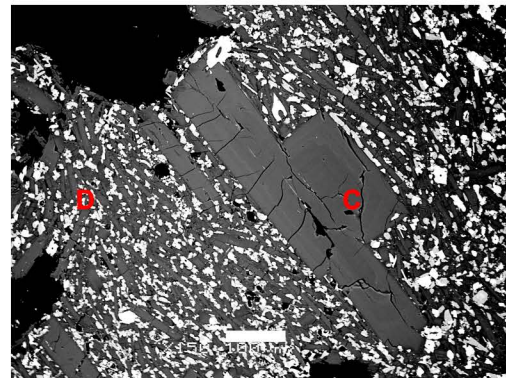
**(a)** Al-522: South Coast [48.9 wt.%]



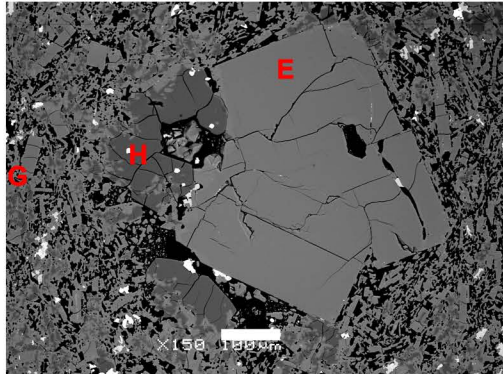
**(b)** Al-522: South Coast [48.9 wt.%]



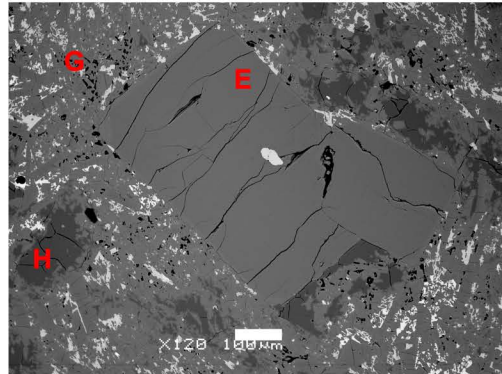
**(c)** Al-423: Letterbox [57.6 wt.%]



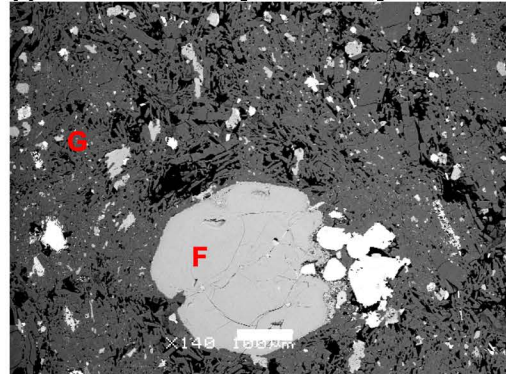
**(d)** Al14-428: White Horse [66.8 wt.%]



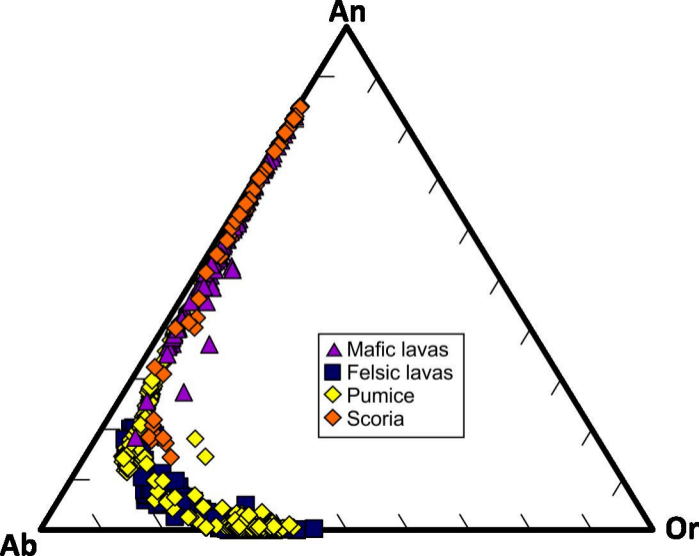
**(e)** Al-511: Cricket Valley [67.3 wt.%]

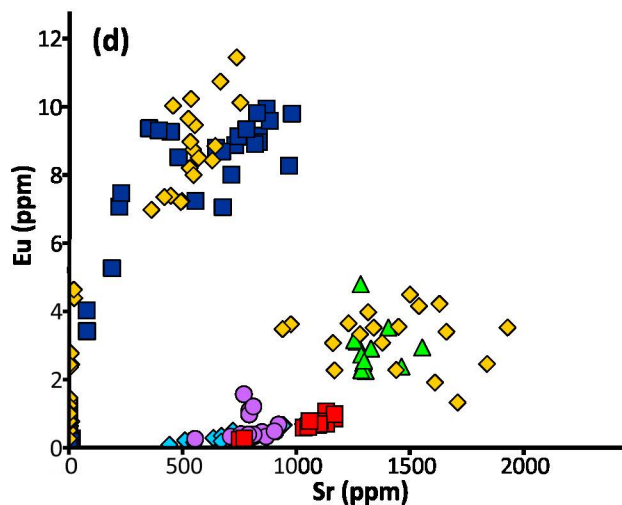
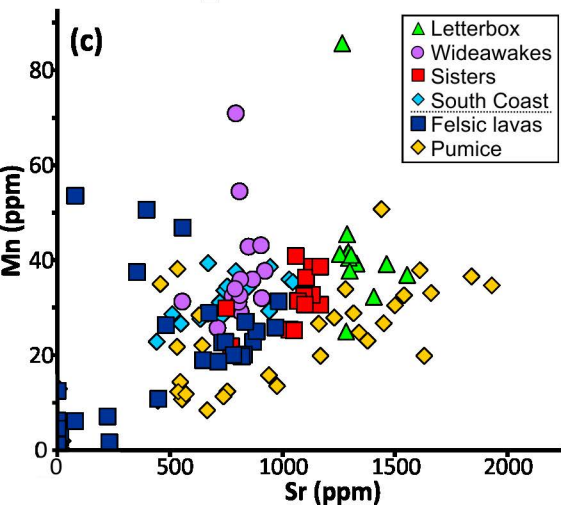
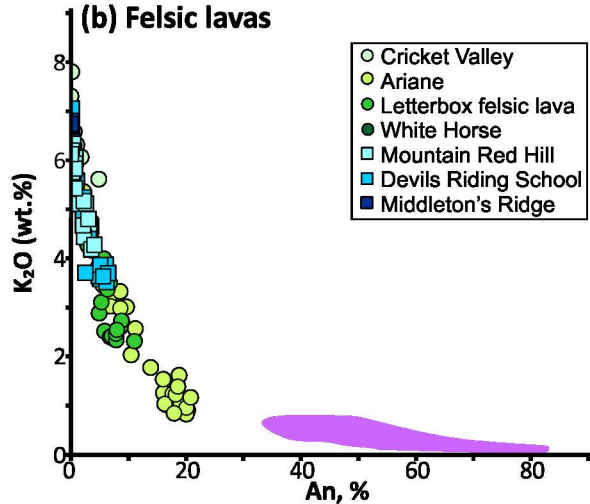
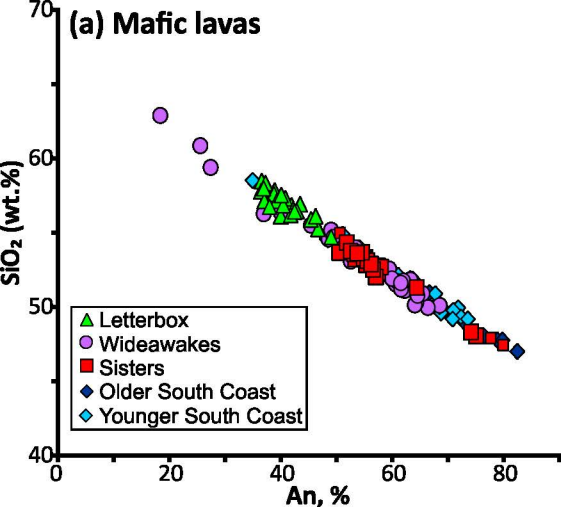


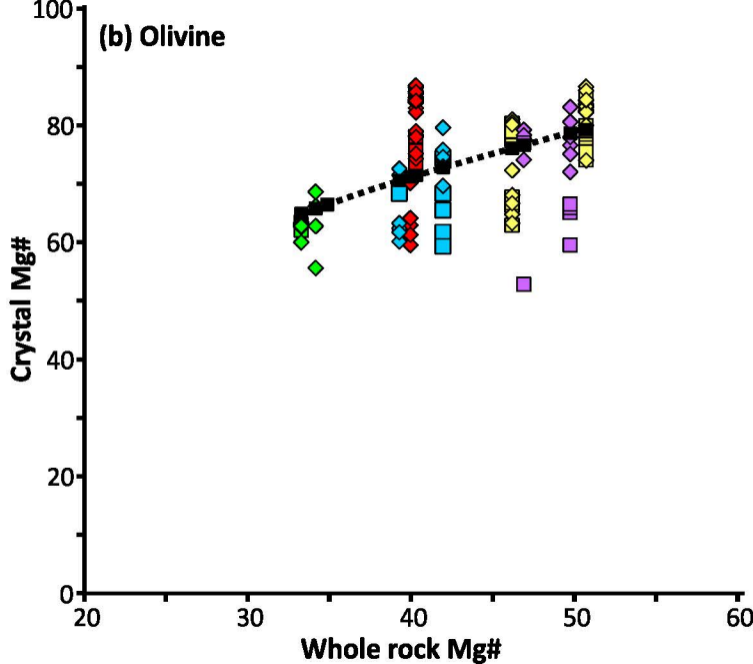
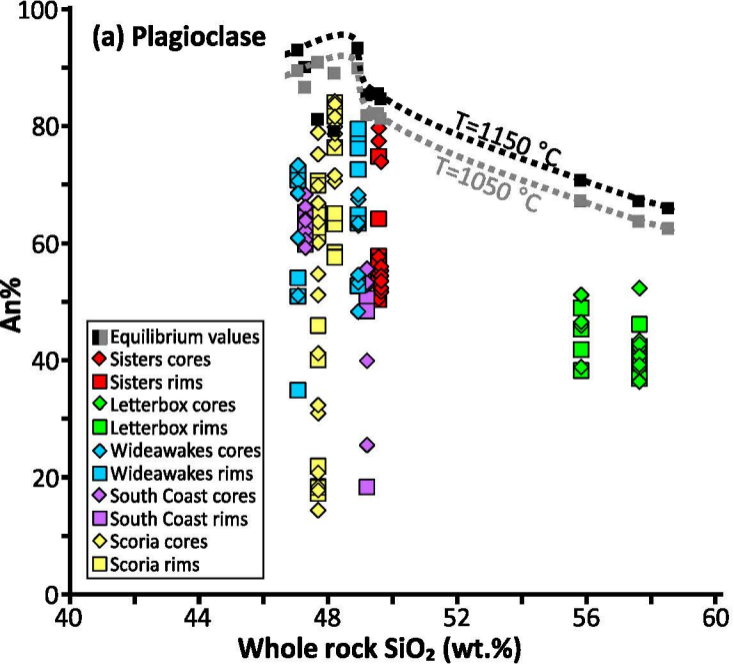
**(f)** Al-485: Ariane [65.9 wt.%]

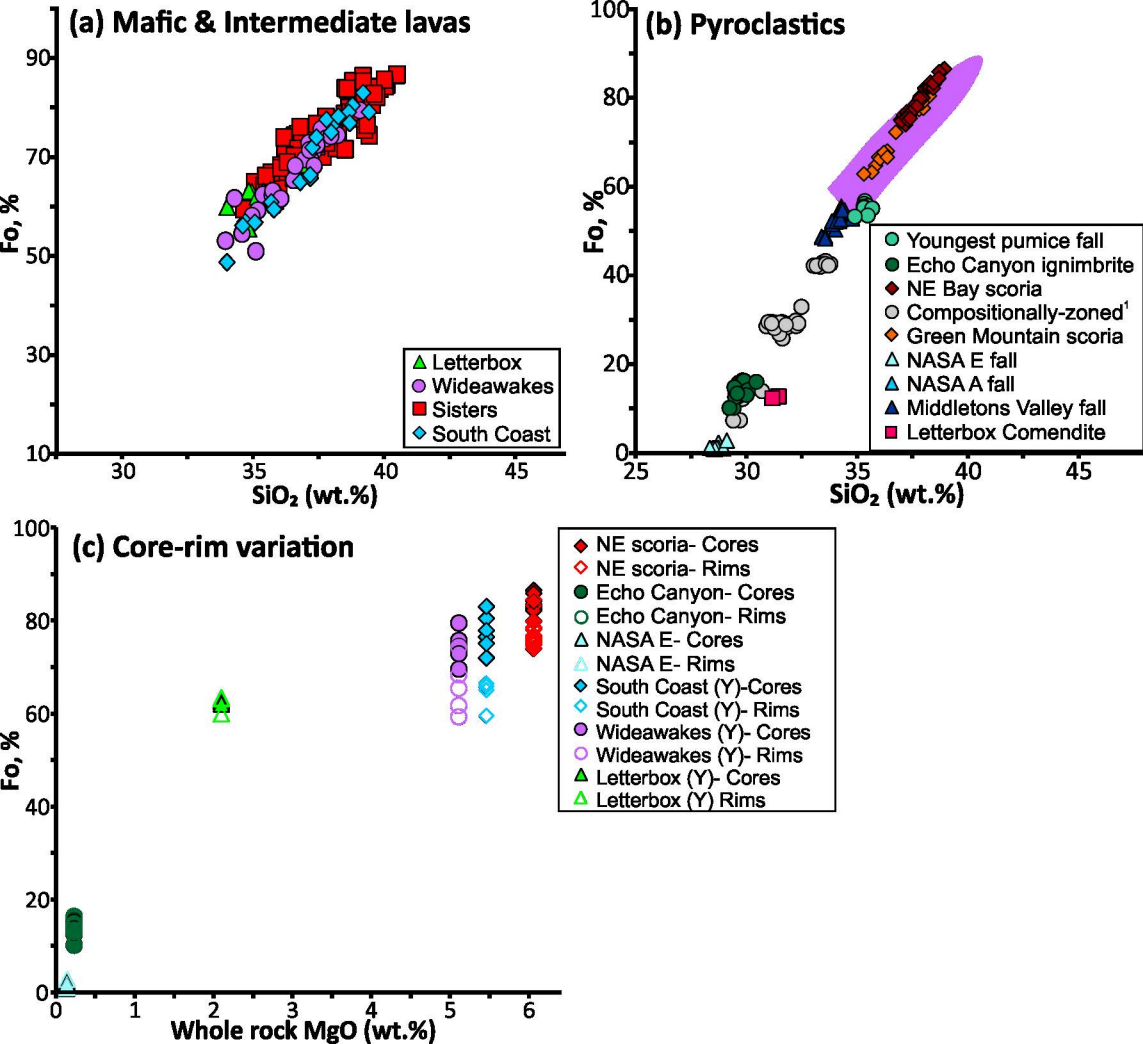


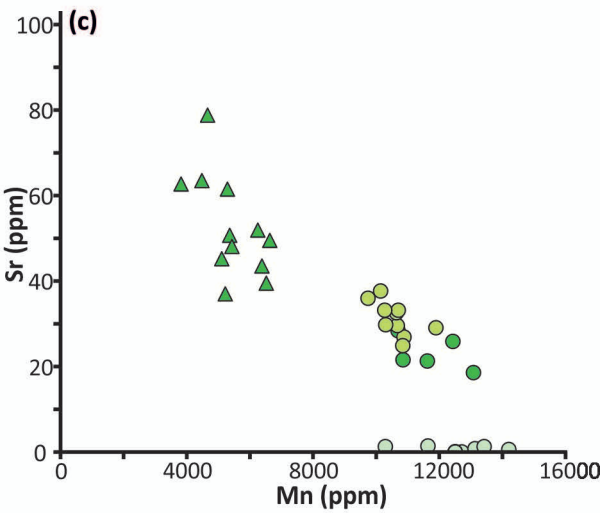
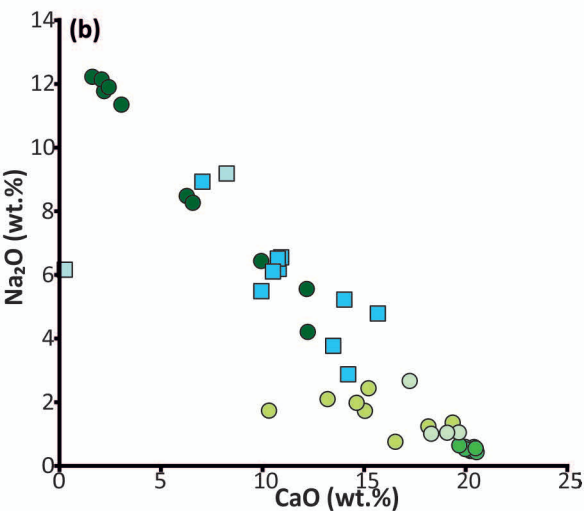
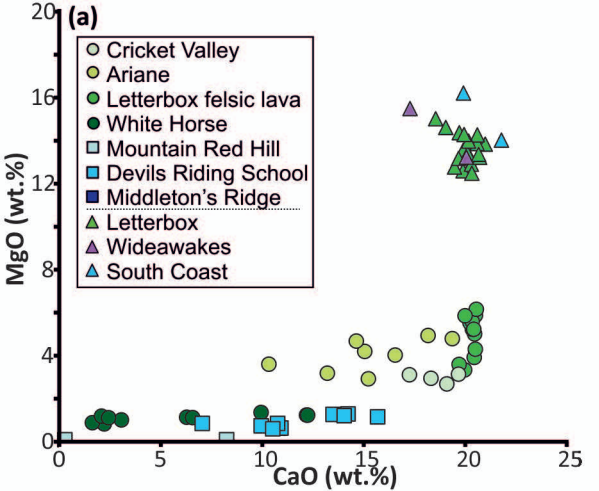


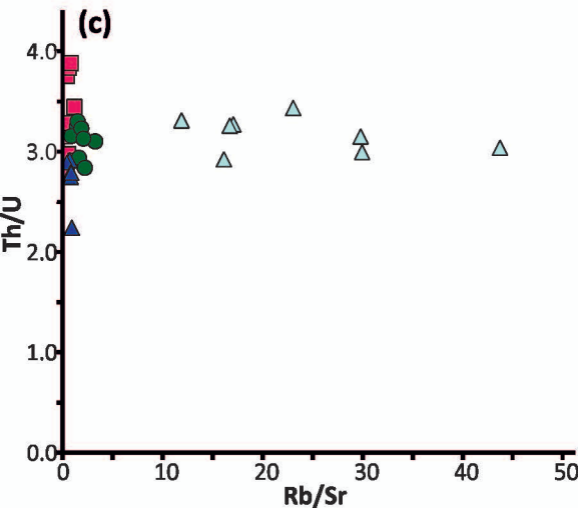
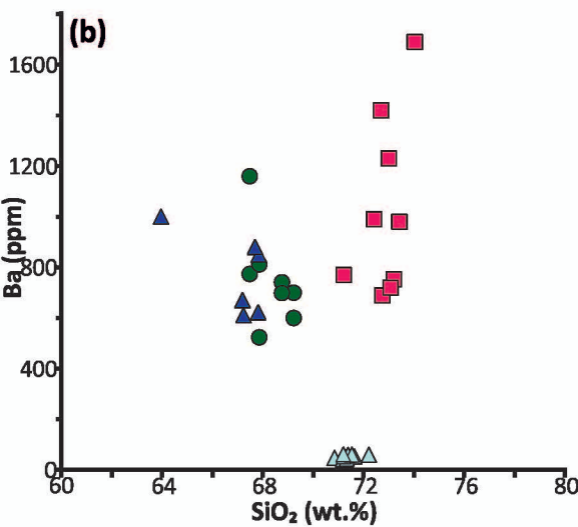
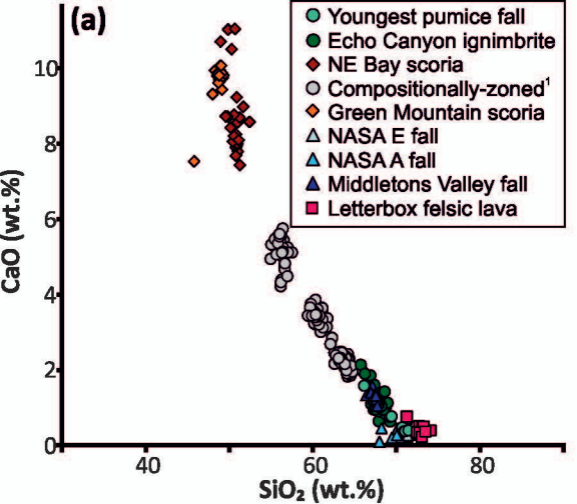


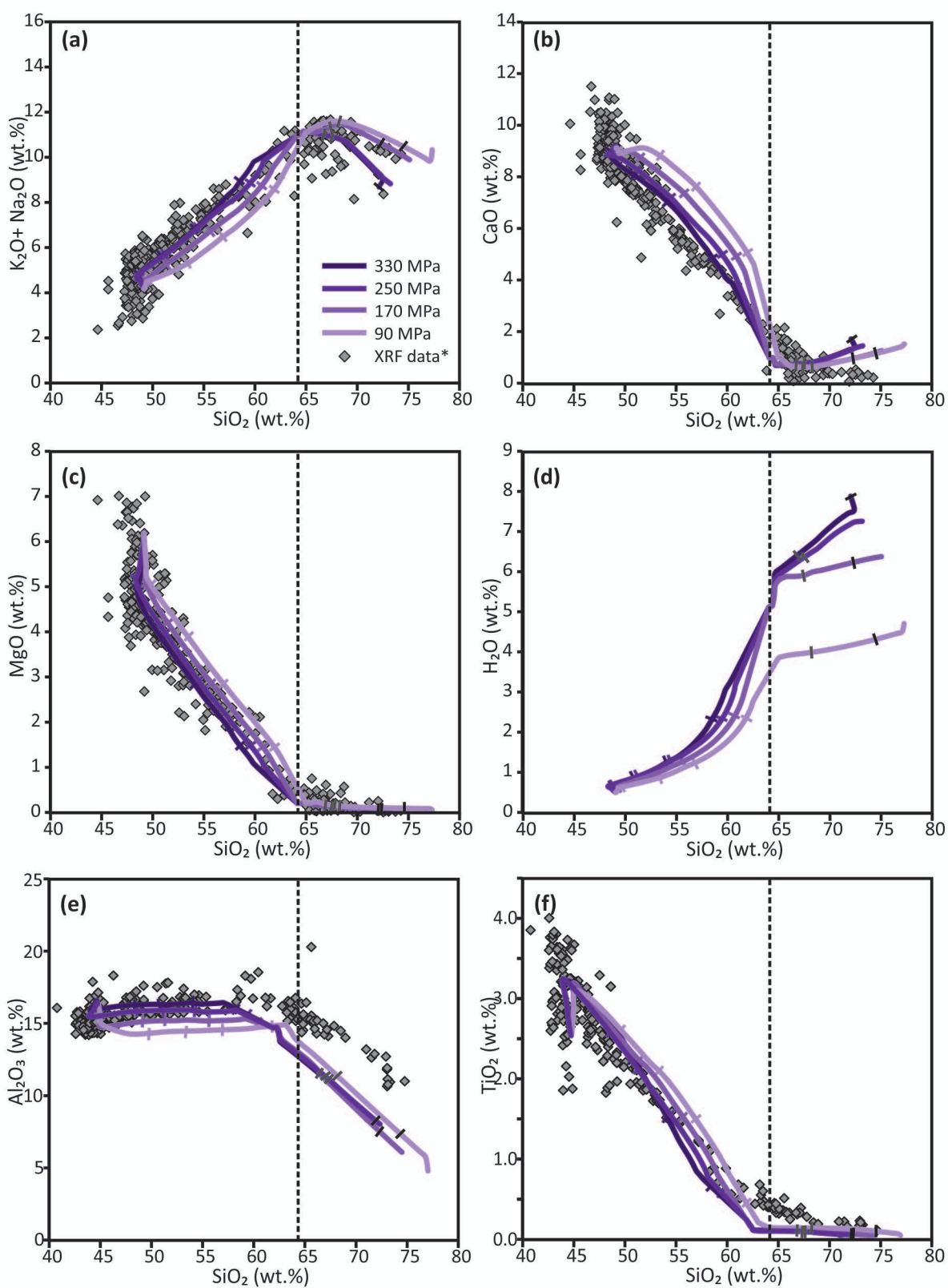


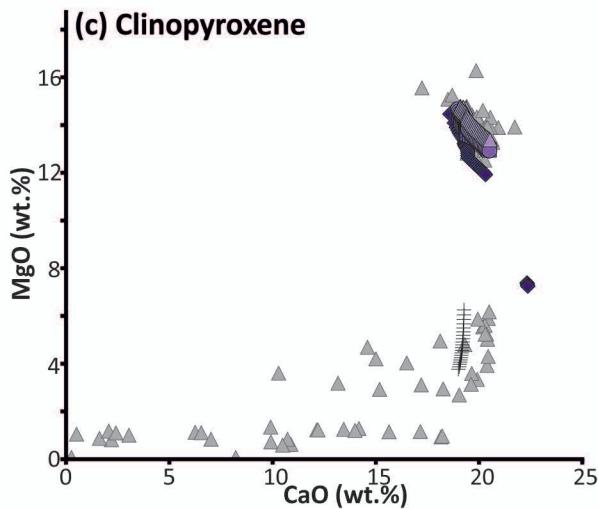
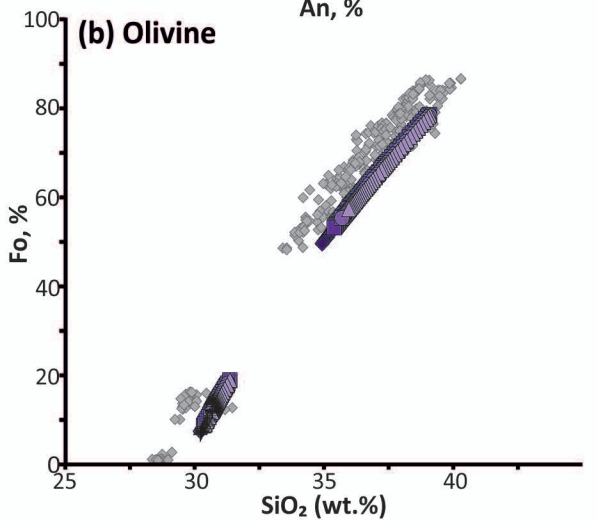
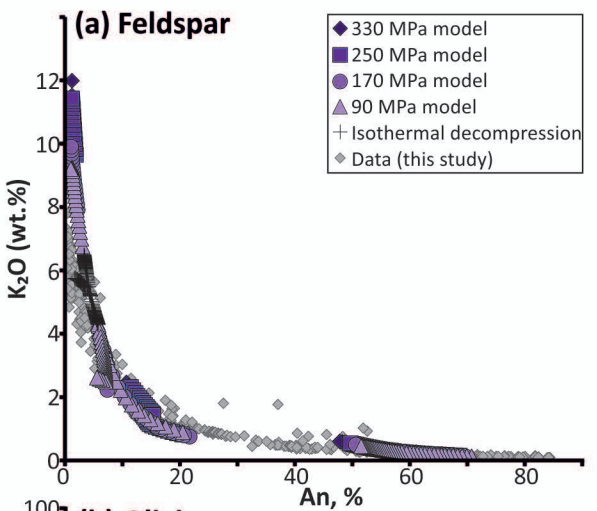




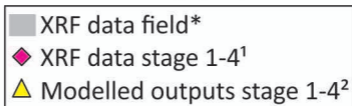
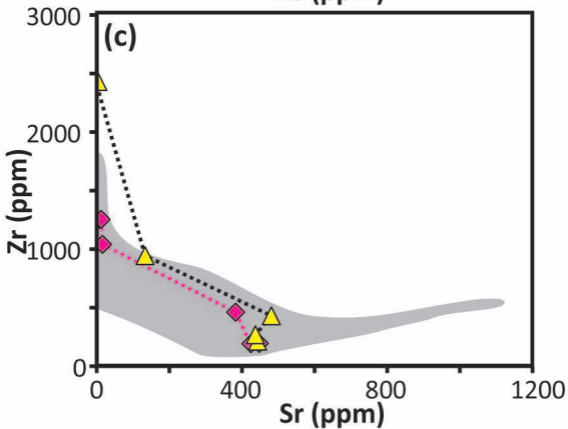
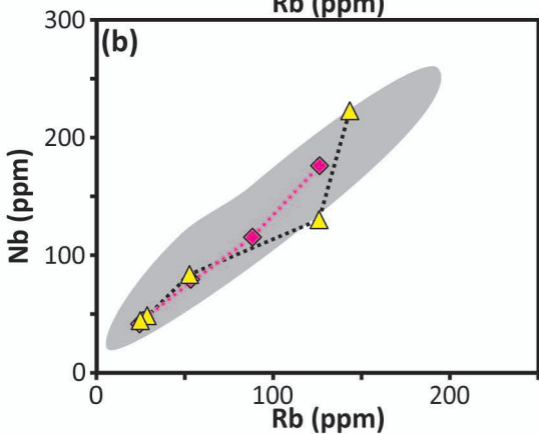
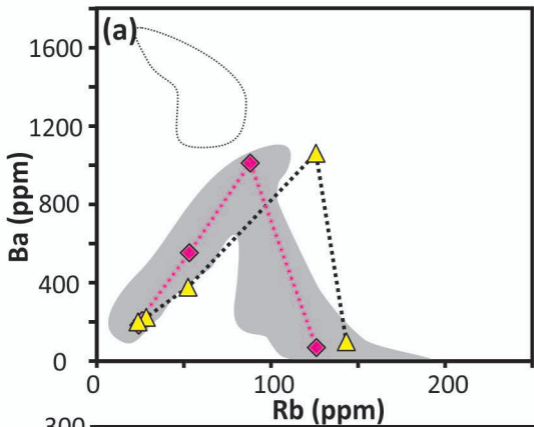


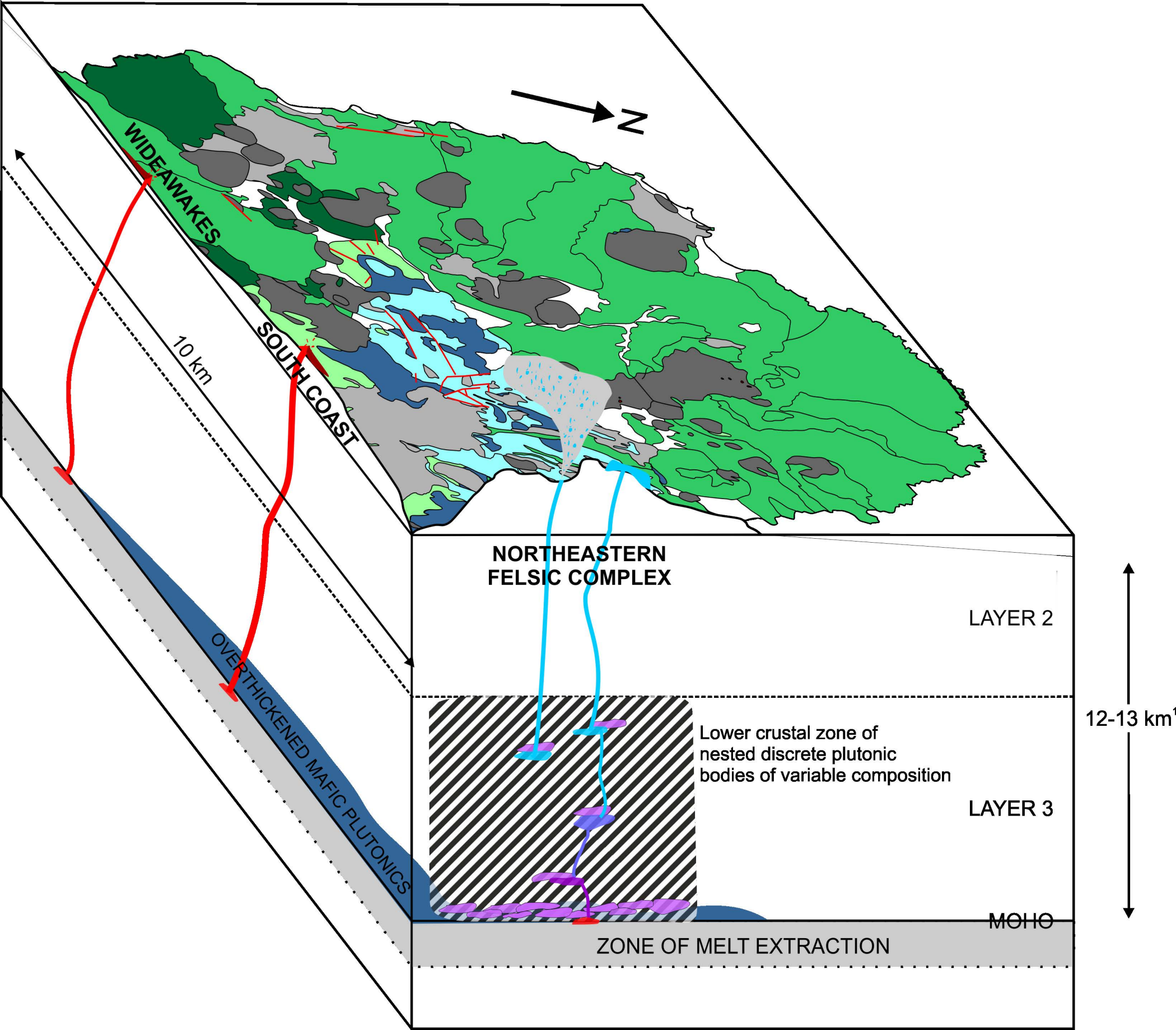












**Table 1:** Samples investigated for this study

Sample number	Region	TAS classification	Sample type	Grid Reference (E, N)	Approximate age of sample
AI14-411	Youngest Sisters	Trachy-basalt	mafic lava	0568706, 9123576	zero age *
AI14-471	Older Sisters	Trachy-basalt	mafic lava	0567966, 9125332	> AI14-411‡
AI14-423	Youngest Letterbox	Trachy-andesite	mafic lava	0577052, 9121328	--
AI14-429	Older Letterbox	Trachy-andesite	mafic lava	0576443, 9121020	> AI14-423‡
AI14-445	Youngest Wideawakes	Basalt	mafic lava	0566907, 9117906	298 ka *
AI14-449	Older Wideawakes	Trachy-basalt	mafic lava	0568052, 9118271	> 298 ka ‡
AI14-522	Youngest South Coast	Basalt	mafic lava	0575056, 9119068	120 ka ""
AI14-514	Older South Coast	Basalt	mafic lava	0574092, 9119604	> 120 ka ‡
AI14-485	Ariane flow	Trachyte	felsic lava	0573518, 9124726	169 ka *
AI14-511	Cricket Valley	Trachyte	felsic dome	0574341, 0122098	52 ka *
AI14-428	White Horse	Trachyte	felsic dome	0575736, 9121232	zero age *
AI-94	Middleton's Ridge	Rhyolite	felsic lava	05693, 91214	1094 ka *
AI-103	Mountain Red Hill	Trachyte	felsic lava	05706, 91202	602 ka *
AI14-419	Letterbox felsic lava	Trachyte	felsic dome	0576780, 9120822	--
AI15-621	Devil's Riding School	Trachyte	felsic dome	0568897, 9120365	652 ka*
AI15-618	Youngest pumice fall	Trachyte	pumice fall	0573646, 9123128	--
AI14-435A	Echo Canyon	Trachyte	pumice flow	0573840, 9124754	--
AI14-438	NE scoria	Basalt	scoria fall	0573786, 9124790	--
AI14-493A	NASA unit E	Trachyte	pumice fall	0569420, 9120262	> AI14-488A ‡
AI14-488A	NASA unit A	Trachyte	pumice fall	0569475, 9120264	--
AI14-459A	Middleton's Valley fall	Trachy-andesite	pumice fall	0569849, 9120661	--
AI14-552	Green Mountain scoria	Basalt	scoria fall	0571584, 9121198	--
AI14-439A <sup>+</sup>	<i>Compositionally-zoned top</i>	<i>Trachy-andesite</i>	<i>zoned fall</i>	<i>0573786, 9124790</i>	--
AI14-439D <sup>+</sup>	<i>Compositionally-zoned mid</i>	<i>Trachy-andesite</i>	<i>zoned fall</i>	<i>0573786, 9124790</i>	--
AI14-439G <sup>+</sup>	<i>Compositionally-zoned base</i>	<i>Trachyte</i>	<i>zoned fall</i>	<i>0573786, 9124790</i>	--

<sup>+</sup> Data presented in Chamberlain et al., 2016.

\* Ar-Ar age data from Jicha et al., 2014

"" K-Ar age data from Harris et al., 1992

‡ Stratigraphic relationship observed in the field

**Table 2:** Whole rock XRF data for Ascension samples, major elements in wt.%, trace elements in ppm.

Sample number	AI114-411	AI14-471	AI14-423	AI14-429	AI14-445	AI14-449	AI14-522	AI14-514	AI14-485	AI14-511	AI14-428	AI-94	AI-103
Location	Young Sisters	Old Sisters	Young Letterbox	Old Letterbox	Younger Wideawakes	Older Wideawakes	Younger South Coast	Older South Coast	Ariane	Cricket Valley	White Horse	Middleton's Ridge	Mountain Red Hill
SiO <sub>2</sub>	49.5	49.6	57.6	55.8	47.3	49.2	48.9	47.0	65.9	67.3	66.8	72.5	66.6
TiO <sub>2</sub>	3.12	2.97	1.42	1.58	3.44	3.26	2.25	2.79	0.42	0.37	0.41	0.23	0.45
Al <sub>2</sub> O <sub>3</sub>	15.5	15.5	16.9	16.7	15.3	15.2	16.1	15.5	16.1	15.3	15.5	12.7	15.5
Fe <sub>2</sub> O <sub>3</sub>	12.5	12.2	8.23	8.63	13.8	12.8	10.8	13.0	4.97	4.62	4.68	3.70	5.85
MnO	0.22	0.22	0.24	0.23	0.20	0.23	0.16	0.18	0.22	0.16	0.18	0.12	0.12
MgO	4.33	4.16	2.10	2.29	5.11	4.24	5.46	5.87	0.28	0.11	0.28	0.06	0.04
CaO	7.55	7.42	4.23	4.62	8.80	7.70	9.8	9.13	1.6	0.88	1.04	0.33	0.38
Na <sub>2</sub> O	4.03	4.18	5.68	5.40	3.45	4.09	2.85	2.76	6.62	6.07	6.73	5.60	6.04
K <sub>2</sub> O	1.41	1.50	2.53	2.37	1.08	1.44	0.80	0.77	3.71	5.11	4.30	4.72	4.94
P <sub>2</sub> O <sub>5</sub>	0.89	1.03	0.60	1.04	0.81	1.18	0.53	0.4	0.09	0.06	0.09	0.03	0.06
LOI	-0.10	-0.02	-0.45	-0.51	-0.21	-0.34	0.24	-0.14	0.12	0.00	0.15	0.37	0.95
Total	99.0	98.8	99.0	98.1	99.1	98.9	97.8	97.3	100.1	100.0	100.2	100.4	101.0
Ba	254	263	553	503	183	266	194	120	780	554	919	69	644
Rb	32	34	53	50	24	29	17	16	76	117	94	126	81
Sr	455	461	381	410	423	458	501	391	135	7	24	1	4
Pb	18	20	20	21	17	19	bdl	bdl	5	8	6	10	5
Th	10	10	13	13	11	10	bdl	bdl	9	16	11	17	10
U	bdl	bdl	1	1	1	1	bdl	bdl	bdl	3.7	1.6	bdl	2.0
Zr	279	291	483	450	214	292	158	182	818	940	823	1266	756
Nb	52	55	80	77	42	54	28	29	135	151	139	177	121
Y	41	44	54	52	27	49	24	30	85	62	45	113	57
La	34	35	64	56	28	41	bdl	bdl	71	53	50	78	45
Ce	96	105	144	140	74	115	61	49	156	119	112	177	100
Sc	21	19	12	12	24	18	26	28	9	9	8	3	12
V	227	203	42	55	317	208	252	314	3	4	7	4	3
Ni	8	5	3	5	24	3	56	65	5	3	4	6	5
Zn	122	128	128	132	115	130	83	105	150	126	136	197	151
<b>Sample number</b>	<b>AI114-419</b>	<b>AI15-621</b>	<b>AI15-618</b>	<b>AI14-435A</b>	<b>AI14-438</b>	<b>AI14-493A</b>	<b>AI14-488A</b>	<b>AI14-459A</b>	<b>AI14-552</b>				
<b>Location</b>	Letterbox Felsic lava	Devils Riding School	Youngest Pumice fall	Echo Canyon	NE scoria	NASA unit E	NASA unit A	Middleton's Valley fall	Green Mountain scoria				
SiO <sub>2</sub>	65.5	66.9	65.0	65.6	48.2	68.1	64.3	58.5	47.7				
TiO <sub>2</sub>	0.32	0.33	0.33	0.31	2.49	0.18	0.33	1.06	2.85				
Al <sub>2</sub> O <sub>3</sub>	15.9	15.4	13.7	15.7	16.1	13.1	14.5	17.6	16.0				
Fe <sub>2</sub> O <sub>3</sub>	4.58	4.41	4.05	4.34	11.5	3.74	5.18	5.59	13.4				
MnO	0.17	0.17	0.16	0.20	0.19	0.11	0.19	0.16	0.18				

MgO	0.15	0.22	0.16	0.23	6.06	0.14	0.46	1.53	5.88
CaO	0.87	1.03	0.80	1.08	8.93	0.21	0.49	2.65	8.90
Na <sub>2</sub> O	6.48	6.76	5.23	6.35	2.95	3.78	3.47	3.84	2.41
K <sub>2</sub> O	4.06	4.68	4.84	4.00	1.15	5.66	4.88	2.72	0.60
P <sub>2</sub> O <sub>5</sub>	0.57	0.04	0.07	0.04	0.94	0.02	0.03	0.37	0.31
LOI	0.27	0.93	4.57	2.70	-0.56	5.73	6.90	4.68	0.15
Total	98.9	100.9	98.9	100.5	97.9	100.8	100.7	98.7	98.3
Ba	898	1011	49	740	207	bdl	68	446	111
Rb	75	88	121	77	26	139	105	61	19
Sr	83	15	50	79	446	bdl	11	338	381
Pb	26	5	bdl	24	18	27	24	22	19
Th	17	10	20	16	10	27	20	14	10
U	2	3.4	bdl	bdl	1	bdl	bdl	1	1
Zr	818	1062	948	770	216	1301	1276	589	171
Nb	114	116	147	114	45	152	140	83	29
Y	76	73	89	72	35	96	88	52	28
La	95	57	109	91	34	135	127	64	17
Ce	157	117	173	162	94	188	186	124	47
Sc	6	10	bdl	bdl	23	bdl	bdl	6	28
V	2	4	bdl	bdl	209	bdl	bdl	34	307
Ni	3	7	bdl	bdl	65	bdl	bdl	6	45
Zn	138	145	141	142	111	171	179	103	105

**Table 3:** Petrography of Ascension lavas

Sample number	Region	TAS classification	Percentage Phenocrysts*	Vesicularity (%)	Fspar	Ol	Cpx	Cbt	Aeg	Glass	Ox+Accessory
Al14-411	Youngest Sisters	Trachy-basalt	<1 (f)	5	67	5	20				8
Al14-471	Older Sisters	Trachy-basalt	<1 (f)	7	68	10	17				5
Al14-445	Youngest Wideawakes	Basalt	3 (f, ol)	3	68	14	12				6
Al14-449	Older Wideawakes	Trachy-basalt	<1 (f, ol)	6	55	9	32				4
Al14-423	Youngest Letterbox	Trachy-andesite	<1 (f)	5	80	6	10				4
Al14-429	Older Letterbox	Trachy-andesite	2 (f)	43	77	8	9				6
Al14-522	Youngest South Coast	Basalt	40 (f, ol)	3	62	31	<1				7
Al14-514	Older South Coast	Basalt	18 (f, ol)	23	56	23					21
Al14-428	White Horse	Trachyte	2 (f)	24	64			29	4		3
Al14-511	Cricket Valley	Trachyte	6 (f, aeg)	1	78			12	9		1
Al14-485	Ariane flow	Trachyte	4 (f)	15	77			19			4
Al14-419	Letterbox felsic lava	Trachyte	<1 (f)	25	60		1	1		37	1
Al-103	Mountain Red Hill	Trachyte	2 (f, ox)	4	70			24	5		1
Al15-621	Devil's Riding School	Trachyte	3 (f)	13	68			27	4		1
Al-94	Middleton's Ridge	Rhyolite	<1 (f)	18	57			35	6		2

Arranged by relative age (see Table 1 and references therein)

Crystal percentages represent groundmass (<500  $\mu\text{m}$ ) crystals as well as those represented as phenocryst phases; Fspar = feldspar (plagioclase and ternary); Ol = olivine; Cpx = clinopyroxene; Cbt = cristobalite; Aeg = aegirine; Ox+Acc = bright in Back Scattered Electron imagery oxides and accessory phases including zircon and apatite.

\*Phenocrysts used to represent phases > 500  $\mu\text{m}$  in thin sectioned area. Phases which constitute phenocryst phases indicated in brackets: f= feldspar, ol = olivine; cpx = clinopyroxene; aeg = aegirine; ox = oxides;

**Table 4:** Representative feldspar data for Ascension samples

Sample	AI114-411				AI14-423		AI14-445		AI14-522		AI14-485		
Location	Young Sisters				Young Letterbox		Young Wideawakes		Young South Coast		Ariane		
Rock type	Trachy-basalt				Trachy-andesite		Basalt		Basalt		Trachyte		
Spot location	F3-C	F3-R	F5-C	F5-R	F1-C	F1-R	F5-C	F5-R	F1-C	F1-R	K3-C	K3-R	K10-C
SiO <sub>2</sub>	53.50	53.29	47.46	52.69	56.56	56.19	52.57	51.22	48.05	51.89	63.33	61.71	62.46
TiO <sub>2</sub>	0.16	0.17	0.05	0.13	0.04	0.06	0.13	0.16	0.05	0.10	0.00	0.01	0.01
Al <sub>2</sub> O <sub>3</sub>	28.26	28.18	32.79	29.25	26.89	26.57	28.59	29.16	32.44	29.44	22.67	23.21	23.19
FeO*	0.91	0.92	0.45	0.77	0.37	0.49	0.77	0.87	0.46	0.66	0.20	0.24	0.25
MgO	0.10	0.09	0.13	0.11	0.04	0.05	0.12	0.13	0.15	0.15	0.00	0.01	0.01
CaO	11.45	11.61	16.66	12.19	8.93	8.88	12.20	12.80	15.92	13.46	3.48	4.44	4.28
Na <sub>2</sub> O	5.04	5.10	2.30	4.74	6.55	6.63	4.48	4.29	2.67	4.17	9.17	8.56	8.87
K <sub>2</sub> O	0.32	0.33	0.07	0.26	0.36	0.36	0.24	0.23	0.10	0.18	1.26	1.17	0.83
Total	99.76	99.70	99.91	100.15	99.75	99.23	99.10	98.87	99.86	100.05	100.11	99.34	99.90
An	54.6	54.7	79.7	57.8	42.1	41.7	59.3	61.4	76.3	63.4	16.1	20.8	20.0
Ab	43.5	43.5	19.9	40.7	55.9	56.3	39.3	37.2	23.1	35.6	76.9	72.7	75.3
Or	1.8	1.9	0.4	1.5	2.0	2.0	1.4	1.4	0.6	1.0	7.0	6.5	4.7
Sc	4.55	4.90	4.64	4.60	4.6	3.11	6.38	12.0	8.48	7.22	6.60	5.20	8.00
Ti	421	612	696	664	254	344	797	4970	508	526	136	137	147
V	0.53	1.60	1.73	1.58	0.42	0.51	3.49	83	3.12	2.41	0.16	bdl	0.48
Mn	32.8	32.6	36.5	38.8	37.7	36.9	37.9	323	37.8	28.3	27.0	25.0	27.0
Ga	21.2	20.0	24.9	23.0	24.8	26.0	24.7	22.9	23.1	23.1	28.2	27.1	26.8
Rb	0.31	0.45	0.28	0.14	0.86	0.56	0.93	2.90	0.24	0.32	1.56	1.64	1.29
Sr	1130	1096	1104	1168	1289	1317	922	811	793	718	834	883	836
Y	0.19	0.18	0.20	0.40	0.64	0.40	0.57	9.00	0.11	0.08	0.02	0.13	0.13
Zr	0.41	0.22	0.00	0.11	2.80	1.54	1.26	31.5	0.50	0.17	bdl	1.86	0.25
Ba	134	137	121	175	295	324	109	131	70.7	86.2	1248	1353	1087
La	3.16	2.76	2.46	2.71	5.88	6.40	2.13	9.23	1.08	1.10	8.29	7.78	7.35
Ce	5.21	3.73	3.61	4.33	8.62	6.47	4.00	20.0	1.93	1.55	9.14	9.42	8.43
Pr	0.54	0.41	0.24	0.42	0.90	0.81	0.37	2.33	0.17	0.18	0.74	0.57	0.60
Nd	2.24	0.99	1.00	1.55	2.10	2.36	1.72	11.4	0.59	0.70	1.71	1.80	1.51
Sm	0.23	0.09	0.34	0.41	0.57	0.19	0.31	2.37	0.04	bdl	0.13	0.00	0.17
Eu	1.07	0.68	0.78	0.98	3.04	2.50	0.68	1.21	0.38	0.39	8.97	9.59	9.15
Gd	0.17	0.27	0	0.25	0.35	0.20	0.25	2.44	bdl	0.06	bdl	0.21	0.17
Yb	bdl	bdl	bdl	bdl	0.13	bdl	0.01	0.61	0.02	bdl	0.00	0.00	0.00
Pb	0.40	0.22	0.26	0.22	0.84	0.65	0.23	0.63	0.22	0.10	1.04	1.01	0.84

Sample	AI114-485	AI114-511		AI114-419		AI114-435A		AI114-493A		AI114-488A		AI114-459A		
Location	Ariane	Cricket Valley		Letterbox felsic lava		Echo Canyon		NASA Unit E		NASA Unit A		Middletons Valley		
Rock type	Trachyte	Trachyte		Trachyte		Trachyte		Trachyte		Trachyte		Trachy-andesite		
Spot location	F10-R	F2-C	F2-R	F3-C	F3-R	F4-C	F4-R	F26-C	F26-R	K13-C	K13-R	K18-C	K18-R	K20-C
SiO <sub>2</sub>	65.60	65.20	65.89	65.20	64.76	65.21	65.91	67.98	68.53	68.51	67.88	62.04	61.39	58.09
TiO <sub>2</sub>	0.03	0.02	0.01	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.01	0.02	0.07	0.04
Al <sub>2</sub> O <sub>3</sub>	19.42	19.33	19.00	20.71	20.90	21.22	20.57	19.24	19.53	19.36	19.31	22.50	23.09	24.64
FeO*	0.53	0.32	0.31	0.17	0.20	0.22	0.18	0.29	0.24	0.28	0.32	0.34	0.49	0.35
MgO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.03	0.03	0.04
CaO	0.75	0.23	0.07	1.44	1.67	3.21	2.59	0.07	0.12	0.13	0.12	5.10	5.86	7.59
Na <sub>2</sub> O	8.56	7.79	7.74	9.19	9.13	8.96	9.28	7.50	7.73	7.53	8.03	8.28	7.96	7.29
K <sub>2</sub> O	4.37	6.30	6.54	2.40	2.46	1.12	1.40	6.64	6.36	6.08	5.92	0.94	0.74	0.48
Total	99.28	99.22	99.57	99.10	99.14	99.95	99.93	101.71	102.52	101.89	101.58	99.25	99.62	98.52
An	3.5	1.1	0.3	6.9	7.9	15.5	12.3	0.3	0.6	0.6	0.6	24.1	27.7	35.5
Ab	72.2	64.6	64.1	79.4	78.2	78.1	79.8	63.0	64.5	64.9	67.0	70.6	68.1	61.8
Or	24.3	34.4	35.6	13.7	13.9	6.4	7.9	36.7	34.9	34.5	32.4	5.3	4.2	2.7
Sc	6.90	6.77	7.44	0.33	6.97	3.90	5.20	6.30	3.20	5.20	6.90	5.20	4.20	3.60
Ti	156	104	76.5	6.89	124	105	104	62.0	84.0	69.8	67.7	229	264	352
V	0.31	0.45	0.17	0.02	bdl	0.11	bdl	bdl	1.70	0.09	0.01	bdl	1.40	1.70
Mn	20.1	1.20	0.60	2.55	7.00	22.1	11.8	1.10	bdl	2.10	bdl	19.9	28.0	34.8
Ga	27.8	25.6	24.5	0.95	28.7	28.0	25.9	32.2	34.7	30.7	35.3	29.5	23.5	33.9
Rb	2.40	39.0	47.3	0.33	10.2	2.06	1.91	51.2	45.7	32.8	33.0	1.56	2.16	1.18
Sr	828	1.31	0.81	13.0	222	644	571	0.79	2.28	2.35	1.46	1169	1230	1930
Y	0.31	0.17	0.03	0.19	0.79	0.04	0.00	0.00	0.00	0.00	0.00	0.10	0.27	0.36
Zr	2.70	3.22	0.45	2.17	bdl	0.00	0.00	bdl	2.90	0.30	0.47	bdl	5.50	1.80
Ba	1261	120	80.6	96.7	3770	1680	1552	66.0	129	194	97.4	533	753	448
La	8.60	0.67	0.42	0.48	6.96	7.35	6.03	0.65	0.66	0.43	0.56	11.6	12.4	11.7
Ce	9.90	0.68	0.23	0.73	8.47	8.37	7.14	0.13	0.30	0.37	0.17	15.2	14.2	15.9
Pr	0.74	0.02	bdl	0.07	0.72	0.48	0.52	0.00	0.00	0.00	0.00	1.01	1.14	0.85
Nd	2.40	0.27	0.04	0.28	2.20	1.47	1.24	0.00	0.00	0.00	0.00	3.10	3.60	3.10
Sm	0.26	0.03	bdl	0.05	0.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.27
Eu	9.82	0.98	0.99	0.28	7.07	8.85	8.50	0.73	1.00	1.25	1.00	2.27	3.65	3.53
Gd	0.18	0.01	bdl	0.04	0.35	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.04	0.00
Yb	0.00	bdl	bdl	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Pb	1.21	1.12	0.93	0.07	1.94	1.29	1.34	0.53	0.88	0.89	0.69	1.91	1.85	1.02

Major element data given as wt.%; Anorthite, albite and orthoclase (An, Ab, Or) contents as mol. %; trace elements in ppm. For full data set, see Electronic Appendix 2. Where elements are below the detection limits of the method, bdl is written. Spot location refers to the analysis being core [C] or rim [R].



**Table 5:** Representative olivine major and trace element data for selected Ascension samples

Sample	AI14-411		AI14-423		AI14-445		AI14-522		AI14-435A			AI14-493A	
Location	Young Sisters		Young Letterbox		Young Wideawakes		Young South Coast		Echo Canyon			NASA unit E	
Rock type	Trachy-basalt		Trachy-andesite		Basalt		Basalt		Trachyte			Trachyte	
Spot tag	16_C	16_R	2_C	2_R	6_C	6_R	6_C	6_R	3_C	3_R	10_C	10_R	
SiO <sub>2</sub>	39.59	38.43	35.49	35.07	38.96	37.31	37.93	37.17	29.42	29.23	28.74	29.11	
Al <sub>2</sub> O <sub>3</sub>	0.06	0.00	0.02	0.02	0.03	0.03	0.03	0.04	0.00	0.00	0.00	0.00	
FeO*	14.67	24.67	32.21	32.30	19.05	28.37	22.89	29.63	59.56	59.01	62.61	61.38	
MnO	0.22	0.76	1.10	1.13	0.25	0.48	0.33	0.56	4.96	4.94	5.53	5.48	
MgO	43.65	34.97	30.98	30.87	41.46	34.30	38.63	32.92	3.77	3.73	0.83	0.98	
CaO	0.22	0.26	0.18	0.18	0.25	0.26	0.34	0.32	0.31	0.32	0.31	0.52	
Cr <sub>2</sub> O <sub>3</sub>	0.03	bdl	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
NiO	0.19	0.00	0.01	0.00	0.15	0.04	0.07	0.05	0.02	0.00	0.00	0.00	
P <sub>2</sub> O <sub>5</sub>	--	--	0.10	0.04	0.01	0.04	0.01	0.03	--	--	--	--	
<b>Total</b>	98.63	99.16	100.15	99.65	100.19	100.83	100.25	100.73	98.04	97.24	98.02	97.48	
Fo (%)	84.1	71.7	63.2	63.0	79.5	68.3	75.1	66.5	10.1	10.1	2.3	2.8	
Fa (%)	15.9	28.3	36.8	37.0	20.5	31.7	24.9	33.5	89.9	89.9	97.7	97.2	

Major element data given as wt.%; Fosterite, fayalite contents as mol. %. For full data set, see Electronic Appendix 2. Spot location refers to the analysis being core [C] or rim [R].

**Table 6:** Representative clinopyroxene major element data for selected Ascension samples

Sample	AI14-423		AI14-429		AI14-449		AI14-522		AI14-485		AI14-511		AI14-428		AI14-419		AI15-621		AI15-618	
Location	Young Letterbox		Old Letterbox		Old Wideawakes		Young South Coast		Ariane	Weatherpost		White Horse		Letterbox bedrock		Devils Riding School		Young pumice fall		
Rock type	Trachy-andesite		Trachy-andesite		Trachy-basalt		Basalt		Trachyte	Trachyte		Trachyte		Trachyte		Trachyte		Trachyte		
Spot tag	2_C	2_R	GM	GM	1_C	1_R	1_C	1_R	GM	GM	GM	GM	GM	GM	1_C	1_R	GM	GM	10_C	10_R
SiO <sub>2</sub>	48.35	50.51	49.37	36.12	50.22	48.46	50.63	47.34	47.79	47.74	47.90	50.81	50.86	48.50	48.20	47.94	49.82	51.82	51.58	
TiO <sub>2</sub>	1.27	0.75	0.97	0.02	0.96	2.25	1.09	1.94	2.05	0.29	0.42	1.95	1.88	0.39	0.53	0.32	0.47	0.63	0.56	
Al <sub>2</sub> O <sub>3</sub>	3.62	1.98	3.03	0.02	2.15	5.37	2.82	5.19	2.59	0.29	0.33	2.35	3.50	0.70	0.89	0.21	1.99	1.59	1.69	
FeO*	9.50	9.65	12.42	31.46	11.76	9.61	8.44	8.11	25.67	26.08	26.72	27.00	24.79	22.35	22.84	27.99	26.35	11.30	10.81	
MnO	0.47	0.61	0.72	1.09	0.67	0.25	0.19	0.15	2.38	1.54	1.64	1.25	1.25	1.52	1.47	1.37	0.91	0.72	0.67	
MgO	13.93	14.33	12.66	30.88	15.56	13.23	16.29	13.92	3.21	2.71	2.97	0.84	1.12	5.04	5.26	1.17	0.86	15.26	13.92	
CaO	20.54	20.55	19.84	0.14	17.24	20.05	19.86	21.74	13.17	19.05	18.26	2.19	2.41	20.41	20.30	15.64	10.73	18.70	20.34	
Na <sub>2</sub> O	0.49	0.50	0.55	0.01	0.46	0.56	0.29	0.39	2.09	1.04	1.01	11.76	11.88	0.51	0.53	4.78	6.50	0.43	0.50	
K <sub>2</sub> O	0.00	0.03	0.00	0.00	0.01	0.00	0.00	0.01	0.09	0.00	0.00	0.70	1.24	0.01	0.01	1.39	2.06	0.00	0.01	
Cr <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.00	0.00	0.00	0.13	0.23	0.00	0.00	0.00	bdl	bdl	0.00	bdl	bdl	0.00	0.00	0.00	
Total	98.16	98.91	99.58	99.74	99.03	99.78	99.76	99.02	99.05	98.74	99.25	98.85	98.94	99.44	100.02	100.81	99.69	100.45	100.09	

Major element data given as wt.%. For full data set, see Electronic Appendix 2. Spot location refers to the analysis being core [C], rim [R] or groundmass [GM].

**Table 7:** Representative glass analyses from Ascension Island

7a: Major element analyses normalised to 100 wt.%

Sample	AI14-419		AI15-618		AI14-552		AI14-438		AI14-435A			AI14-493A		AI14-488A		AI14-459A	
Location	Letterbox felsic lava		Young pumice		Green Mountain sc.		NE scoria		Echo Canyon			NASA unit E		NASA unit A		Middelton's Valley fall	
Spot Tag	419-1	419-2	15-5	15-6	1-1	1-4	14-1	14-2	10-2	10-3	10-4	8-1	8-4	15-1	15-2	4-1	4-3
SiO <sub>2</sub>	71.21	72.69	70.64	70.74	48.91	49.14	50.79	50.99	67.86	69.22	67.98	71.17	71.30	71.07	70.11	67.23	67.60
TiO <sub>2</sub>	0.27	0.31	0.19	0.19	3.68	3.72	3.61	3.52	0.10	0.29	0.22	0.21	0.22	0.17	0.18	0.48	0.49
Al <sub>2</sub> O <sub>3</sub>	12.04	10.48	13.26	12.91	13.51	13.87	13.54	12.90	17.98	13.88	16.01	12.33	11.78	13.67	13.60	16.80	16.64
FeO*	5.64	5.77	4.50	4.89	13.53	13.27	12.02	11.97	1.38	4.47	3.43	4.77	4.87	3.28	3.96	2.78	2.94
MnO	0.27	0.28	0.27	0.25	0.15	0.23	0.19	0.20	0.08	0.19	0.16	0.18	0.16	0.12	0.20	0.07	0.06
MgO	0.06	0.07	0.03	0.02	5.32	5.41	3.78	4.52	0.02	0.08	0.09	0.02	0.02	0.00	0.00	0.54	0.50
CaO	0.77	0.51	0.48	0.46	9.70	9.43	7.91	8.56	0.65	0.64	0.90	0.29	0.28	0.46	0.45	1.32	1.23
Na <sub>2</sub> O	5.15	4.44	6.14	6.11	3.34	3.16	3.48	3.22	7.32	5.43	6.09	5.52	4.42	4.42	4.95	5.88	5.73
K <sub>2</sub> O	4.56	5.41	4.46	4.40	1.20	1.10	2.19	2.21	4.36	5.03	4.47	4.75	5.94	6.14	5.74	4.41	4.43
BaO	0.00	0.00	0.00	0.00	0.05	0.07	0.08	0.06	0.11	0.06	0.09	0.04	0.00	0.00	0.00	0.09	0.04
P <sub>2</sub> O <sub>5</sub>	0.03	0.04	0.03	0.01	0.57	0.56	1.97	1.74	0.04	0.01	0.00	0.03	0.01	0.04	0.01	0.11	0.10
Cl	0.00	0.00	0.00	0.00	0.04	0.05	0.11	0.12	0.11	0.43	0.34	0.54	0.60	0.56	0.54	0.25	0.24
F	0.00	0.00	0.00	0.00	0.00	0.00	0.33	0.00	0.00	0.27	0.22	0.14	0.40	0.07	0.26	0.03	0.00
Total	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100

Major element data given as wt.%. For full data set, see Electronic Appendix 2.

**Table 7b:** Selected trace element data from glass in felsic samples (all elements in ppm); where elements are below the detection limits of the method, bdl is written. Analyses in italics previously published in Chamberlain et al., 2016.

Sample	AI14-419		AI14-435A		AI14-493A		AI14-459A		AI14-439D			AI14-439G		
Location	Letterbox felsic lava		Echo Canyon		NASA unit E		Middelton's Valley fall		<i>Compositionally-zoned mid (D) and upper (G)</i>					
Tag	G1	G2	G1	G2	G1	G2	G1	G2	G3	G1	G2	G1	G2	G3
Li	10.0	15.0	10.0	17.0	15.5	29.0	24.0	bdl	13.0	<i>13.6</i>	<i>18.0</i>	<i>29.0</i>	<i>20.0</i>	<i>18.0</i>
Sc	13.6	15.9	13.8	12.8	9.70	9.40	5.00	11.1	13.0	<i>11.2</i>	<i>21.0</i>	<i>27.0</i>	<i>10.1</i>	<i>27.0</i>
Ti	994	1020	1705	1300	1448	1437	5400	4960	5400	<i>2360</i>	<i>5690</i>	<i>4200</i>	<i>4530</i>	<i>2900</i>
V	1.60	1.20	0.71	bdl	4.37	6.80	10.1	10.0	21.0	<i>bdl</i>	<i>5.30</i>	<i>1.10</i>	<i>33.8</i>	<i>11.0</i>
Mn	863	820	1544	1290	1259	1270	1720	1060	1190	<i>1160</i>	<i>2720</i>	<i>2130</i>	<i>1290</i>	<i>1550</i>
Zn	84	90	154	215	268	239	178	113	94	<i>97</i>	<i>164</i>	<i>134</i>	<i>107</i>	<i>155</i>
Ga	34	32	30	32	39	42	57	38	33	<i>16</i>	<i>37</i>	<i>35</i>	<i>27</i>	<i>17</i>
Rb	101	58	90	90	169	182	166	134	161	<i>44</i>	<i>73</i>	<i>97</i>	<i>56</i>	<i>64</i>
Sr	90	106	50	44	10	8	246	175	220	<i>175</i>	<i>390</i>	<i>184</i>	<i>117</i>	<i>159</i>
Y	76	49	77	80	124	129	52	44	38	<i>34</i>	<i>74</i>	<i>70</i>	<i>41</i>	<i>79</i>
Zr	930	584	791	868	1507	1661	1180	1000	1310	<i>286</i>	<i>620</i>	<i>760</i>	<i>490</i>	<i>830</i>
Nb	133	89	147	161	234	247	160	135	141	<i>64</i>	<i>138</i>	<i>141</i>	<i>83</i>	<i>98</i>
Ba	746	1240	790	720	55	61	950	651	1020	<i>470</i>	<i>900</i>	<i>1290</i>	<i>840</i>	<i>1300</i>
La	76	51	83	83	138	145	93	65	71	<i>41</i>	<i>73</i>	<i>81</i>	<i>54</i>	<i>79</i>
Ce	160	113	169	178	274	283	197	124	145	<i>75</i>	<i>160</i>	<i>153</i>	<i>102</i>	<i>153</i>
Pr	18	12	19	20	30	30	17	12	9	<i>7</i>	<i>19</i>	<i>15</i>	<i>12</i>	<i>19</i>

<b>Nd</b>	67	50	73	77	112	115	57	43	45	33	85	60	43	75
<b>Sm</b>	16.9	9.3	15.3	14.8	24.2	23.9	13.6	8.2	7.8	8	12.9	14.8	8.5	10.2
<b>Eu</b>	2.67	3.92	3.24	2.52	2.61	2.40	2.57	2.40	2.60	1.72	4.60	4.40	3.06	3.60
<b>Gd</b>	11.6	8.8	13.8	14.9	21.9	22.4	10.6	6.1	10.0	7.0	12.0	11.6	10.2	8.9
<b>Dy</b>	13.3	11.0	14.4	14.1	23.6	24.2	9.4	7.3	10.0	7.7	11.2	11.8	7.8	14.6
<b>Er</b>	8.10	5.66	8.45	7.91	12.93	13.39	5.30	5.20	4.70	3.70	8.10	7.40	4.40	4.90
<b>Yb</b>	8.70	6.70	7.97	7.48	11.91	13.00	4.60	5.20	6.20	3.60	7.80	6.70	2.50	5.50
<b>Lu</b>	1.18	1.00	1.15	1.15	1.61	1.76	1.18	0.81	0.60	0.27	0.99	0.59	0.37	0.51
<b>Hf</b>	18.6	12.7	17.1	19.2	30.3	33.8	26.8	21.0	23.5	6.9	12.8	14.5	9.2	13.2
<b>Ta</b>	7.2	5.0	8.5	9.0	13.3	13.4	11.1	9.5	6.9	3.2	6.8	6.1	4.5	6.9
<b>W</b>	2.50	0.79	1.88	1.95	3.58	4.00	5.50	2.77	1.90	0.54	2.00	1.03	1.00	1.14
<b>Pb</b>	5.91	4.65	6.75	6.79	14.37	13.06	13.50	10.20	4.30	2.90	4.10	6.30	3.50	6.20
<b>Th</b>	10.9	7.8	10.8	10.7	21.1	21.8	20.5	16.0	16.0	4.2	8.7	7.9	5.7	9.1
<b>U</b>	3.15	2.02	3.33	3.40	6.42	6.32	7.00	5.80	5.70	1.13	2.71	2.20	1.82	2.77

**Table 8:** Intensive variables modelled for Ascension Island samples

Sample number	Region	FeTi-Oxide Thermometry <sup>1</sup>		Feldspar-melt Thermometry <sup>2,3</sup>	
		T (°C)	fO <sub>2</sub> (ΔNNO)	T (°C)	wt.% H <sub>2</sub> O <sup>4</sup>
AI14-411	Youngest Sisters	935	-0.4	1150	
AI14-471	Older Sisters	--	--	1152	
AI14-423	Youngest Letterbox	990	-0.05	1093	
AI14-429	Older Letterbox	1025	+0.08	1110	
AI14-445	Youngest Wideawakes	985	-0.34	1167	
AI14-511	Cricket Valley	884	-1.6	886	5.2
AI14-428	White Horse	--	--	837	5.5
AI-94	Middleton's Ridge	--	--	772	4.7
AI-103	Mountain Red Hill	967	-1.9	801	7.2
AI14-419	Letterbox felsic lava	--	--	837	
AI15-621	Devil's Riding School	--	--	871	5.1
AI15-618	Youngest pumice fall	--	--	847	6.4
AI14-435A	Echo Canyon	--	--	993	
AI14-438	NE scoria	--	--	1164	
AI14-493A	NASA unit E	--	--	811	7.1
AI14-488A	NASA unit A	--	--	813	8.1
AI14-459A	Middleton's Valley fall	950	-0.2	1034	
AI14-552	Green Mountain scoria	--	--	1174	
AI14-439A*	Compositionally-zoned scoria	845	-2.28		
AI14-439G*	Compositionally-zoned pumice	866	-1.94		

<sup>1</sup>Using the calibration of Ghiorso & Evans, 2008, on oxides which have passed the equilibrium test of Bacon & Hirschmann (1988).

<sup>2</sup>Plagioclase-melt thermometry from Putirka (2008), only analyses where  $K_D(\text{Ab-An})$  was  $0.1 \pm 0.11$  for  $T < 1050$  °C, or  $0.27 \pm 0.05$  at  $T > 1050$  °C were used (Putirka, 2008).

<sup>3</sup>Alkali-feldspar-melt thermometry (Putirka, 2008). Only analyses within the equilibrium bounds outlined in Mollo et al. (2015) were used.

<sup>4</sup>Alkali-feldspar-melt hygrometry (Mollo et al., 2015). Only analyses within the equilibrium bounds outlined in Mollo et al. (2015) were used. Uncertainties of  $\pm 0.7$  wt.% consistent with that published in Mollo et al. (2015) are assumed.