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#### Paper:

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3	Volcanoes (Ascension Island, South Atlantic)
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### 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 volcano which has produced a continuum of eruptive compositions from basalt to rhyolite in 47 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

et al., 2016) or through a combination of these processes (e.g. Bohrson & Reid, 1997;

Wiesmaier et al., 2013; Sliwinski et al., 2015). Understanding the relative importance of these
processes in any one setting has significant implications for understanding the relationships
between the timescales of magma genesis, magmatic heat flux and potential triggering
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.

#### 116 ASCENSION ISLAND

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;
- 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
- 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
- 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

within the middle crust, which were inferred to be the crystallised remains of a relict magmachamber.

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# 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 products of Ascension: <sup>143</sup>Nd/<sup>144</sup>Nd varies between 0.51292 and 0.51310, and significant 148 149 variation in <sup>87</sup>Sr/<sup>86</sup>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 the significant variation in <sup>87</sup>Sr/<sup>86</sup>Sr coupled to variations in <sup>143</sup>Nd/<sup>144</sup>Nd (Fig. 2a). The large 155 variation in <sup>87</sup>Sr/<sup>86</sup>Sr has been suggested to reflect post-emplacement alteration of samples 156 157 (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).

Previous studies utilised the trace element variation in basaltic magmas erupted on Ascension Island to infer source composition and to define three main magmatic groups, 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

The magmatism on Ascension Island defines a transitional to mildly-alkaline, silica undersaturated array from olivine basalt - hawaiite - mugearite - benmoreite - trachyte - rhyolite (Daly, 1925; Weaver et al., 1996; Fig. 3). Mafic volcanic products are erupted all across the island (Fig. 1), while felsic products are limited to central and eastern areas (Fig. 1). Previous authors have divided the silicic eruptive products into two main centres: the older (Kar et al., 1998; Hobson, 2001; Jicha et al., 2013) central felsic region, which contains the oldest dated exposed lava on the island (at 1094 ka, Jicha et al., 2013), and the younger eastern complex
(youngest published Ar-Ar date of 52 ± 3 ka; Jicha et al., 2013; Fig. 1). Felsic magmas are
inferred to be the product of high degrees of fractional crystallisation (Fig. 2c; Weis et al.,
1987; Kar et al., 1998; Webster & Rebbert, 2001; Jicha et al., 2013) originating chiefly from
similar mafic melts to those erupted around the peripheries of the felsic complexes (both high
and intermediate Zr/Nb basalts have been suggested as parental melts; Figs. 1, 2c; Weaver et
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.

#### 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

less than 16 mm) and all lithic clasts were removed by hand. These samples were then
thoroughly cleaned by removing any adhering matrix or oxidised rind followed by soaking in
(frequently changed) milli-RO water for a minimum of one week. Lava was treated by
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	Brucker-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 $P_2O_5$ 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 <i>in-situ</i> 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 <i>in-situ</i> 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

major elements (>1 wt.% concentration) were always within 2 relative % (2 s.d.); with slightly higher uncertainties for minor elements (see Electronic Appendix 1 for further details on accuracy and precision of secondary standards). Due to their hydrous nature, only glass analyses with totals of <93 wt.% were set aside; values for the remaining analyses were then 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 µm spot (for glass) or 50 µm spot (for crystals). The LA-ICPMS data were internally normalized to <sup>29</sup>Si or <sup>43</sup>Ca from EPMA analyses. Abundances of single trace elements were 274 275 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.% SiO<sub>2</sub>, 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.% SiO<sub>2</sub>, 10.3 wt.% TA; Fig. 3a, 286 Table 2). When selected whole rock trace element data are normalised to primitive mantle (Palme & O'Neill, 2003) felsic lava and pumice samples are clearly depleted in Sr, Ti and Ba 287 288 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 Fo49 – Fo89. Where groundmass olivine can be identified separately to phenocrysts, 338 groundmass olivines have higher CaO and lower MnO concentrations at any given value of

Fo number. Olivine crystals are faintly normally-zoned, if they are zoned at all, with noevidence 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<sub>20</sub>Ab<sub>75</sub>Or<sub>5</sub> and Ab<sub>70</sub>Or<sub>30</sub> (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<sub>20</sub>Ab<sub>75</sub>Or<sub>5</sub>) through to anorthoclase (Ab<sub>70</sub>Or<sub>30</sub>; 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. Again and the second state of the second state 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 Fo12.5 (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>-

 $Ab_{70}Or_{30}$ ) 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  $(An_7Ab_{73}Or_{20} - Ab_{70}Or_{30})$  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  $\pm$  clinopyroxene  $\pm$  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<sub>14</sub>Ab<sub>72</sub>Or<sub>14</sub> and An<sub>84</sub>Ab<sub>15</sub>Or<sub>1</sub>. 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<sub>2</sub> (< 50 wt.%) and high CaO (> 6 wt.%), (Fig. 12, Table 8); the bulk 400 scoria composition is slightly less-evolved  $(45.8 - 49.3 \text{ wt.}\% \text{ SiO}_2)$  than the NE Bay scoria 401 (AI14-438, 49.0 - 52.4 wt.% SiO<sub>2</sub>).

402

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

404Pumice clasts are typically crystal poor, with < 5% crystals. Ternary feldspar is the dominant</th>405crystal phase (oligoclase to anorthoclase composition  $\pm$  sanidine) with one ferromagnesian406phase of either amphibole or fayalitic olivine, with minor magnetite  $\pm$  ilmenite  $\pm$  apatite, and407rarely augitic clinopyroxene is present. One exception to this is the intermediate sample408AI14-459 which has ~15% crystals, with large amphibole (up to 3 mm) phenocrysts. Crystal409phases are not zoned in BSE, and preserve euhedral crystal habits. Groundmass is typically410glassy, with varying amount of feldspar microlites.

Feldspar compositions from these pumice fall deposits intersect and straddle those from the evolved effusive eruptions (Fig. 7, Table 4): with compositions ranging between An<sub>51</sub>Ab<sub>47</sub>Or<sub>2</sub> and An<sub>0.5</sub>Ab<sub>58,5</sub>Or<sub>41</sub>. Some eruptions have relatively restricted feldspar 414 compositions (e.g. AI14-488: An<sub>0.5</sub>Ab<sub>65.5</sub>Or<sub>34</sub> - An<sub>1</sub>Ab<sub>69</sub>Or<sub>30</sub>) whereas other extend over wide 415 ranges (e.g. AI14-435: An<sub>2</sub>Ab<sub>73</sub>Or<sub>25</sub> – An<sub>18</sub>Ab<sub>76</sub>Or<sub>6</sub>). 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<sub>51</sub>Ab<sub>47</sub>Or<sub>2</sub>-419 An<sub>20</sub>Ab<sub>74</sub>Or<sub>6</sub>; rims An<sub>43</sub>Ab<sub>54</sub>Or<sub>2</sub> – An<sub>20</sub>Ab<sub>74</sub>Or<sub>6</sub>). 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  $\sim 66$  wt.% to 75 wt.% 427 SiO<sub>2</sub> (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 (see464 Electronic Appendix 2 for full XRF results).

465

484

#### 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,

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

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

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

485 be partly responsible for the radiogenic Sr isotopic compositions of more felsic melts (Kar et

evolved melts at Ascension. Some minor assimilation and contamination has been inferred to

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 <sup>87</sup>Sr/<sup>86</sup>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 TiO<sub>2</sub>, K<sub>2</sub>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.% H<sub>2</sub>O, and fO<sub>2</sub> 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  $\epsilon$ 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.% H<sub>2</sub>O) from feldspar-518 melt hygrometry, and  $fO_2$  -1.5  $\Delta$ 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.% SiO<sub>2</sub>), 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 H<sub>2</sub>O with increasing SiO<sub>2</sub> support the modelled high melt H<sub>2</sub>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*

Fractional crystallisation, as modelled by MELTS, has demonstrated that the observed whole rock compositional range within the Ascension Island magmatic suite can be reproduced in this process. For the mafic and intermediate magmas, the crystal compositions measured overlap well with the modelled crystallising phases from MELTS (Fig. 14), and up to 70% total fractionation has occurred to produce the most evolved intermediate magmas (Fig. 13). Whilst MELTS details proportions of phases fractionating, previous studies have
demonstrated some limitations on the model's ability to accurately predict crystallisation of
water-bearing phases (Gualda et al., 2012), clinopyroxene (Fowler & Spera, 2010) and apatite
(Rooney et al., 2012). Therefore, to yield further insights into proportions of phases
crystallising least-squares modelling of the major elements has been undertaken, constrained
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 Electonic 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  $\sim 8\%$  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  $\sim 4 - 6$  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 favalitic 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 Kd 623 of ~ 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<sub>2</sub>O feldspar and aegirine-augite are only found in felsic 630 lavas; in felsic pyroclastic samples no feldspar has 4 - 6 wt.% K<sub>2</sub>O 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),

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

In other alkaline systems it has been shown that variations in  $fO_2$  at constant temperatures can result in variable phase assemblages (White et al., 2009; Markl et al., 2010). Given the large variation in calculated  $fO_2$  within Ascension Island magmas (Table 8), some of the small variation around liquid lines of descent, and mismatch between the MELTSmodelled vs. observed phases, may stem from variable phase crystallisation at differing  $fO_2$ 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 An82, and Fo87; Fig 8, 10), whereas the Letterbox

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

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.

Mafic lavas, excluding those from the South Coast group, are clearly in chemical
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 g/cm<sup>3</sup>, 689 remarkably similar to that of plagioclase feldspar  $(2.6 - 2.7 \text{ g/cm}^3, \text{ Scoates}, 2000; \text{ Ghiorso } \&$ 690 Gualda, 2015), yet less dense than olivine and clinopyroxene, (~3.6 g/cm<sup>3</sup> and ~3.2 g/cm<sup>3</sup> 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).

*Role of crystal entrainment in mafic to intermediate magma genesis:* Excluding the
South Coast group lavas, all other mafic and intermediate lavas have crystallinities less than
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<sub>2</sub>O 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

highlight the lateral variability in plutonic or mushy regions within the Ascension Islandcrust.

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.

The presence of plutonic rocks in ocean island crust has long been known to affect the evolution of magmas prior to eruption—the volcanic islands of Terceira (Mungall & Martin, 1995) and Oki-Dōzen (Brenna et al., 2015), among others, have evidence for central felsic complexes (both as plutonic rocks, and as storage regions for felsic volcanic eruptions) which 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).

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

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

813

# 814 The magmatic plumbing system

Here we combine our new results with all previously published data to present a model for
the magmatic plumbing system underlying Ascension Island (Fig. 16) during the subaerial
phase of activity from ~1 Ma to present. The key features of the model are as follows:
1) A zone of melt extraction from which all Ascension Island volcanism is sourced, at
varying degrees of partial melt (Fig. 16; Paulick et al., 2010; Jicha et al., 2013). This

melt extraction zone is geochemically distinct from the mantle melting responsible for
the significantly larger volume submarine volcanism of Ascension which was active
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

deep mafic plutonic rocks is reflected in the proportions of antecrysts incorporatedduring ascent of mafic magmas.

3) A central nested region of small discrete plutonic bodies of varying composition (of a
higher proportion of felsic bodies), which forces the majority of ascending mafic
magmas to stall and fractionate until they reach a level where their buoyancy has
increased enough to allow them to ascend further. This is supported by the felsic
plutonic lithic clasts within the Green Mountain scoria, erupted in the central felsic
complex, and by mechanically incorporated plutonic lithic fragments in many felsic
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 6.8 – 10.2 km, assuming a crustal density of 3000 kgm<sup>-3</sup>; Webster & Rebbert, 2001). 849 850 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.

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

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

6) The magmas that fed explosive and effusive felsic eruptions exhibit differences in the presence or absence of low pressure phases (Fig. 14) perhaps suggestive of variation in the location of their storage regions and ascent rates, and warrants further investigation. However, fractional crystallisation (with minor crystal entrainment) of predominantly feldspars induced by stalling (at varying depths) in the nested (more felsic) plutonic region, remains the dominant evolutionary process for all felsic 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 et907 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, Cole et al., 1995]), then  $\sim 2 \text{ km}^3$  of fractionated crystals, preserved as plutonic rocks, remain 919 920 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,

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

antecrysts during ascent, consistent with the observation of increased numbers ofcoherent plutonic lithic fragments in these lava flows.

Felsic melt evolution at Ascension Island is dominantly controlled by crystal
fractionation of ternary feldspar with minor fayalite or sodic clinopyroxene,

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).

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

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## **1317 FIGURE CAPTIONS**

1318 Figure 1: Geological map of Ascension Island, located 90 km from the Mid Atlantic Ridge

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;
- eruptive centres identified in grey font. Surface expressions of known faults outlined in
- 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 <sup>143</sup>Nd/<sup>144</sup> Nd with degree of evolution (MgO). All
- data presented is a combination of Weaver et al. (1995, grey diamonds), Kar et al.
- (1998, orange diamonds), Paulick et al. (2010, blue squares) and Jicha et al. (2013,crosses).
- 1332 Figure 3: (a) Whole rock compositions of selected samples compared with all known
- Ascension Island whole rock data in grey field- (data from Weaver et al., 1996, Kar et
- 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).
- Figure 4: Whole rock variation in major and trace elements for all studied samples of
  Ascension Island mafic-intermediate lavas (purple triangles), felsic lavas (blue
  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 $SiO_2$
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 $\pm$ clinopyroxene $\pm$
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

1366compositions of feldspars from mafic to intermediate lava samples. The pink field on1367panel (b) shows the Mid Atlantic Ridge ocean crust feldspars from the Kane Fracture1368Zone (Coogan et al., 2000). Core-rim differences within individual samples shown by1369filled (core) and open (rim) symbols in (c); sample colours and shapes refer to those1370listed 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)

equilibrium compositions of feldspar are calculated at 1150 °C and 330 MPa (black)

and 1050 °C and 330 MPa (grey) respectively, based on feldspar-melt and Fe-Ti oxide

thermometry (see Table 8), and the pressure at the base of the crust at Ascension Island(Klingelhöfer, 2001).

Figure 10: Compositions of olivines from (a) mafic to intermediate lavas; (b) all pyroclasticsamples. 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

1381those 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.

1383Figure 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

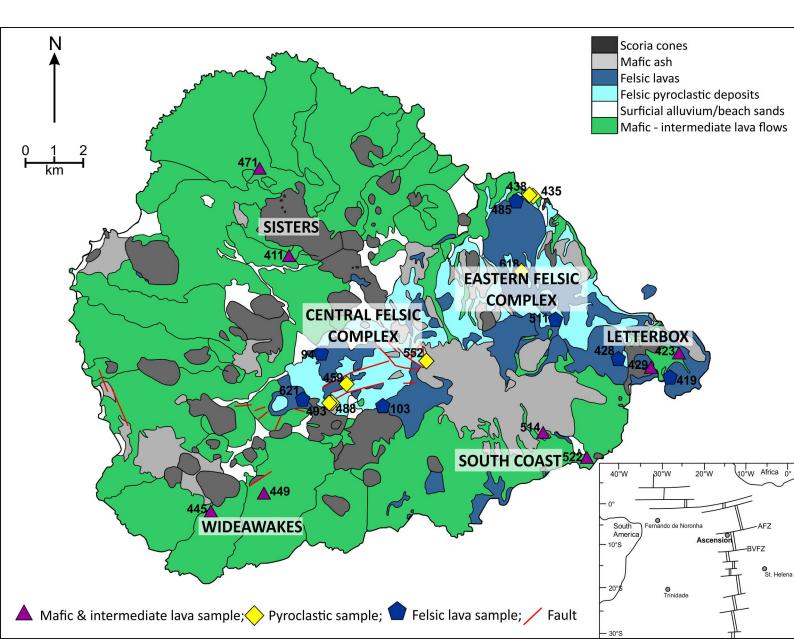
1386Appendix 2.

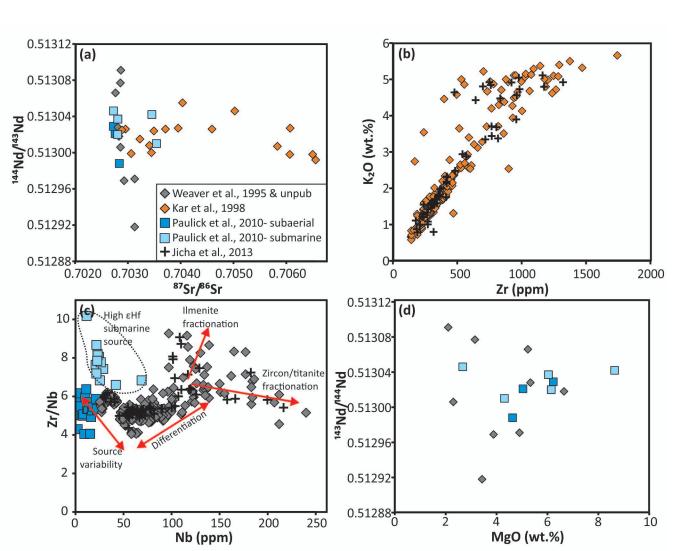
Figure 12: Matrix glass compositions from all Ascension Island samples. Sample areas relateto those used in Table 1. For full data set see Electronic Appendix 2.

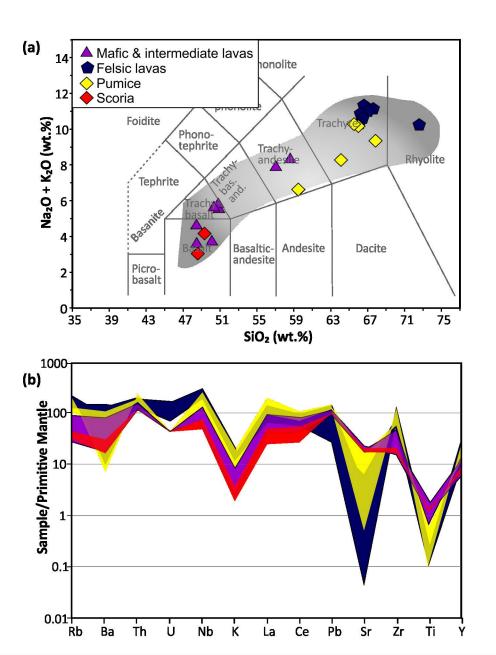
1389 Figure 13: Combined liquid lines of descent modelled from AI14-438 (to 950 °C, end marked

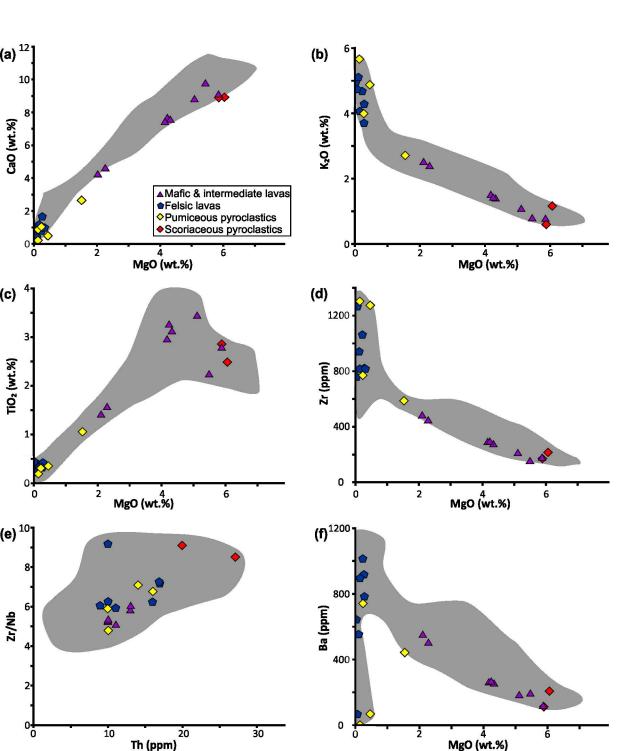
1390 by dashed line) and AI-621 (< 950 °C, start from dashed line) using rhyolite-MELTS

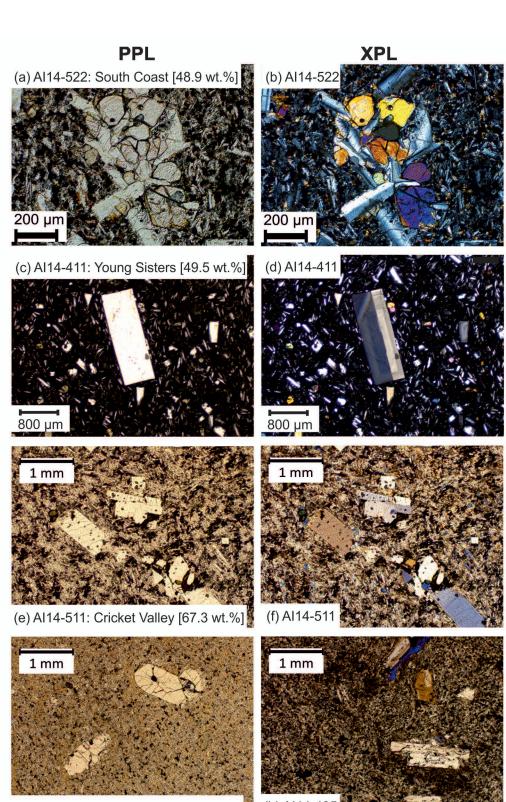
1391	(Gualda & Ghiorso, 2015) at 330 MPa, 250 MPa, 170 MPa and 90 MPa. Purple ticks
1392	mark 20% increments of melt fraction remaining; grey ticks mark 10% melt fraction
1393	remaining, black ticks show liquid composition at 5% melt fraction remaining. Initial
1394	water concentrations for AI-438 were 0.5 wt.% $H_2O$ , 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 $(1 - 4, \text{ connected by})$
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</sup> , <sup>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).





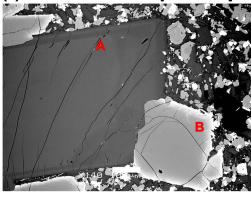




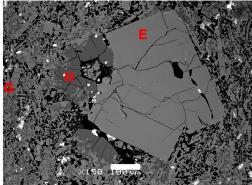


(g) AI14-428: White Horse [66.8 wt.%] 🦉 (h) AI14-485

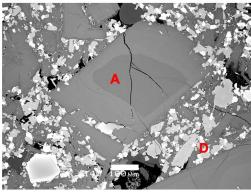
(a) AI-522: South Coast [48.9 wt.%]



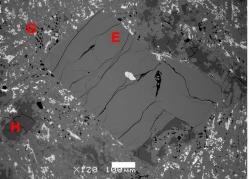
(d) AI14-428: White Horse [66.8 wt.%]



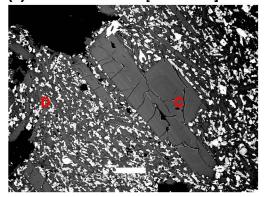
(b) AI-522: South Coast [48.9 wt.%]



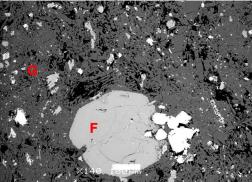
(e) AI-511: Cricket Valley [67.3 wt.%]

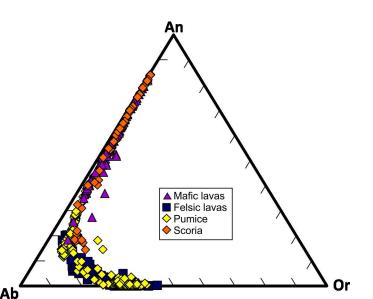


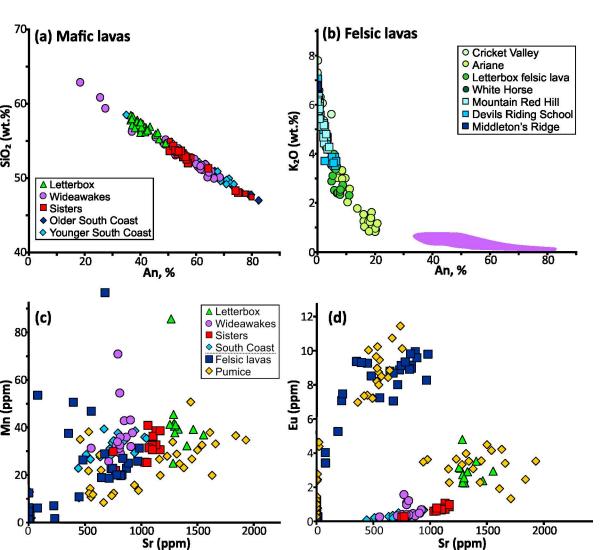
(c) AI-423: Letterbox [57.6 wt.%]

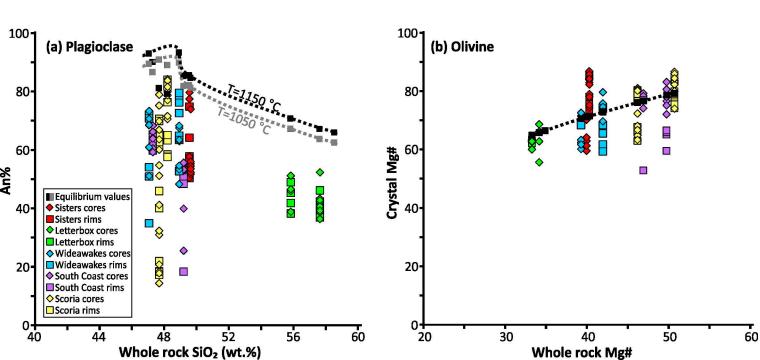


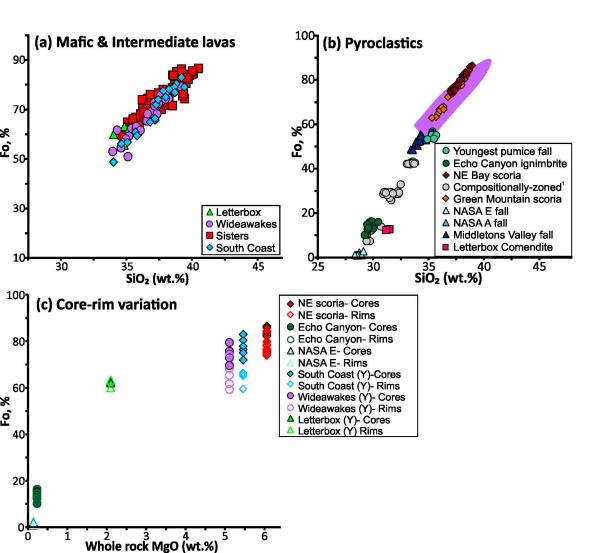
(f) AI-485: Ariane [65.9 wt.%]

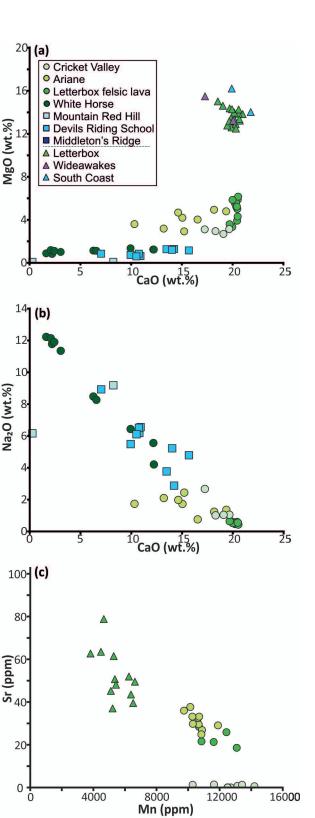


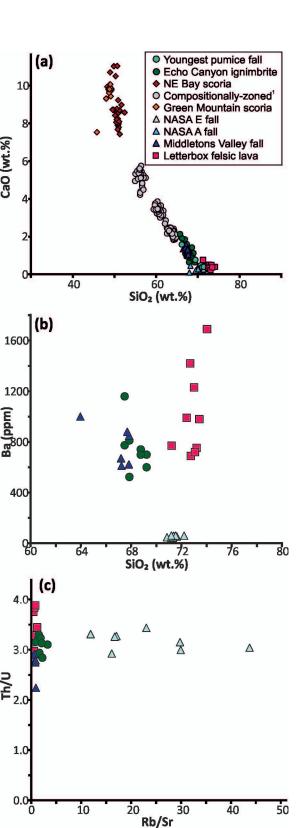


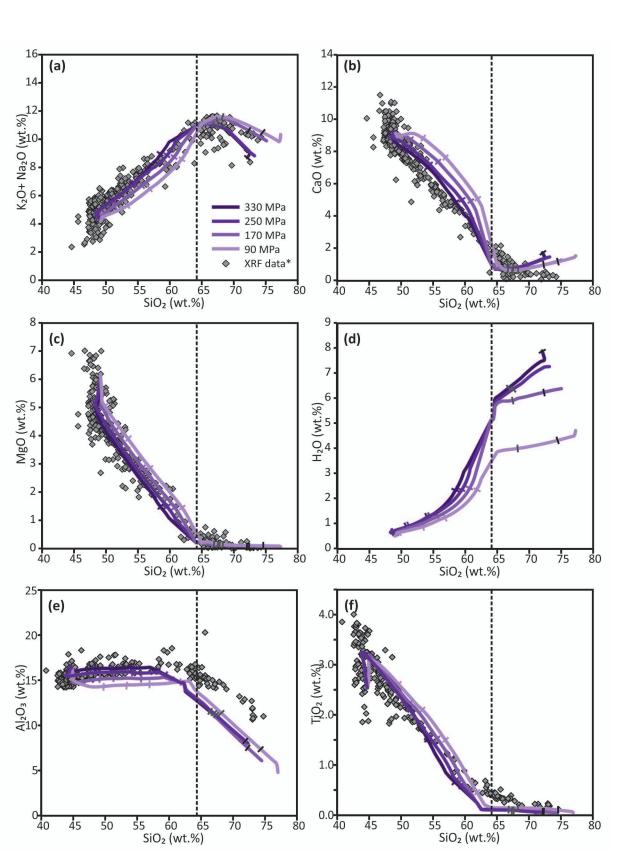


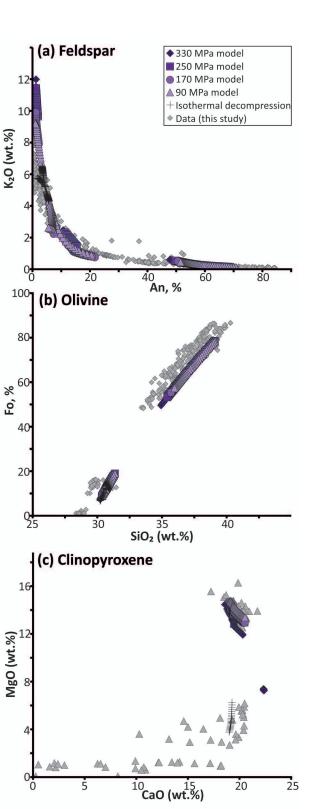


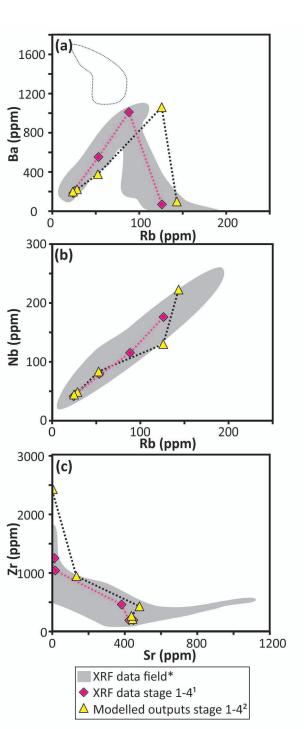


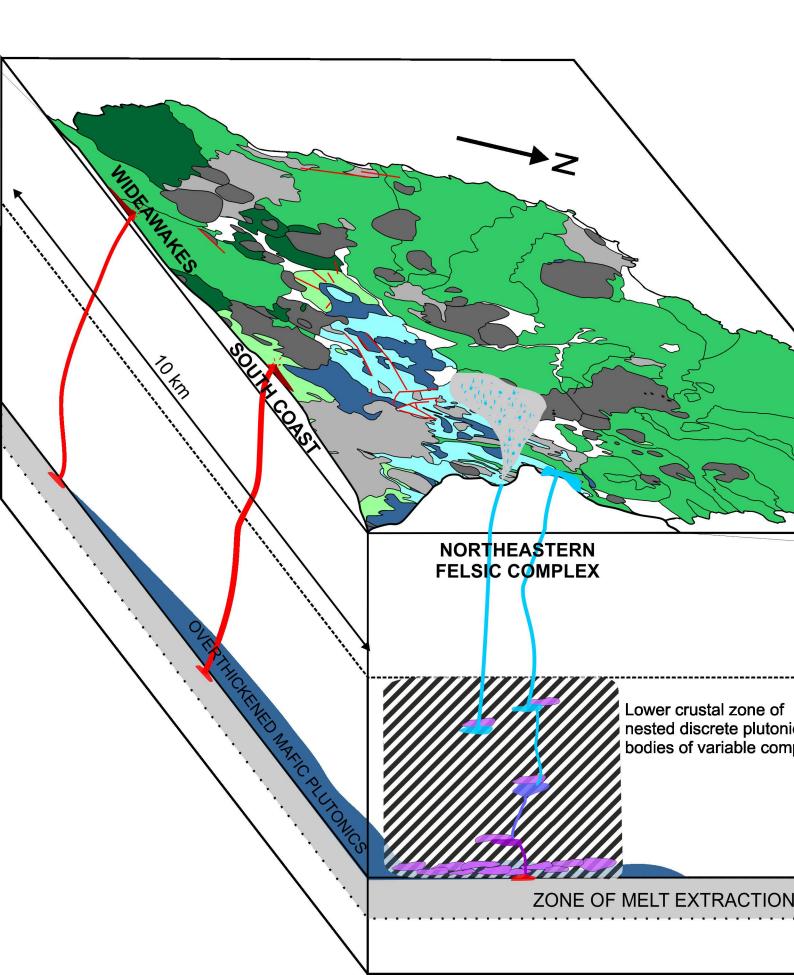












Sample	Region	TAS	Sample	Grid Reference	Approximate
number		classification	type	(E, N)	age of sample
AI14-411	Youngest Sisters	Trachy-basalt	mafic lava	0568706, 9123576	zero age *
AI14-471	Older Sisters	Trachy-basalt	mafic lava	0567966, 9125332	> AI14-411¥
AI14-423	Youngest Letterbox	Trachy-andesite	mafic lava	0577052, 9121328	
AI14-429	Older Letterbox	Trachy-andesite	mafic lava	0576443, 9121020	> AI14-423¥
AI14-445	Youngest Wideawakes	Basallt	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+	Compositionally-zoned top	Trachy-andesite	zoned fall	0573786, 9124790	
AI14-439D+	Compositionally-zoned mid	Trachy-andesite	zoned fall	0573786, 9124790	
AI14-439G⁺	Compositionally-zoned base	Trachyte	zoned fall	0573786, 9124790	

 Table 1: Samples investigated for this study

\* Ar-Ar age data from Jicha et al., 2014
\* K-Ar age data from Harris et al., 1992

¥ Stratigraphic relationship observed in the field

Sample number	AI114-411	AI14-471	AI14-423	AI14-429	AI14-445	AI14-449	AI14-522	AI14-514	AI14-485	AI14-!
Location	Young Sisters	Old Sisters	Young Letterbox	Old Letterbox	Younger Wideawakes	Older Wideawakes	Younger South Coast	Older South Coa	Ariane	Cricke
SiO <sub>2</sub>	49.5	49.6	57.6	55.8	47.3	49.2	48.9	47.0	65.9	67.3
TiO <sub>2</sub>	3.12	2.97	1.42	1.58	3.44	3.26	2.25	2.79	0.42	0.37
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
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
MnO	0.22	0.22	0.24	0.23	0.20	0.23	0.16	0.18	0.22	0.16
MgO	4.33	4.16	2.10	2.29	5.11	4.24	5.46	5.87	0.28	0.11
CaO	7.55	7.42	4.23	4.62	8.80	7.70	9.8	9.13	1.6	0.88
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
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
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
LOI	-0.10	-0.02	-0.45	-0.51	-0.21	-0.34	0.24	-0.14	0.12	0.00
Total	99.0	98.8	99.0	98.1	99.1	98.9	97.8	97.3	100.1	100.0
Ва	254	263	553	503	183	266	194	120	780	554
Rb	32	34	53	50	24	29	17	16	76	117
Sr	455	461	381	410	423	458	501	391	135	7
Pb	18	20	20	21	17	19	bdl	bdl	5	8
Th	10	10	13	13	11	10	bdl	bdl	9	16
U	bdl	bdl	1	1	1	1	bdl	bdl	bdl	3.7
Zr	279	291	483	450	214	292	158	182	818	940
Nb	52	55	80	77	42	54	28	29	135	151
Y	41	44	54	52	27	49	24	30	85	62
La	34	35	64	56	28	41	bdl	bdl	71	53
Ce	96	105	144	140	74	115	61	49	156	119
Sc	21	19	12	12	24	18	26	28	9	9
V	227	203	42	55	317	208	252	314	3	4
Ni	8	5	3	5	24	3	56	65	5	3
Zn	122	128	128	132	115	130	83	105	150	126
Sample number	AI114-419	AI15-621	AI15-618	AI14-435A	AI14-438	AI14-493A	AI14-488A	AI14-459/	4	AI14-!
Location	Letterbox	Devils	Youngest	Echo Canyon	NE scoria	NASA unit E	NASA unit A	Middletor	n's	Green
	Felsic lava	<b>Riding School</b>	Pumice fall					Valley fall		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

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

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
$P_2O_5$	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
Ва	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

Sample	Region	TAS	Percentage	Vesicularity	Fspar	OI	Срх	Cbt	Aeg
number	0	classification	Phenocrysts*	(%)	•		•		0
AI14-411	Youngest Sisters	Trachy-basalt	<1 (f)	5	67	5	20		
AI14-471	Older Sisters	Trachy-basalt	<1 (f)	7	68	10	17		
AI14-445	Youngest Wideawakes	Basallt	3 (f, ol)	3	68	14	12		
AI14-449	Older Wideawakes	Trachy-basalt	<1 (f, ol)	6	55	9	32		
AI14-423	Youngest Letterbox	Trachy-andesite	<1 (f)	5	80	6	10		
AI14-429	Older Letterbox	Trachy-andesite	2 (f)	43	77	8	9		
AI14-522	Youngest South Coast	Basalt	40 (f, ol)	3	62	31	<1		
AI14-514	Older South Coast	Basalt	18 (f, ol)	23	56	23			
AI14-428	White Horse	Trachyte	2 (f)	24	64			29	4
AI14-511	Cricket Valley	Trachyte	6 (f, aeg)	1	78			12	9
AI14-485	Ariane flow	Trachyte	4 (f)	15	77			19	
AI14-419	Letterbox felsic lava	Trachyte	<1 (f)	25	60		1	1	
AI-103	Mountain Red Hill	Trachyte	2 (f, ox)	4	70			24	5
AI15-621	Devil's Riding School	Trachyte	3 (f)	13	68			27	4
AI-94	Middleton's Ridge	Rhyolite	<1 (f)	18	57			35	6

**Table 3**: Petrography of Ascension lavas

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

Crystal percentages represent groundmass ( $<500 \mu m$ ) crystals as well as those represented as phenocryst phases; ternary); Ol = olivine; Cpx = clinopyroxene; Cbt = cristobalite; Aeg = aegirine; Ox+Acc = bright in Back Scatter accessory phases including zircon and apatite.

\*Phenocrysts used to represent phases > 500  $\mu$ m in thin sectioned area. Phases which constitute phenocryst phase of = olivine; cpx = clinopyroxene; aeg = aegirine; ox = oxides;

Sample Location Rock type	<b>Al114-411</b> Young Sisters Trachy-basalt				<b>AI14-423</b> Young Letterbox Trachy-andesite		<b>AI14-445</b> Young Wideawake Basalt	es	<b>AI14-522</b> Young So Basalt	
Spot location	F3-C	F3-R	F5-C	F5-R	F1-C	F1-R	F5-C	F5-R	F1-C	F1-R
SiO <sub>2</sub>	53.50	53.29	47.46	52.69	56.56	56.19	52.57	51.22	48.05	51.89
TiO <sub>2</sub>	0.16	0.17	0.05	0.13	0.04	0.06	0.13	0.16	0.05	0.10
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
FeO*	0.91	0.92	0.45	0.77	0.37	0.49	0.77	0.87	0.46	0.66
MgO	0.10	0.09	0.13	0.11	0.04	0.05	0.12	0.13	0.15	0.15
CaO	11.45	11.61	16.66	12.19	8.93	8.88	12.20	12.80	15.92	13.46
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
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
Total	99.76	99.70	99.91	100.15	99.75	99.23	99.10	98.87	99.86	100.0
An	54.6	54.7	79.7	57.8	42.1	41.7	59.3	61.4	76.3	63.4
Ab	43.5	43.5	19.9	40.7	55.9	56.3	39.3	37.2	23.1	35.6
Or	1.8	1.9	0.4	1.5	2.0	2.0	1.4	1.4	0.6	1.0
Sc	4.55	4.90	4.64	4.60	4.6	3.11	6.38	12.0	8.48	7.22
Ti	421	612	696	664	254	344	797	4970	508	526
V	0.53	1.60	1.73	1.58	0.42	0.51	3.49	83	3.12	2.41
Mn	32.8	32.6	36.5	38.8	37.7	36.9	37.9	323	37.8	28.3
Ga	21.2	20.0	24.9	23.0	24.8	26.0	24.7	22.9	23.1	23.1
Rb	0.31	0.45	0.28	0.14	0.86	0.56	0.93	2.90	0.24	0.32
Sr	1130	1096	1104	1168	1289	1317	922	811	793	718
Y	0.19	0.18	0.20	0.40	0.64	0.40	0.57	9.00	0.11	0.08
Zr	0.41	0.22	0.00	0.11	2.80	1.54	1.26	31.5	0.50	0.17
Ва	134	137	121	175	295	324	109	131	70.7	86.2
La	3.16	2.76	2.46	2.71	5.88	6.40	2.13	9.23	1.08	1.10
Ce	5.21	3.73	3.61	4.33	8.62	6.47	4.00	20.0	1.93	1.55
Pr	0.54	0.41	0.24	0.42	0.90	0.81	0.37	2.33	0.17	0.18
Nd	2.24	0.99	1.00	1.55	2.10	2.36	1.72	11.4	0.59	0.70
Sm	0.23	0.09	0.34	0.41	0.57	0.19	0.31	2.37	0.04	bdl
Eu	1.07	0.68	0.78	0.98	3.04	2.50	0.68	1.21	0.38	0.39
Gd	0.17	0.27	0	0.25	0.35	0.20	0.25	2.44	bdl	0.06
Yb	bdl	bdl	bdl	bdl	0.13	bdl	0.01	0.61	0.02	bdl
Pb	0.40	0.22	0.26	0.22	0.84	0.65	0.23	0.63	0.22	0.10

Table 4: Representative feldspar data for Ascension samples

Sample	AI114-485	AI14-511		AI14-419		AI14-4		AI14-49		AI14-488		AI14-4
Location	Ariane	Cricket Valley	/	Letterbox fel	sic lava	Echo C	•	NASA U		NASA Un		Middl
Rock type	Trachyte	Trachyte <b>F2-C</b>	F2-R	Trachyte <b>F3-C</b>	F3-R	Trachy F4-C	te F4-R	Trachyt <b>F26-C</b>		Trachyte		Trachy
Spot location	65.60	65.20	<b>г2-к</b> 65.89	<b>F3-C</b> 65.20	<b>гз-к</b> 64.76	65.21	<b>г4-к</b> 65.91	67.98	68.53	<b>K13-C</b> 68.51	<b>K13-R</b> 67.88	<b>К18-С</b> 62.04
SiO₂ TiO₂	0.03	0.02	0.01	0.00	0.00	0.00	0.01	0.00	08.55	0.00	07.88	0.02
Al <sub>2</sub> O <sub>3</sub>	0.03 19.42	19.33	19.00	20.71	20.90	21.22	20.57	0.00 19.24	0.00 19.53	0.00 19.36	19.31	
FeO*												22.50 0.34
	0.53	0.32	0.31	0.17	0.20	0.22	0.18	0.29	0.24	0.28	0.32	
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
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
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
K₂O	4.37	6.30	6.54	2.40	2.46	1.12	1.40	6.64	6.36	6.08	5.92	0.94
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
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
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
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
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
Ti	156	104	76.5	6.89	124	105	104	62.0	84.0	69.8	67.7	229
V	0.31	0.45	0.17	0.02	bdl	0.11	bdl	bdl	1.70	0.09	0.01	bdl
Mn	20.1	1.20	0.60	2.55	7.00	22.1	11.8	1.10	bdl	2.10	bdl	19.9
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
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
Sr	828	1.31	0.81	13.0	222	644	571	0.79	2.28	2.35	1.46	1169
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
Zr	2.70	3.22	0.45	2.17	bdl	0.00	0.00	bdl	2.90	0.30	0.47	bdl
Ва	1261	120	80.6	96.7	3770	1680	1552	66.0	129	194	97.4	533
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
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
Pr	0.74	0.02	bdl	0.07	0.72	0.48	0.52	0.00	0.00	0.00	0.00	1.01
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
Sm	0.26	0.03	bdl	0.05	0.22	0.00	0.00	0.00	0.00	0.00	0.00	0.00
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
Gd	0.18	0.01	bdl	0.04	0.35	0.00	0.00	0.00	0.00	0.00	0.00	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
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

Major element data given as wt.%; Anorthite, albite and orthoclase (An, Ab, Or) contents as mol. %; trace elements are below the detection limits of the method, bdl is written. Spot loca core [C] or rim [R].

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

Sample Location	AI14-411 Young Sisters		AI14-423 Young Let	terbox	AI14-44 Young	15	AI14-522 Young South Coa	ast	AI14-439 Echo Car		<b>AI14-49</b> NASA ur	
Location	Toung Sisters		Toung Let		Wideav	vakes	Toung South Col		Leno ca	iyon		
Rock type	Trachy-basalt		Trachy-an	desite	Basalt		Basalt		Trachyte	2	Trachyte	ž
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				
Total	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 App analysis being core [C] or rim [R].

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

Sample Location Rock type	AI14-423 Young Le Trachy-ai	tterbox	AI14-429 Old Lette Trachy-a	erbox	<b>AI14-44</b> 9 Old Wide Trachy-b	eawakes	<b>Al14-52</b> Young Se Basalt	<b>2</b> outh Coast	<b>AI14-485</b> Ariane Trachyte	AI14-51 Weather Trachyte	rpost	<b>Al14-42</b> White H Trachyte	orse	AI14-41 Letterbo Trachyte
Spot tag	2_C	2_R	GM	GM	1_C	1_R	1_C	1_R	GM	GM	GM	GM	GM	1_C
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
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
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
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
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
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
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
Na₂O	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
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
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
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

Major element data given as wt.%. For full data set, see Electronic Appendix 2. Spot location refers to the anal groundmass [GM].

**Table 7:** Representative glass analyses from Ascension Island**7a:** Major element analyses normalised to 100 wt.%

Sample	AI14-419		AI15-618	1	AI14-552	2	AI14-438	3	AI14-43	5A		AI14-49	3A	AI14-48
Location	Letterbox f	elsic lava	Young pu	imice	Green M	lountain sc.	NE scoria	3	Echo Ca	nyon		NASA ur	nit E	NASA ur
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
SiO2	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
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
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
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
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
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
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
Na₂O	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
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
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
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
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
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
Total	100	100	100	100	100	100	100	100	100	100	100	100	100	100
Maior	alamant	t data a	ivon og	wt 0/	For full	data sat	soo El	aatroni	a Annai	div 2				

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 b method, bdl is written. Analyses in italics previously published in Chamberlain et al., 2016.

Sample	AI14-419	)	AI14-43	5A	AI14-49	3A	AI14-45	9A		AI14-43	9D	AI14-43	9G	
Location	Letterbox	x felsic lava	Echo Ca	nyon	NASA ur	nit E	Middlet	on's Valley	fall	Compos	itionally-zo	ned mid (D)	and upper	· (G)
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	13.6	18.0	29.0	20.0	18.0
Sc	13.6	15.9	13.8	12.8	9.70	9.40	5.00	11.1	13.0	11.2	21.0	27.0	10.1	27.0
Ti	994	1020	1705	1300	1448	1437	5400	4960	5400	2360	5690	4200	4530	2900
v	1.60	1.20	0.71	bdl	4.37	6.80	10.1	10.0	21.0	bdl	5.30	1.10	33.8	11.0
Mn	863	820	1544	1290	1259	1270	1720	1060	1190	1160	2720	2130	1290	1550
Zn	84	90	154	215	268	239	178	113	94	97	164	134	107	155
Ga	34	32	30	32	39	42	57	38	33	16	37	35	27	17
Rb	101	58	90	90	169	182	166	134	161	44	73	97	56	64
Sr	90	106	50	44	10	8	246	175	220	175	390	184	117	159
Y	76	49	77	80	124	129	52	44	38	34	74	70	41	79
Zr	930	584	791	868	1507	1661	1180	1000	1310	286	620	760	490	830
Nb	133	89	147	161	234	247	160	135	141	64	138	141	83	98
Ва	746	1240	790	720	55	61	950	651	1020	470	900	1290	840	1300
La	76	51	83	83	138	145	93	65	71	41	73	81	54	79
Ce	160	113	169	178	274	283	197	124	145	75	160	153	102	153
Pr	18	12	19	20	30	30	17	12	9	7	19	15	12	19

Nd	67	50	73	77	112	115	57	43	45	33	85	60	43	75
Sm	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
Eu	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
Gd	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
Dy	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
Er	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
Yb	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
Lu	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
Hf	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
Та	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
W	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
Pb	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
Th	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
U	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 Is	sland samples
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Sample number Al14-411 Al14-471 Al14-423 Al14-429 Al14-445	Region Youngest Sisters Older Sisters Youngest Letterbox Older Letterbox Youngest Wideawakes	FeTi-Oxide T (°C) 935  990 1025 985	Thermometry <sup>1</sup> fO <sub>2</sub> (ΔΝΝΟ) -0.4  -0.05 +0.08 -0.34	Feldspar-melt T T (°C) 1150 1152 1093 1110 1167	hermometry <sup>2,3</sup> wt.% H <sub>2</sub> O <sup>4</sup>
Al14-511 Al14-428 Al-94 Al-103 Al14-419 Al15-621	Cricket Valley White Horse Middleton's Ridge Mountain Red Hill Letterbox felsic Iava Devil's Riding School	884  967  	-1.6  -1.9 	886 837 772 801 837 871	5.2 5.5 4.7 7.2 5.1
Al15-618 Al14-435A Al14-438 Al14-493A Al14-488A Al14-459A Al14-552	Youngest pumice fall Echo Canyon NE scoria NASA unit E NASA unit A Middleton's Valley fall Green Mountain scoria	    950 	   -0.2	847 993 1164 811 813 1034 1174	6.4 7.1 8.1
AI14-439A⁺ AI14-439G⁺	Compositionally-zoned scoria Compositionally-zoned pumice	845 866	-2.28 -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  $K_D(Ab-An)$  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.