

Constraining magma storage conditions at a restless volcano in the Main Ethiopian Rift using phase equilibria models

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ABSTRACT

The Main Ethiopian Rift hosts a number of peralkaline volcanic centres, several of which show signs of recent unrest. Due to the low number of historical eruptions recorded in the region and lack of volcanic monitoring, conditions of magma storage in the Main Ethiopian Rift remain poorly constrained. Aluto is one of these restless volcanic centres and identifying magma storage conditions is vital for evaluating the significance of recent periods of unrest. Using Aluto as a case study, we explore magma storage conditions using Rhyolite-MELTS thermodynamic modelling software. We performed ~150 fractional crystallisation models using a primitive basalt as the starting composition, and for a range of pressures (50 – 300 MPa), initial H₂O contents (0.5 – 3 wt%) and oxygen fugacities (QFM-2 – QFM+1). Predicted liquid lines of descent from these models are compared with published whole-rock data and, together with new observations of natural phase assemblages and erupted mineral compositions, provide constraints on magma storage conditions.

Using a statistical approach to compare empirical data and thermodynamic model outputs, we find that compositions of evolved peralkaline rhyolites from Aluto are best reproduced by protracted (90%) isobaric fractional crystallisation from a rift-related basaltic composition, without the need for

significant crustal assimilation. The required extent of fractional crystallisation suggests that much of the magmatic system may exist as a highly crystalline mush with only a small lens of rhyolitic melt. The best agreement between models and natural samples is at low pressures (150 MPa), low initial H₂O concentrations (0.5 wt%) and an oxygen fugacity near the QFM buffer. The depth of magma storage derived from these results ($\sim 5.6 \pm 1$ km) is consistent with the source depths modelled from measured ground deformation. Data from other peralkaline volcanic centres in the Main Ethiopian Rift (Boset and Gedemsa), and other locations globally (e.g. Pantelleria, Italy) suggest that these storage conditions are a common feature of many peralkaline volcanic centres.

Key Words: Rhyolite-MELTS; Magma storage; Aluto; Main Ethiopian Rift

1 INTRODUCTION

At the northern end of the East African Rift system, the active Main Ethiopian Rift (MER) and the Afar Depression represent two distinct stages in the transition from a continental rift to a rift system that shares many characteristics with a slow-spreading oceanic ridge (Ebinger, 2005). Volcanic features in the MER include both focussed, caldera-forming silicic centres, and elongate rift-controlled basaltic fields, dominantly composed of monogenetic centres (Abebe et al., 2007; Corti, 2009). Volcanic activity is generally concentrated within localised segments, controlled by dense fault swarms (Corti, 2009). In the Afar region, incipient seafloor-spreading centres characterise the initiation of the Red Sea Rift, with extension largely accommodated by dikes (Wright et al., 2006; Keir et al., 2009), indicative of a more mature rift-setting.

In this study, we investigate the magmatic evolution of Aluto volcano, Ethiopia. Aluto is a peralkaline silicic volcano located in the MER, which has been active for ~ 500 ka and underwent a phase of major caldera-forming volcanism at ~ 310 ka (Hutchison et al., 2016a). Following a hiatus in activity after these caldera-forming events, volcanic activity at Aluto resumed at ~ 60 ka and has since been characterised by the eruption of evolved peralkaline rhyolites (Hutchison et al., 2015, 2016a,b,c). Aluto is one of a number of prominent restless caldera systems in the MER, showing evidence for

recent ground deformation (Biggs et al., 2011; Hutchison et al., 2016b), and hosts an active hydrothermal system (Hutchison et al., 2015). Volcanic hydrothermal systems offer geothermal resources that may provide significant economic benefits for Ethiopia (Kebede, 2012; Younger, 2014). Evaluating the potential risks associated with developing geothermal energy plants on these restless volcanic systems requires investigation of the magma storage conditions during the most recent eruptive periods. Improving our understanding of how and where magmas evolve beneath peralkaline volcanic centres is particularly important, given the potential hazards posed by the infrequent explosive eruptions at these volcanic systems. Aluto is currently showing signs of unrest (Biggs et al., 2011; Hutchison et al., 2016b), and recent geophysical observations allow comparison of the present magmatic activity with that inferred for past eruptive episodes.

Here we explore the hypothesis that the composition of Aluto's volcanic products can be generated by closed-system, isobaric fractional crystallisation (i.e. without crustal assimilation) and aim to determine the conditions of magma storage before recent eruptions. We approach this by calculating potential liquid lines of descent across a wide range of potential magmatic storage conditions (i.e. pressure, oxygen fugacity [f_{O_2}], initial H_2O content) using the Rhyolite-MELTS thermodynamic modelling software (Gualda et al., 2012) and comparing model outputs with whole-rock compositions of natural samples. We develop a statistical test to quantitatively assess the fit between modelled liquid lines of descent, and empirical whole rock data. On identifying the best fitting magma storage conditions (i.e. the model parameters that best predict the natural melt compositions), we compare these petrological constraints with the interpreted depth of melt accumulation from geophysical measurements of recent ground deformation and seismic activity at Aluto. A comparison between current activity and the petrologically derived magma storage conditions of past eruptions has not previously been attempted at Aluto and may provide important information regarding the potential scale and eruptive style of future activity.

1.1 GENESIS OF PERALKALINE MAGMAS

Peralkaline magmas are defined as having a peralkalinity index ($PI = (K_2O + Na_2O)/Al_2O_3$) greater than 1 (Macdonald, 1974), with peralkaline rhyolites further sub-divided into comendites (high Al_2O_3 and low FeO) and pantellerites (low Al_2O_3 and high FeO). Peralkaline magmas occur globally and are commonly associated with ocean islands (e.g. Socorro Island, Mexico; Bohrson and Reid, 1997), localised extension in collisional settings (e.g. Pantelleria; Civetta et al., 1988; White et al., 2009) and continental rifts (e.g. Gedemsa; Peccerillo et al., 2003). Isotopic and trace element analysis of peralkaline volcanic products suggests that fractional crystallisation is the dominant mechanism by which peralkaline rhyolites are generated (e.g. Peccerillo et al., 2003; Ronga et al., 2009; LeMasurier et al., 2011; Field et al., 2012; Neave et al., 2012; Rooney et al., 2012), though in some systems (e.g. Gedemsa and Olkaria) radiogenic isotope ratios point to a limited role for crustal contamination (e.g. Davies and Macdonald, 1987; Black et al., 1997; Peccerillo et al., 2003; Giordano et al., 2014).

Phase equilibrium experiments indicate that the generation of peralkaline magmas may be favoured by low pressure conditions, characteristic of the upper crust (Caricchi et al., 2006). Experimental data indicates that fractional crystallisation from a transitional basaltic parent cannot produce melt compositions that cross the subaluminous/peralkaline divide at $P > 0.5$ GPa. Phase equilibrium experiments by Di Carlo et al. (2010) suggest that pantellerite magmas may evolve at depths as shallow as 5 ± 1 km. Low pressures stabilise plagioclase feldspar and inhibit amphibole crystallisation, leading to an increase in the peralkalinity index of the residual melt (Caricchi et al., 2006). The phase relationships of peralkaline rhyolites are also consistent with crystallisation under relatively reduced conditions (<QFM – Quartz-Fayalite-Magnetite buffer), since the peralkalinity of the magma increases as crystallisation of calcic clinopyroxene is inhibited under these conditions (Scaillet and Macdonald 2001, 2003).

99 **2 GEOLOGICAL SETTING**

100 The MER is a ~500 km long zone of active continental rifting between the Nubian and Somalian plates
101 (Fig. 1; Corti, 2009), and is considered a type example of an active continental rift (Ebinger, 2005). It
102 can be split into three sections: the Northern, Central, and Southern MER. These developed
103 diachronously and display significant along-axis variation in structure and morphology (Keir et al.,
104 2015).

105 The initial Miocene-Pliocene rifting in the MER occurred due to displacement along large boundary
106 faults, resulting in the subsidence of asymmetric basins (Corti, 2009), with relatively diffuse, bimodal
107 volcanism (Abebe et al., 2005). At present, the deformation style changes along the MER as rift
108 maturity increases northward (Agostini et al., 2011); in the Southern MER, active extension is largely
109 controlled by displacement along boundary faults (Corti et al., 2013), whereas, during the Pleistocene,
110 the main boundary faults of the Central MER were deactivated and deformation is now localised on
111 dense fault swarms, along en-echelon segments within the thinned rift axis (Corti, 2009; Agostini et
112 al., 2011). These volcano-tectonic segments focussed the ascending magma and corresponding
113 volcanic centres (Ebinger and Casey, 2001). In contrast, the Northern MER is more closely analogous
114 to the Afar region where extension is achieved via dike emplacement, similar to oceanic spreading
115 centres (Ebinger et al, 2010; Ferguson et al., 2013).

116 **2.1 ALUTO**

117 Aluto volcano is located in the Central MER, around 100 km south of Addis Ababa. The complex
118 comprises a 700 m high, 14 km wide edifice predominantly composed of peralkaline pumice cones,
119 ignimbrites, lava flows and domes (Hutchison et al., 2015).

120 Exploration wells drilled at Aluto in the 1980s revealed the major structural features and underlying
121 volcanic stratigraphy (Gianelli and Teklemariam, 1993; Gizaw, 1993). For most of the Pleistocene,
122 volcanic activity was dominated by rift-related basaltic volcanism (Hutchison et al., 2016c). The early
123 Aluto edifice comprises a series of trachytic tuffs and lacustrine sediments that were emplaced before

deposition of a thick welded ignimbrite sequence at ~310 ka, which was likely accompanied by caldera collapse. The current (post-caldera) phase of volcanism started at ~60 ka and is characterised by rhyolitic pumice cones and obsidian coulees (Hutchison et al., 2016c). Aluto remains active, with current estimates indicating an eruption frequency of 1 every 1000 years in the last 10 ka (Hutchison et al., 2016c). Here we focus on the most recent (i.e. <60 ka) phase of volcanism at Aluto. These erupted products have a narrow compositional range, with nearly all analysed samples classified as pantellerites, according to the scheme of Macdonald (1974). Very few basaltic or intermediate-composition ejecta are known to have erupted within the past 60 ka, and these deposits are confined to a number of small vents located to the NW of the main volcanic complex. Recent vents are largely located along pre-existing faults, including a major NNE-SSW-trending fault zone (the Artu Jawe fault zone [AJFZ]) and a hypothesised ring-fault produced during caldera formation (Hutchison et al., 2015). Hutchison et al. (2016c) published major and trace element data for a large suite of Aluto samples and compiled whole-rock data from previous studies (Di Paola, 1972; Mamo, 1985; Teklemariam, 1996; Teklemariam et al., 1996; Yimer, 1984). These data are summarised in Table 1, and provide the basis for our geochemical analysis and thermodynamic modelling of magmatic evolution at Aluto. Trace element modelling by Hutchison et al. (2016c) suggested that the compositional diversity of Aluto pantellerites is consistent with fractional crystallisation of a rift-related basalt. However, they did not explore the major element liquid line of descent, phase equilibria constraints, mineral compositions or the physical conditions under which magmas may have been stored. Here, we build on the work of Hutchison et al. (2016c), first characterising the petrology of Aluto pantellerites, then using whole-rock and mineral analyses to investigate the plausibility of magma genesis through closed-system fractional crystallisation and to determine the conditions of magma storage.

3 PETROLOGY.

Thin sections of representative basaltic and pantelleritic samples were analysed using an FEI Quanta FEG-scanning electron microscope to characterise mineral textures and identify compositional zoning.

Phenocryst phase proportions in basaltic sample 17-01-05 were determined by mapping and subsequent phase analysis (Supplementary Material Table S.1 and Fig S.1) using an Oxford Instruments Energy-Dispersive X-ray Spectrometry (EDS) (Supplementary Material Table S.1 and Fig. S.1).

Major element compositions of olivine, alkali feldspar and plagioclase feldspar phenocrysts were analysed using a Cameca SXFive FEG-electron microprobe (EPMA) at the Department of Earth Sciences, University of Oxford, calibrated using a range of oxide and metal (Mn) micro-analytical standards. Matrix glass compositions in pantelleritic samples were determined using a JEOL 8600 EPMA at the Research Laboratory for Archaeology and the History of Art, University of Oxford.

Mineral analyses were collected using a 15 kV, 10 nA, 10 μm defocussed spot electron beam. All elements were counted on-peak for 30s with the exceptions of Na (12s) and Mn, P and Ti (60s). Matrix glass analyses were collected using a 15 kV, 6 nA, 10 μm defocussed spot electron beam, with SiO_2 and Na_2O analysed first to avoid potential alkali-loss during analysis (Humphreys et al., 2006). The count time on Mn was reduced to 50s on peak. For all elements in minerals and glasses, backgrounds were collected either side of the peak for half of the on-peak count time. Data quality was routinely monitored using secondary glass and/or mineral standards analysed at the start and end of each run. In general, the measured values of the secondary standards fall within 2σ of the published values (Jochum et al., 2006). A labradorite secondary standard (from the University of Bristol collection) was used before and after analysis of mineral phases, the composition of these standards are reported alongside the results from the EPMA analysis in Supplementary Material A.1; A.2; A.3. Since the mineral data collected by the Cameca SXFive EPMA are the first data to be collected by this particular instrument, repeat analysis of alkali feldspar or olivine phenocrysts were carried out on the JOEL 8600 EPMA. These repeat analyses confirm the reproducibility of the results gathered on the Cameca SXFive EPMA (see Supplementary Material A.4).

3.1 BASALTS

A representative sample of a Quaternary rift-related basalt (17-01-05) was collected from a scoria cone to the north-east of the main Aluto complex. It is porphyritic with the phenocryst assemblage: plagioclase feldspar + olivine + clinopyroxene + minor orthopyroxene. Magnetite is present in the microcrystalline groundmass. Olivine phenocrysts are typically 0.25-1 mm in diameter and are generally euhedral or subhedral. EDS mapping of an area of ca. 28 mm² of sample 17-01-05 and phase analysis was used to estimate the proportion of olivine phenocrysts in the sample. Olivine phenocrysts account for ~10% of the sample (Supplementary Table S.1) and are zoned, with narrow (~20-50 µm) Fe-rich rims (Fo₆₆₋₈₀) and more Mg-rich cores (Fo₈₀₋₈₇) (Supplementary Material A.3). EPMA analysis reveals that olivine cores have a uniform composition. The Mg-rich cores formed in a more primitive (Mg-rich) magma than the surrounding groundmass, which was compositionally similar to the whole-rock composition of sample 17-01-05 (Supplementary Material Fig. S.2).

Clinopyroxene and plagioclase phenocrysts in sample 17-01-05 are unzoned and subhedral. The sample has a subophitic texture, indicating that clinopyroxene grew contemporaneously or after plagioclase crystallisation (Fig. 2). A small number of large (>1 mm diameter) clinopyroxene and plagioclase phenocrysts have Fe or Na-rich cores overgrown by Mg or Ca-rich rims (<300 µm wide), respectively. However, these form only a minor component of the sample (<2%; see Supplementary Material).

3.2 PERALKALINE RHYOLITE

The pantellerite lavas are mildly (PI = 1.23) to strongly peralkaline (PI = 2.04) and are variably porphyritic (5-40% phenocrysts by volume), with the phase assemblage: alkali feldspar + quartz ± aenigmatite ± clinopyroxene ± apatite ± biotite ± Fe-Ti oxides (Fig. 3). All phenocryst phases are unzoned and lack disequilibrium textures (e.g. sieve textures). Alkali feldspar and quartz have a granophyric texture, attesting to their co-precipitation (Fig. 2). Alkali feldspar compositions cover a restricted compositional range within individual samples (e.g. Or₄₅ to Or₄₉), but vary from Or₃₀ to Or₄₉ between samples, crossing the divide between Anorthoclase and Sanidine (Supplementary Material

A.1). Alkali feldspar phenocrysts that are not intergrown with quartz crystals in a granophyric texture are typically euhedral to subhedral, and form elongate crystals (up to 3mm long). Little to no chemical zonation is apparent in the alkali feldspar phenocrysts. Matrix glass compositions (measured by EPMA; Supplementary Material A.2) are indistinguishable from the whole-rock composition of the pantellerites (see Supplementary Material).

3.3 THERMOMETRY

Temperature estimates were obtained from EPMA analysis of the rim compositions of alkali feldspar phenocrysts and matrix glass from 6 pantellerite lava samples (Supplementary Material A.1; A.2), using the alkali-feldspar – melt geothermometer of Putirka (2008; eqn 24b) assuming $P = 150$ MPa. These temperatures represent the crystallisation temperature of the alkali feldspar rims, and range from 718-765°C, with a standard error of $\pm 23^\circ\text{C}$ (Putirka, 2008). We assume that the alkali feldspar rims are in equilibrium with the surrounding matrix glass, consistent with the lack of disequilibrium textures or zoned rims. In no sample did the calculated temperature vary by more than 30°C , and most vary by $<15^\circ\text{C}$ (Supplementary Fig S.6). Rhyolite-MELTS models indicate that alkali-feldspar dominates the crystallising assemblage below $\sim 800^\circ\text{C}$ (see section 5). Despite the potential for Rhyolite-MELTS to overestimate crystallisation temperatures by $\sim 40^\circ\text{C}$ (Gualda et al., 2012; Gardner et al., 2014), the modelled alkali feldspar saturation temperature is consistent with that calculated by alkali-feldspar – melt geothermometry.

4 THERMODYNAMIC MODELLING

Thermodynamic modelling of the compositional evolution of liquid and mineral phases in Aluto magmas was carried out using Rhyolite-MELTS (Gualda et al., 2012). This model calculates the stable phase assemblage of a system under given conditions (e.g. P , T , $f\text{O}_2$) through free-energy minimisation. Rhyolite-MELTS is calibrated from experimental datasets for fluid-bearing silicic systems (Gualda et al., 2012). It been used successfully to investigate magma storage conditions in metaluminous and

peralkaline systems (e.g. Fowler et al. 2007; Fowler and Spera, 2010; Cannatelli, 2012; Pamukcu et al., 2015; Stock et al., 2016).

Here, we use Rhyolite-MELTS to investigate phase equilibria during isobaric crystallisation of primitive melts at Aluto. Pressure is held constant in each model run, and phase proportions and solid/liquid compositions are calculated as the temperature drops (starting at the liquidus), with crystal phases extracted from the bulk composition at each temperature step (i.e. fractional crystallisation). We ran simulations with 2°C temperature intervals because larger temperature intervals would influence the resolution of the fractional crystallisation models. Pressure, initial H₂O concentration (i.e. the H₂O concentration of the silicate liquid at the liquidus, equivalent to the H₂O concentration of the parental melt feeding the system) and fO_2 were varied systematically over a realistic range (50-300 MPa, 0.5-3 wt% and -2 – +1 log units below/above the QFM buffer, respectively), covering the range of potential magma storage conditions identified previously in peralkaline systems (Table 2). Model outputs are compared with previously determined whole-rock geochemical data to determine the parameters that represent the most likely magma storage conditions during pre-eruptive crystallisation at Aluto (section 4.2).

We used basalt sample 17-01-05 as a starting composition for these models. This represents the most primitive basalt sample from Aluto, as defined by its Mg number ($Mg\# = MgO / [FeO_t + MgO]$), and has been used as the starting composition for trace element modelling of Aluto magmas (Hutchison et al., 2016c). One hundred and forty four fractional crystallisation models were run, covering the full range of P, initial H₂O and fO_2 conditions.

We assume that whole-rock compositions of pantelleritic samples from Aluto are representative of melt (liquid) composition. Since the samples are generally phenocryst-poor, and measured matrix glass compositions are indistinguishable from whole rock in terms of their major element chemistry, this assumption is reasonable. Our statistical analysis (section 4.2) gives identical results regardless of whether the matrix glass or whole-rock compositions are used as the target composition (see

Supplementary Material for full discussion). The anhydrous component of the modelled compositions are normalised to 100% for comparison between empirical data and modelled compositions.

4.1 MODEL LIMITATIONS

Gualda et al. (2012) note that Rhyolite-MELTS does not accurately model the formation of hydrous phases such as amphibole. This influences the accuracy of Rhyolite-MELTS when modelling the evolution of intermediate magma compositions with high H₂O contents (Gualda et al., 2012). Biotite and apatite are the only hydrous minerals present in Aluto, and are volumetrically minor: biotite generally accounts for <2% of the phenocryst assemblage (Fig. 3), and apatite only occurs as an accessory phase (<1%; Fig.3). Consequently, we expect that inaccuracies in modelling the stability of hydrous minerals will have a negligible effect on our model outputs.

Rooney et al. (2012) note that Rhyolite-MELTS consistently over-predicts magmatic P₂O₅ concentrations, due to inaccuracies in the apatite solubility model. However, as apatite is only a minor accessory phase, this is also unlikely to have a significant effect on the major element evolution of the melt in our models. Fowler and Spera (2010) identified that Rhyolite-MELTS often over predicts melt CaO contents compared to measured compositions of natural samples. This is attributed to the under-stabilisation of clinopyroxene (Fowler and Spera, 2010), but does not seem to have a significant effect on the concentration of other major oxides in the melt.

Other potential limitations relate specifically to peralkaline systems. While multiple studies have used MELTS to investigate magmatic evolution in peralkaline volcanoes (Peccerillo et al., 2003; Ronga et al., 2009; Rooney et al., 2014), it is clear that some common peralkaline mineral phases (notably aenigmatite [(Na,Ca)₄(Fe²⁺,Ti,Mg)₁₂Si₁₂O₄₀]) are not accurately modelled. This is because there is insufficient experimental data on aenigmatite stability in peralkaline melts to accurately constrain its thermodynamic properties. Although aenigmatite is present in most obsidian samples from Aluto, it only occurs in minor modal quantities (Fig. 4), and is considered to be a late-crystallising phase, forming only at low temperatures, after a drop in melt Al₂O₃ following alkali feldspar precipitation (Di

Carlo et al., 2010; Neave et al., 2012). Consequently, inaccuracies in modelling aenigmatite should not influence the liquid line of descent until very late in fractional crystallisation, and thus do not significantly affect our results.

4.2 A STATISTICAL METHOD FOR IDENTIFICATION OF BEST FIT CONDITIONS IN RHYOLITE-MELTS MODELS

Previous studies have determined magmatic crystallisation conditions by comparing Rhyolite-MELTS outputs run at different conditions with empirical datasets (e.g. Fowler et al., 2007; Fowler and Spera, 2010; Cannatelli, 2012). Typically, the best-fit models are identified qualitatively by visual assessment. We have developed a statistical routine to quantify the fit between the modelled composition of the melt and the geochemical data. This approach is based on a least-squares analysis, where the sum of the squared residuals between the measured whole-rock data and modelled magma compositions is used to identify the model that most accurately reproduces the composition of natural samples. This method is explained in Supplementary Table S.3. The sum of the squared residuals is calculated from:

$$\sum_X^m \left(\sum_i^n (X_i - \hat{X})^2 / X_{SD} \right)_X \quad (\text{Eqn. 1})$$

where, n is the number of natural samples whose compositions represent the target compositions for the fractional crystallisation models (i.e. the compositions that we are attempting to reproduce via fractional crystallisation); m is the number of oxides considered in the analysis; X_i is the concentration of oxide X determined by whole-rock analysis of sample i ; \hat{X} is the concentration of oxide X predicted by Rhyolite-MELTS at a particular temperature; X_{SD} is the standard deviation of oxide X in all the empirically determined compositions of natural samples representing the target compositions. In all cases, elemental concentrations are in oxide wt%.

This analysis is carried out for every point in a single fractional crystallisation model (i.e. at every temperature step the composition generated by Rhyolite-MELTS is compared to the whole-rock data; Table S.3). The temperature point with the lowest calculated residual is then used to represent how closely that model matches the composition of the natural samples (Table S.3). We refer to this

method as the minimum Weighted Squared Residual (mWSR) method. However, taking only one point of a Rhyolite-MELTS model to represent the fit may give anomalous results (Fig. 4a). We therefore developed a second method, which averages the 15 lowest residuals for each model (referred to as the average Weighted Squared Residual method – aWSR). Using greater than 15 analyses tends to give erroneous results because some of our models terminate at a melt fraction of $f \sim 0.1$ (after which Rhyolite-MELTS failed to converge), whereas others were able to progress to $f < 0.05$ (explained in Fig. 4b). A MATLAB routine implementing this analysis is available in the Supplementary Files.

The aWSR method provides a means to quickly and easily evaluate intrinsic variables of magma storage, and determine the models that most likely represent the conditions of natural magma storage. The strength of this method is not only that it is a statistically robust, quantitative determination of magma storage conditions from modelling data, but also the speed and ease of analysis. In previous studies (e.g. Fowler et al., 2007; Fowler and Spera, 2010; Cannatelli, 2012), mineral abundances and compositions predicted by Rhyolite-MELTS are also compared with those observed in natural samples. These factors are not included in the statistical routine described above and are difficult to apply to Aluto, due to low crystallinity and the formation of certain phenocryst phases (e.g. aenigmatite) which are not accurately modelled by Rhyolite-MELTS. Comparison of modelled and measured feldspar compositions was attempted but this was made difficult by the over prediction of CaO in the melt, which caused Rhyolite-MELTS models to over predict the CaO content of the alkali feldspars (see Supplementary Information).

5 RESULTS FROM RHYOLITE-MELTS MODELLING

5.1 STARTING COMPOSITION

Using basalt sample 17-01-05 as a parental magma composition we are able to successfully recreate most aspects of the major element composition of pantelleritic samples from Aluto. Hutchison et al. (2016c) were able to successfully model trace element variation during magmatic evolution at Aluto using the same sample, and we suggest that the whole-rock composition of sample 17-01-05 is the

best estimate of the Aluto parental magma. Future work to determine the composition of primitive mafic samples in the region would greatly aid future investigations of magmatic differentiation at peralkaline volcanic centres in the MER.

5.2 LIQUID LINE OF DESCENT

All models using 17-01-05 as a starting composition display comparable geochemical trends as minerals are predicted to crystallise in a similar order and at similar temperatures (Fig. 5), although it is important to note that Rhyolite-MELTS is known to overestimate saturation temperatures, as mentioned previously (Gualda et al., 2012; Gardner et al., 2014). Olivine is the first mineral predicted to precipitate at the liquidus (1260-1213°C), causing a large decrease in the MgO content and a smaller increase in the SiO₂ concentration of the residual liquid over a short crystallisation interval (Fig. 5e). Clinopyroxene is the next phase to appear on the liquidus (1185°C at fO_2 = QFM; P = 150 MPa; initial H₂O = 0.5 wt%; Fig. 6b), at which point melt SiO₂ concentrations begins to decrease along with CaO. Melt MgO content continues to decrease markedly over a short crystallisation interval and the concentrations of other key oxides (Na₂O, K₂O, Al₂O₃ and TiO₂) increase slightly.

Spinel group minerals (predominantly ulvospinel – magnetite solid solution) and plagioclase are the next minerals that are predicted to precipitate, but the order in which they come onto the liquidus depends on the magma storage conditions. Plagioclase will generally precipitate first, but at high fO_2 (QFM+1) and high initial H₂O contents (≥ 1 wt%) spinel precipitates first. At fO_2 = QFM; P = 150 MPa; initial H₂O = 0.5 wt%, plagioclase precipitates at 1159°C and the spinel group minerals precipitate at 1117°C (Fig. 6b). However, at fO_2 = QFM+1; P = 150 MPa; initial H₂O = 1 wt%, plagioclase precipitates at 1116°C, after spinel group minerals (predominantly magnetite) at 1134°C. Crystallisation of these minerals has a major influence on the composition of the residual liquid. Notably, when spinel group minerals start to precipitate, melt SiO₂ concentrations increase significantly over a short crystallisation interval, while TiO₂ concentrations decrease. Models predict that plagioclase feldspar is initially Ca-

rich ($An_{80}-An_{90}$), causing a continual decrease in melt CaO content, and Al_2O_3 . After *plagioclase*- and *spinel-in*, there is a reduction in the magnitude of the decrease in MgO per unit crystallisation.

As fractional crystallisation continues, plagioclase, olivine, clinopyroxene and spinel group minerals dominate the mineral assemblage, leading to a continual decrease in MgO, FeO_t , Al_2O_3 and TiO_2 and an increase in the Na_2O , K_2O and SiO_2 contents of the residual liquid over a protracted temperature interval (i.e. 1150-900°C). The SiO_2 content of the residual liquid can be used as a fractionation index to measure the degree of differentiation (higher SiO_2 represents more differentiated melts).

As the residual liquid evolves towards more SiO_2 -rich compositions, modelled feldspar compositions become progressively more sodic. Consequently, the degree of Na_2O enrichment in the residual liquid per unit crystallisation decreases during magmatic evolution. Rhyolite-MELTS predicts sanidine crystallisation after ~90% crystallisation (Fig. 6). At this point, K_2O becomes compatible with the crystallising mineral assemblage, and rapidly becomes depleted in the residual liquid (Fig. 5b). Modelled sanidine compositions are more calcic than those measured in natural samples. This is likely due to known inaccuracies in the modelling of magmatic CaO evolution in Rhyolite-MELTS (Section 4.1; Fowler and Spera, 2010).

Melt FeO_t concentrations predicted in Rhyolite-MELTS models at high SiO_2 (>70 wt%) do not match those observed in Aluto pantellerites (Fig 5h). This may be due to inaccuracies in modelling of aenigmatite stability in pantelleritic melts; Rhyolite-MELTS predicts crystallisation of Fayalite and Fe-Ti oxides instead of aenigmatite during magmatic differentiation. These minerals contain more Fe than aenigmatite, resulting in lower FeO_t values. As stated previously, aenigmatite is only likely to form very late during fractional crystallisation (following alkali feldspar formation when the magma becomes enriched in Na_2O relative to Al_2O_3 ; Di Carlo et al., 2010; Neave et al., 2012) and is a relatively minor modal component. Therefore, inaccuracies in aenigmatite modelling will only have an important influence on the concentration of the major elements at very low residual melt fractions ($f < 0.05$).

5.3 EFFECTS OF OXYGEN FUGACITY

The stability of spinel group minerals in Rhyolite-MELTS models is affected by changes in the system fO_2 . At higher fO_2 , the stability of spinel minerals (specifically magnetite) increases in the melt, and they precipitate at higher temperatures. Consequently, the increase in magmatic SiO_2 concentration that accompanies *spinel-in* begins at higher temperatures. Enhanced magnetite and ulvospinel crystallisation at elevated fO_2 , causes lower Na_2O and higher MgO concentrations in the residual melt at a given SiO_2 concentration (Fig. 7). At low fO_2 (QFM-2), *plagioclase-in* can occur up to 80°C before crystallisation of spinel group minerals, causing a reduction in the Al_2O_3 content of the magma during the early stages of differentiation. At higher fO_2 , plagioclase forms alongside or after spinel group minerals. As a result, the Al_2O_3 concentration of the residual melt does not decrease prior to an increase in the SiO_2 concentration caused by magnetite and ulvospinel fractionation, resulting in a higher Al_2O_3 concentration at similar SiO_2 content in models at higher fO_2 .

Earlier formation of spinel group minerals explains the smaller increase of TiO_2 during magmatic cooling in models at $fO_2 = QFM+1$, relative to models at $fO_2 = QFM-2 - QFM$. Variability in fO_2 also influences the composition of spinel group minerals. At $fO_2 = QFM+1$, spinel minerals contain less TiO_2 than at lower fO_2 . Therefore, at $fO_2 = QFM-2 - QFM$ there is greater reduction of TiO_2 after *spinel-in*.

Overall, models at $fO_2 = QFM - QFM+1$ provide a better match to the geochemical data of eruptive products from Aluto than those at $fO_2 < QFM$. Models at low fO_2 (QFM-2 - QFM-1) under predict the Al_2O_3 and TiO_2 contents of the pantelleritic rhyolites at Aluto, and over predict the K_2O and Na_2O contents. This is confirmed by very high residuals for models at $fO_2 < QFM$ in our aWSR statistical analysis (Fig. 8a). This analysis also reveals that models at $fO_2 = QFM$ provide a better match to the compositional data than those at $fO_2 = QFM+1$.

5.4 PRESSURE

At pressures >200 MPa, leucite is predicted as a stable mineral phase in Rhyolite-MELTS models. As leucite is not present in any Aluto samples, isobaric fractional crystallisation at >200 MPa is considered

implausible. At lower pressures, variability in the modelled liquid line of descent is largely the result of variable feldspar stability. Plagioclase stability is inversely correlated with pressure, causing an increase in Al_2O_3 and a decrease in the MgO content of the melt at a given SiO_2 concentration in higher pressure models (Fig. 9). Conversely, as pressure increases the stability of sanidine increases, in agreement with previous work (Blundy and Cashman, 2001). Earlier sanidine crystallisation at elevated pressures causes K_2O and Al_2O_3 to decrease in the melt at higher temperatures (and SiO_2 concentrations) as crystallisation progresses. At pressures of ~200 MPa, Rhyolite-MELTS predicts formation of an Al- and Ti-rich clinopyroxene, causing a decrease in the MgO, Al_2O_3 and TiO_2 contents of the residual liquid. Models at lower pressures (50 MPa) slightly over predict the MgO and under predict the K_2O content of the natural pantelleritic samples. Therefore, the composition of the natural samples from Aluto appear to be best reproduced by models at 100 - 150 MPa. Using our statistical analysis, we are able to quantitatively show that the best fitting pressure lies within the range 100 - 150 MPa, and is dependent on the initial H_2O content of the parental melt (Fig. 8b).

5.5 INITIAL WATER CONTENT

Higher initial H_2O contents reduce plagioclase stability, resulting in magmas with higher Al_2O_3 and lower MgO contents than those with lower initial H_2O at equivalent SiO_2 concentrations. Furthermore, higher initial H_2O content (>1 wt%) induces crystallisation of a second high Ti-clinopyroxene phase, in addition to the relatively Ti-poor clinopyroxene that is seen in all other models. This causes a decrease in the TiO_2 content of the melt at a given temperature, relative to models with lower initial H_2O concentrations. As a result, models with lower initial H_2O contents (0.5 wt%) best reproduce the composition of the basaltic and intermediate samples from Aluto (Fig 10).

In models with initial H_2O contents ≥ 1 wt%, the Na_2O content of the melt is under predicted at pressures >100 MPa, due to increased stability of more albitic plagioclase. However, when $f\text{O}_2 = \text{QFM}$ and $P = 100$ MPa, the composition of the pantelleritic rhyolites from Aluto can be reproduced

reasonably well, regardless of the initial H₂O content. At low initial H₂O contents (0.5 wt%), changing the crystallisation pressure only has a minor effect on the liquid line of descent.

There are a number of initial conditions which appear to provide a reasonably good match to the compositional data from Aluto. However, using our statistical analysis, we are able to determine that the best fitting storage conditions are at low initial H₂O content (0.5 wt%) and pressures of 150 MPa (Fig. 8c). In summary, the aWSR analysis verifies that the Rhyolite-MELTS models that best reproduce the natural composition of Aluto whole-rock samples were run at $fO_2 = \text{QFM}$; $P = 150 \text{ MPa}$; initial H₂O = 0.5 wt%.

6 DISCUSSION

6.1 MAGMA STORAGE CONDITIONS

Rhyolite-MELTS modelling of major elements shows that the composition of the pantellerite lavas from Aluto can be generated by protracted fractional crystallisation without the need for crustal assimilation. Around 90% fractional crystallisation is required to produce these compositions, in agreement with previous trace element investigations (Hutchison et al., 2016c). This represents a similar degree of fractional crystallisation to that required for rhyolite genesis at other peralkaline volcanic centres (Peccerillo et al., 2003; White et al., 2009).

Our aWSR analysis indicates that Aluto magmas crystallise with fO_2 close to the QFM buffer (Fig. 8a). This is supported by the presence of Ti-bearing aenigmatite, which is stable at fO_2 between the QFM and Nickel-Nickel Oxide buffers and a pressure of ~100 MPa (Scaillet and Macdonald, 2001; Ronga et al., 2009). Crystallisation at $fO_2 = \text{QFM}$ is consistent with other peralkaline systems (White et al., 2005; Macdonald, 2012).

The statistical results show that models with initially low H₂O contents (0.5 wt%) generally have a better match to the pantelleritic rhyolites from Aluto than models run with higher starting H₂O contents (Fig 8c). Low initial H₂O contents (0.5 wt%) produce a better match to the basaltic and

intermediate products at Aluto (Fig. 10). These low H₂O contents in Aluto parental magmas are consistent with results from previous studies on peralkaline volcanism in the MER and elsewhere (section 6.3). For example, melt inclusion analysis in olivine and clinopyroxene hosts from the Cuddie Rosse scoria deposits, Pantelleria, indicate dissolved H₂O contents of 0.9-1.6 wt% at an MgO content of ~5 wt% (Gioncada and Landi, 2010). Our models predict that when the residual liquid has reached 5 wt% MgO, the dissolved volatile content is ~0.85 wt%, only marginally lower than those measured at Pantelleria. Low initial H₂O contents increase plagioclase stability in the magma, resulting in a greater reduction of Al₂O₃ due to plagioclase crystallisation and increasing the peralkalinity of the residual melt.

The predicted low initial H₂O content of Aluto melts implies that saturation of a H₂O-rich volatile phase is only expected to occur at low pressures or after significant crystallisation (~90-95% crystallisation, similar to the amount of crystallisation required to produce the composition of pantellerites from Aluto; Fig. 6b). This may play an important role in priming the system for eruption (e.g. Stock et al., 2016); late-stage exsolution of a magmatic vapour phase would increase the overpressure of the magma chamber (Tait et al., 1989; Fowler and Spera, 2008) and the buoyancy of the magma (Degruyter and Huber, 2014). Hence, saturation of a H₂O-rich volatile phase may increase the eruptibility of highly silicic pantelleritic magmas by driving a rapid rise in the volume fraction of bubbles in the magma (Fowler and Spera, 2008; Stock et al., 2016). Whilst many peralkaline systems have been shown to be volatile saturated with respect to a Cl-rich brine well before eruption (e.g. Lanzo et al., 2013) further work on melt inclusion studies is required to determine whether or not this is the case for Aluto.

At low initial H₂O contents (0.5 wt%), pressure variations only play a minor role in determining the liquid line of descent. However, our statistical analysis shows that the best match to the compositional data is at ~150 MPa (section 5.6). These relatively low pressures are supported by the presence of granophyric textures in rhyolitic magmas (Fig. 2), which are indicative of crystallisation at shallow

depths (~3 km) where undercooling can occur due to volatile exsolution during magma ascent (Lowenstern et al., 1997). Geobarometry using the method of Gualda and Ghiorso (2014) was attempted for the matrix glass compositions in this study. We found that this method was not applicable here, as the liquid needs to be saturated in both quartz and feldspar, which is not the case for these matrix glass compositions. We find that these compositions are under saturated with respect to quartz, which may explain why quartz is predominantly found as in granophyric intergrowths that formed as a result of undercooling (Lowenstern et al., 1997).

Mineral-melt equilibria using rim compositions from the alkali feldspar phenocrysts provides magmatic temperature estimates shortly before eruption. In Aluto, these mineral-melt equilibria reveal a relatively narrow temperature range between 718-765°C (section 3.3). This is a smaller eruptive temperature range than that observed in peralkaline rhyolites erupted from other peralkaline silicic centres (e.g. Pantelleria, White et al., 2009; Dabbahu, Field et al., 2012). Whilst this may simply reflect a sampling bias (temperature was calculated for only 6 samples at Aluto compared to 27 at Dabbahu; Field et al., 2013), this potentially reflects the restricted compositional diversity of Aluto products from the last ~60 ka, relative to the compositional range of Dabbahu and Pantelleria eruptive products in the same age range (White et al., 2005; 2009; Field et al. 2012; 2013). Additionally, Dabbahu volcano displays several key differences to Aluto, for example, magma mixing has been shown to be an important process at Dabbahu, and magma is considered to be stored in a series of stacked sills, rather than a compositionally zoned magma reservoir (Field et al., 2012; 2013).

Independent experimental studies on peralkaline magmas have shown that the phase assemblage of alkali feldspar + clinopyroxene + aenigmatite + ilmenite is stable at 100-150 MPa at near H₂O saturated conditions (Di Carlo et al., 2010) and temperatures of 680-725°C. However, it is argued that the low crystal content and high proportion of alkali feldspar phenocrysts in natural samples from Pantelleria (and Aluto) indicate that eruption temperatures are at the top of the range suggested by Di Carlo et al. (2010). These experiments agree with our modelling results, although the temperature range is

slightly below that estimated for our pantellerite samples, based on alkali feldspar – melt equilibria. This may be due to slight differences in the composition of the samples, and that our temperatures represent the temperature of crystallisation rather than temperature of eruption. As alkali feldspar is shown to form before aenigmatite and quartz (Di Carlo et al., 2010), this may also explain the slight temperature discrepancy.

The stratigraphy beneath Aluto is well-constrained (see Hutchison et al., 2015), and we infer an average crustal density of $\sim 2625 \text{ kg/m}^3$ up to 2.5 km beneath the volcano ($\sim 65 \text{ MPa}$). At greater depths, regional gravity surveys reveal that the crustal density beneath MER volcanoes (i.e. Boset) ranges from $2770\text{--}3000 \text{ kg/m}^3$ (Cornwell et al., 2006). Assuming an average crustal density of 2800 kg/m^3 at depths $>2.5 \text{ km}$, the depth of magma storage at Aluto (i.e. crystallisation at 150 MPa) is $\sim 5.6 \pm 1 \text{ km}$ (where errors represent uncertainty in the crustal density).

The extent of crystallisation ($\sim 90\%$) required to generate the rhyolitic melt compositions erupted at Aluto indicates that much of the magma reservoir beneath the volcano likely exists as a highly crystalline magmatic mush. It is extremely difficult to extract melt from the interstices of a magma reservoir with a crystallinity as high as 90% (Dufek and Bachmann, 2010). However, we argue that differentiation of silicic melts beneath Aluto may be the result of crystal settling and *in-situ* crystallisation along the crystal mush-melt interface, as suggested for other peralkaline-to-agpaitic magmas (Hunt et al., 2017). The low viscosity of peralkaline magmas would aid magmatic differentiation via crystal settling (Neave et al., 2012). Crystal-poor silicic magmas erupted at Aluto may therefore be extracted from a melt-rich lens or cupola near the roof of the magma reservoir, as envisaged for other rhyolitic volcanoes (Bachmann and Bergantz, 2008), rather than from melts distributed between crystals throughout the mush pile. This is supported by variations in the incompatible trace element compositions of melts erupted at Aluto since $\sim 60 \text{ ka}$, which are consistent with crystal fractionation in the compositionally zoned cap of a shallow magmatic reservoir (Hutchison et al., 2016c). Additionally, magnetotelluric data is inconsistent with the presence of a large,

dominantly liquid magma body at the crystallisation depth indicated by our Rhyolite-MELTS models ($\sim 5.6 \pm 1$ km; Samrock et al., 2015).

Previous studies have suggested the importance of polybaric crystallisation during magmatic differentiation in MER peralkaline volcanoes (Rooney et al., 2012; 2014). Our models demonstrate that complex magma plumbing systems, with crystallisation at different crustal levels, are not necessary to generate the compositional diversity of Aluto samples, which is consistent with simple isobaric differentiation. The lack of petrographic or geochemical evidence for magma mixing in pantellerite samples from Aluto is in contrast with other peralkaline volcanic centres, which show evidence for interaction between rhyolites and more mafic magmas (e.g. Boset; Ronga et al., 2009 and Dabbahu; Field et al., 2012). This may indicate more complex magmatic plumbing systems beneath these volcanoes (i.e. multiple regions of magma storage). However, petrological evidence does not require or support significant polybaric crystallisation at Aluto.

6.2 COMPARISON WITH RECENT ACTIVITY

Using a Mogi point source model Hutchison et al. (2016b) observed that a period of ground deformation in 2008 (i.e. inflation of ~ 10 cm) was best modelled by a deformation source at $\sim 5.1 \pm 0.5$ km depth (Hutchison et al., 2016b). $\delta^{13}\text{C}$ analysis of CO_2 in soil gas and fumaroles from Aluto overlap magmatic values and provide evidence for a degassing magmatic body at depth (Hutchison et al., 2016b). The 5.1 km deformation source is thought to represent the roof of a magmatic storage region (Hutchinson et al., 2016b), consistent with our petrological modelling results (Fig. 11).

Recent measurements of seismicity at Aluto also point to the presence of a shallow storage region. Data from a local seismic network deployed around Aluto from 2012-2014 reveals a decrease in the number of seismic events recorded from 4 – 7 km depth directly beneath the Aluto caldera (Wilks, 2016). This depth range corresponds with our predicted depth of magma storage, and the decrease in seismicity in this region may be related to elevated temperatures close to a magma body (Fig. 11).

6.3 COMPARISON TO OTHER MER VOLCANIC CENTRES

Boset and Gedemsa, two peralkaline silicic centres in the Northern MER, have been the subject of previous petrological and geochemical studies (Peccerillo et al., 2003; Ronga et al., 2009). These volcanoes have phase assemblages and peralkaline magmatic compositions that are similar to those seen at Aluto, and MELTS modelling has been used to evaluate magmatic storage conditions at these locations (Peccerillo et al., 2003; Ronga et al., 2009). These studies indicate that storage conditions at Boset and Gedemsa are similar to those identified at Aluto ($P = 50\text{--}100$ MPa; $fO_2 = \text{QFM}$; initial $H_2O = 1$ wt%). We have compiled data on magmatic storage conditions from a number of other peralkaline volcanoes in the MER and elsewhere in Table 4. Although slight differences in the magmatic storage conditions can be identified between the different systems, this compilation indicates that peralkaline magmas tend to be generated by magmatic differentiation in the shallow crust (commonly <200 MPa) from parental melts with relatively low initial H_2O contents (typically ≤ 1 wt% at $MgO = 5\text{--}10$ wt%) and at an oxygen fugacity close to the QFM buffer, as hypothesised previously (Peccerillo et al., 2003; Caricchi et al., 2006; White et al., 2009).

Regional gravity surveys in the MER reveal the presence of positive gravity anomalies within the shallow crust (Mahatsente et al., 1999; Cornwell et al., 2006). These are interpreted to represent crystallised magmatic intrusions, which often occur at depths of <4 km (Mahatsente et al., 1999). These separate lines of evidence indicate that shallow magma storage systems are likely to be a common feature of many peralkaline silicic centres along the MER and suggest that silicic volcanism at MER volcanic centres is commonly fed by mafic intrusion and subsequent crystal fractionation in the shallow crust.

6.4 FORMATION OF A DALY GAP

At Aluto there is a notable absence of erupted samples with intermediate magma compositions, which are predicted to form during protracted fractional crystallisation from basalts to evolved pantellerites. This lack of intermediate products (i.e. a “Daly Gap”; Daly, 1925) is observed on a regional scale across the MER, and may relate to density stratification of magma reservoirs (Peccerillo et al., 2003; Ronga

et al., 2009; Rooney et al., 2012); low-density silicic magmas occupy the shallower parts of magma reservoirs and inhibit the eruption of higher density intermediate and basaltic compositions from deeper parts of the system at the central volcanic centre (Peccerillo et al., 2003; Neave et al., 2012). This scenario may apply at Aluto if pantelleritic eruptions are sourced from the top of a compositionally zoned chamber that lies directly beneath the caldera and could explain why eruption of basaltic and intermediate composition lavas are restricted to a small number of volcanic cones to the NW of the main complex (Hutchison et al., 2016c). At Aluto density stratification likely involves a rhyolitic liquid cap underlain by basaltic and/or intermediate composition mush. The highly crystalline mush is unlikely to be erupted due to its high viscosity, and there is no evidence for magma mixing and mush rejuvenation at Aluto (e.g. Burgisser and Bergantz, 2011).

Our thermodynamic modelling of Aluto magmas shows that SiO_2 does not change linearly with decreasing temperature. SiO_2 increases from 50 to ~64 wt% over a very short crystallisation interval, mainly due to the crystallisation of SiO_2 -poor phases such as spinel (White et al., 2009); this causes the system to evolve through intermediate compositions over a short temperature interval (Fig. 12) and has been proposed previously as a mechanism for generating a Daly Gap (Mushkin et al. 2002; White et al., 2009). As magma storage conditions for peralkaline systems tend to be very similar (section 6.3), we may expect the crystallising assemblage to be similar for many peralkaline systems. Therefore, it may not be surprising that many peralkaline volcanoes lack intermediate composition magmas (Macdonald, 2012). The occurrence of a Daly Gap at Aluto is likely due to a combination of multiple factors, including density stratification of a zoned magma reservoir beneath the caldera and the scarcity of intermediate composition magmas produced by the fractionation process.

7 CONCLUSIONS

1. Magma storage conditions can be evaluated using Rhyolite-MELTS to determine the simplest scenario (i.e. the scenario with the least number of assumptions) under which magmatic differentiation can produce the compositions of the eruptive products. We have developed a

quantitative statistical routine that allows identification of the model input parameters (i.e. the magma chamber conditions) that most closely predicts the compositions of natural erupted products. This approach could be easily applied in other volcanic systems to identify best fitting storage conditions.

2. We have constrained the pre-eruptive magma storage conditions of peralkaline rhyolites at Aluto by comparing liquid lines of descent modelled at different pressures, initial H₂O concentrations and fO_2 (using Rhyolite-MELTS) with the compositions of natural eruptive products. Our results suggest that erupted melts at Aluto formed through fractional crystallisation of a parental basaltic magma with a low initial H₂O content (0.5 wt%), an fO_2 near the QFM buffer, and at depths of $\sim 5.6 \pm 1$ km (150 MPa). These results are consistent with pantelleritic phase equilibria experiments, and textural evidence (e.g. granophyric texture), and are in agreement with the depth of the current melt zone beneath Aluto interpreted from InSAR observations and seismic data.
3. Modelling isobaric fractional crystallisation using Rhyolite-MELTS reveals that Aluto pantellerite magmas can be generated by $\sim 90\%$ fractional crystallisation of a parental melt represented by basaltic sample 17-01-05 (a rift-related basalt collected to the east of the main Aluto complex; Table 1). These models reveal that assimilation of crustal material is not required to produce the composition of the pantellerite samples at Aluto.
4. The formation of a Daly Gap at Aluto is likely due to a combination of factors, including density stratification and compositional zonation within the magma reservoir beneath the main caldera, and the short crystallisation interval over which SiO₂ increases from 50 to ~ 64 wt%.
5. In comparison with experimental data and observations from other volcanic systems along the MER (e.g. Boset and Gedemsa) and in other tectonic settings (e.g. Pantelleria), our results indicate that magma storage in the upper crust, low parental melt H₂O contents (≤ 1 wt%) and fO_2 near the QFM buffer are characteristic features of many peralkaline systems globally.

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856

857 **FIGURE CAPTIONS**

858 **Figure 1** – Topographic map of the East African Rift system. The location of the Main Ethiopian Rift and Afar
 859 depression are identified by the black lines. Red dashed lines indicate the divisions between the South Main
 860 Ethiopian Rift (SMER), Central Main Ethiopian Rift (CMER), North Main Ethiopian Rift (NMER) and the Afar region
 861 (after Corti., 2009). Volcanic centres are represented by the small grey triangles (data from the Smithsonian
 862 Global Volcanism Program; Siebert and Simkin, 2002). The location of Aluto is marked by the larger red triangle.
 863 Inset is taken from GeoMapApp (Ryan et al., 2009; <http://www.geomapapp.org/>).

864 **Figure 2** – Photomicrographs (**a, b, d, e**) and electron backscatter images (**c, f**) of basaltic and pantelleritic
 865 samples from Aluto showing key textural features. (**a-c**) Images from basaltic sample 17-01-05. (**a**) Subophitic
 866 texture indicating that plagioclase grew before or contemporaneously with clinopyroxene phenocrysts. (**b**) Large
 867 reversely zoned (confirmed by SEM) clinopyroxene phenocryst. Zoned clinopyroxene phenocrysts are
 868 volumetrically minor (only two crystals identified in the sample). (**c**) Olivine phenocrysts with Fe-rich rims (lighter
 869 grey colour in the back scatter image). (**d, e**) Granophyric texture in sample 01-02-14, with an alkali feldspar core.
 870 (**f**) Aenigmatite phenocryst in sample 30-01-10. Scale bars in all images are 500 μm , except **f** where the scale bar
 871 is 100 μm .

872 **Figure 3** – Modal proportions of phenocryst phases in pantelleritic lavas from Aluto determined by point counting
 873 using JMicroVision 1.2.7 software. The dominant crystal phases are alkali feldspar and quartz. Apatite is also
 874 present in some samples but is an accessory phase, typically comprising <1% of the phenocryst population.
 875 Sample names and whole-rock compositions are found in Table 1.

876 **Figure 4** – Graphs showing the advantage of average Weighted Squared Residual (aWSR) analysis over minimum
 877 Weighted Squared Residual (mWSR) analysis (**a**), and why using >15 points in the aWSR analysis may give
 878 erroneous results (**b**). Black lines show hypothetical modelled liquid lines of descent (e.g. Rhyolite-MELTS
 879 outputs), with starting compositions represented by the black points and the 'target' compositions (i.e. the
 880 composition of the natural samples) marked by the green area. (**a**) For the two modelled liquid lines of descent,
 881 the points recorded by mWSR analysis are represented by the red dots and the area measured by aWSR is shown
 882 by the blue section of the line. Using mWSR analysis, Model 2 more closely reproduces the 'target' composition.
 883 However, when the entire 'target' area is considered by the aWSR analysis, it is clear that Model 1 provides the
 884 better match to the target region. (**b**) At low melt fractions ($f < 0.1$) some Rhyolite-MELTS models fail to converge.
 885 This leads to a bias in the aWSR analysis, as models that are able to progress to low melt fractions are more likely
 886 to pass through the entire 'target' region (e.g. Model 1). To limit this effect, aWSR analysis is confined to the
 887 minimum 15 data points (i.e. Rhyolite-MELTS temperature steps) for each model. Additionally, for Aluto, 15 data
 888 point corresponds to a temperature variation of $\sim 30^\circ\text{C}$, similar to the range of eruption temperatures calculated
 889 in all but one sample from Aluto. Therefore, measuring the fit over a larger temperature interval is not justified.

890 **Figure 5** – Comparison of Rhyolite-MELTS model outputs and empirical data from Aluto. The black line shows the
 891 modelled compositional evolution of the silicate melt during fractional crystallisation under the best fit conditions
 892 according to the aWSR analysis ($P = 150 \text{ MPa}$, $f\text{O}_2 = \text{QFM}$, initial $\text{H}_2\text{O} = 0.5 \text{ wt\%}$). Data points show the
 893 compositions of Aluto whole rock samples from Hutchison et al. (2016c) and are distinguished by composition
 894 and the timing of eruption (see legend). The points at which olivine (Ol); Clinopyroxene (Cpx); Plagioclase (Plag);
 895 Spinel (Spl); Apatite (Apa); Rhombohedral-Oxide (Rhm-oxide); and Sanidine (San) come onto the liquidus are
 896 highlighted. Rhyolite-MELTS does not accurately reproduce the FeO , P_2O_5 or CaO content of the pantelleritic
 897 samples. The dashed lines in (**e**) show 20% crystallisation intervals. Errors for whole-rock analysis are typically
 898 smaller than the size of the data points and are reported in the caption of Table 1.

899 **Figure 6** – Mineral phase proportions versus temperature, as predicted by Rhyolite-MELTS models during
 900 fractional crystallisation at 0.5 wt% (**a,b**) and 1 wt% (**c,d**) initial H_2O , and at 100 MPa (**a,c**) and 150 MPa (**b,d**). In
 901 all models $f\text{O}_2$ is set at the QFM buffer. These results are for the storage conditions that produce the lowest
 902 residuals in the aWSR analysis. The model in (**b**) represents the best fit conditions, as determined by the aWSR
 903 analysis. Vertical dashed black line represents the temperature of H_2O saturation in each model.

Figure 7 – Liquid lines of descent (black lines) produced by Rhyolite-MELTS under different fO_2 conditions but identical pressure and initial H_2O concentrations (see legend). Data points show the compositions of Aluto whole rock samples from Hutchison et al. (2016c) and are distinguished by composition and the timing of eruption (see legend). The best match to the measured whole-rock compositions from Aluto are at $fO_2 = QFM - QFM+1$. Labels in (c) show where plagioclase and spinel come onto the liquidus in the $fO_2 = QFM-2$ model. Labels in (d) highlight spinel-in in the $fO_2 = QFM$ and $QFM+1$ models, and rhm-oxide-in in the $fO_2 = QFM+1$ model. Labels in (e) show spinel-in for all models, apart from those at $fO_2 = QFM-2$. This modelling demonstrates that spinel saturation occurs later during fractional crystallisation (i.e. at lower temperatures) at lower fO_2 .

Figure 8 – aWSR results showing the effect of varying fO_2 (a), pressure (b) and initial H_2O concentration (c) in Rhyolite-MELTS on the correlation between modelled liquid compositions and the compositions of natural pantellerites from Aluto. Lower residuals indicate a better match between model outputs and natural sample compositions. These results show that Aluto magmas are stored at an fO_2 near the QFM buffer and pressures between 100 and 150 MPa. The lowest residuals are produced by models at 150 MPa and low initial H_2O concentrations (0.5 wt%).

Figure 9 – Liquid lines of descent (black lines) produced by Rhyolite-MELTS at different pressures but identical fO_2 and initial H_2O concentrations (see legend). Data points show the compositions of Aluto whole rock samples from Hutchison et al. (2016c) and are distinguished by composition and the timing of eruption (see legend). At low initial H_2O concentrations (0.5 wt%), the effect of pressure on the liquid line of descent is minor. Labels in (b) show that sanidine-in occurs at lower SiO_2 concentrations (i.e. higher temperatures) as pressure increases. Labels in (c) highlight plagioclase-in in different models, which occurs at lower Al_2O_3 concentrations in lower pressure models. Labels in (d) show how Al- and Ti-rich clinopyroxene precipitation causes a decrease in the melt TiO_2 concentration in models at $P > 50$ MPa.

Figure 10 – Liquid lines of descent (black lines) produced by Rhyolite-MELTS at different initial H_2O concentrations but identical pressure and fO_2 conditions see legend. Data points show the compositions of Aluto whole rock samples from Hutchison et al. (2016c) and are distinguished by composition and the timing of eruption (see legend). Panel (a) shows that the Na_2O content of the melt decreases at >65 wt% SiO_2 for models at $H_2O > 1$ wt%. Labels in (c) show plagioclase-in and demonstrate that plagioclase precipitates at higher Al_2O_3 and SiO_2 concentrations (i.e. lower temperatures) at higher initial H_2O concentrations. Labels in (d) show where Ti-rich clinopyroxene saturates in models where initial $H_2O > 1$ wt%, resulting in a poor match between model outputs and basaltic-to-intermediate composition samples from Aluto.

Figure 11 – Schematic diagram showing the proposed structure of the sub-volcanic magma plumbing system at Aluto, as interpreted from combined InSAR data (Hutchison et al., 2016b), seismicity (Wilks, 2016) and petrologic constraints (this study), adapted from Hutchison et al. (2016b). The panel on the right shows the number of recorded seismic events versus depth beneath the volcano (red line). Comparison of our Rhyolite-MELTS fractional crystallisation models with the compositions of natural erupted samples, suggests that the most likely depth of magma storage at Aluto is at $\sim 5.6 \pm 1$ km. This is consistent with the depth of the deformation source responsible for ground uplift at Aluto in 2008 (~ 5.1 km, represented by the red dot; Hutchison et al., 2016b) and a region low seismicity beneath the volcano recorded between 2012 and 2014 (4 to 7 km; Wilks, 2016).

Figure 12 – The rate of SiO_2 change in the silicate melt during fractional crystallisation (i.e. SiO_2 variation as a function of time; $d[SiO_2]/dt$) versus magmatic temperature. $d[SiO_2]/dt$ is calculated after Mushkin et al. (2002). All variables in this model are from Rhyolite-MELTS outputs, apart from T_o (the wall rock temperature). We use three possible T_o values in our calculations, following the method of White et al. (2009), to simulate the range of realistic crystallisation scenarios at Aluto. $d[SiO_2]/dt$ increases dramatically after spinel and apatite-in, causing the SiO_2 content of the magma to increase rapidly through intermediate magma compositions.

- 951 **Figure 1** – colour, 2 column fitting
- 952 **Figure 2** – colour (online only), 2 column fitting
- 953 **Figure 3** – black and white, 1.5 column fitting
- 954 **Figure 4** – colour, 1 column fitting
- 955 **Figure 5** – black and white, 2 column fitting
- 956 **Figure 6** – black and white, 2 column fitting
- 957 **Figure 7** – black and white, 2 column fitting
- 958 **Figure 8** – black and white, 1 column fitting
- 959 **Figure 9** – black and white, 2 column fitting
- 960 **Figure 10** – black and white, 2 column fitting
- 961 **Figure 11** – colour, 2 column fitting
- 962 **Figure 12** – black and white, 1 column fitting

963 **TABLE TITLES AND CAPTIONS**

964 **Table 1** - Whole rock compositions of samples erupted at Aluto and the surrounding area, from Hutchison et al. (2016c). All data were measured by X-ray Fluorescence
965 Spectroscopy.

Sample	Rock Type	Concentration (wt%)												
		SiO ₂	TiO ₂	Al ₂ O ₃	FeO _t	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	SO ₃	LOI	Total
17-01-05	Basaltic lava	46.41	2.089	13.7	12	0.185	9.95	11.24	2.37	0.627	0.361	0.009	0.98	99.92
18-11-01	Basaltic lava	50.24	2.75	15.25	12	0.262	3.77	7.54	4.03	1.649	1.29	0.024	0.49	99.29
15-01-01B	Lithic clast from PDC	52.53	2.462	15.53	11.37	0.245	3.4	6.59	4.98	1.816	1.235	<0.002	0.28	100.44
15-01-07B	Scoria	59.85	1.645	15.13	8.15	0.144	2.53	4.83	4.64	2.705	0.271	0.065	0.57	100.53
15-02-09	Trachyte lava	65.18	0.89	15.46	6.36	0.222	0.59	2.14	5.46	3.38	0.206	0.005	0.31	100.21
18-11-06	Trachyte lava	66.38	0.53	14.78	6.39	0.318	0.17	1.05	6.34	3.215	0.063	-0.007	0.76	99.98
18-11-07b	Trachyte tuff	65.92	0.64	15.9	5.97	0.1	0.12	0.94	5.92	3.315	0.072	-0.008	1.14	100.04
18-01-04	Pumice from PDC	69.37	0.289	8.44	6.92	0.287	0.04	0.28	5.35	4.277	0.013	0.011	3.67	98.95
30-01-03B	Pumice from PDC	70.74	0.241	8.49	6.46	0.262	<0.004	0.2	5.81	4.275	0.012	<0.002	3.21	99.69
18-01-11	Pumice from PDC	71.04	0.214	8.6	6.29	0.238	<0.004	0.21	4.93	4.573	0.01	0.005	3.54	99.64
30-01-01B	Pumice from PDC	71.33	0.233	8.92	5.97	0.223	<0.004	0.2	5.7	4.364	0.011	0.004	2.83	99.8
26-01-21A	Pumice from PDC	71.79	0.273	8.72	6.37	0.254	<0.004	0.23	5.74	4.319	0.009	<0.002	2.22	99.92
02-02-03	Pumice from PDC	71.83	0.173	8.48	6.54	0.272	<0.004	0.19	6.17	4.268	0.01	0.007	1.91	99.84
02-02-02	Pumice from PDC	72.25	0.164	8.52	6.6	0.275	<0.004	0.19	5.98	4.289	0.009	<0.002	2.04	100.31
30-01-06B	Pumice from PDC	72.41	0.233	9.05	5.98	0.223	<0.004	0.2	5.22	4.41	0.01	<0.002	2.19	99.92
16-01-05	Pumice from PDC	72.43	0.168	8.14	6.31	0.261	0.01	0.24	5.51	4.155	0.084	<0.002	2.68	99.99
27-01-03	Pumice airfall	70.61	0.286	10.3	5.71	0.21	<0.004	0.25	5.45	4.387	0.009	<0.002	2.65	99.87
03-02-23	Pumice airfall	70.86	0.167	7.78	7.2	0.324	<0.004	0.14	6.69	4.147	0.014	<0.002	2.14	99.47
17-01-01K	Pumice from tuff cone	71.46	0.211	10.88	3.78	0.134	0.07	0.27	2.66	4.739	0.011	<0.002	5.68	99.9
17-01-01G	Pumice from tuff cone	72.03	0.289	9.17	5.14	0.184	0.01	0.25	2.72	4.968	0.008	<0.002	5.15	99.91
03-02-28	Pumice cone	71.34	0.215	8.63	6.28	0.239	<0.004	0.19	5.63	4.292	0.01	0.008	3.16	100
18-02-04	Aphanitic rhyolite lava	70.02	0.43	12.08	6.2	0.19	0.1	0.18	5.32	4.719	0.011	-0.01	0.82	100.05
02-02-01	Aphyric obsidian	70.71	0.352	9.73	6.63	0.275	<0.004	0.28	6.4	4.391	0.016	<0.002	-0.32	98.46

02-02-12	Aphyric obsidian	72.68	0.3	10.03	5.84	0.222	<0.004	0.24	6.15	4.389	0.011	<0.002	-0.22	99.65
01-02-14	Porphyritic obsidian	71.3	0.171	8.02	7.41	0.318	<0.004	0.18	6.86	4.284	0.011	<0.002	-0.29	98.26
26-01-SMP	Porphyritic obsidian	71.77	0.279	8.69	6.38	0.254	<0.004	0.25	5.87	4.281	0.01	<0.002	1.94	99.72
01-02-24	Porphyritic obsidian	72.7	0.18	8.44	7.13	0.296	<0.004	0.2	6.52	4.315	0.01	<0.002	-0.21	99.58
18-01-08	Porphyritic obsidian	73.15	0.222	8.88	6.48	0.246	<0.004	0.2	6.31	4.274	0.009	<0.002	-0.04	99.74
01-02-13	Porphyritic obsidian	73.3	0.243	8.79	6.65	0.268	<0.004	0.2	6.19	4.352	0.01	<0.002	-0.17	99.83
30-01-LNE	Porphyritic obsidian	73.36	0.245	8.99	6.29	0.236	<0.004	0.2	6.01	4.359	0.009	<0.002	-0.14	99.56
31-01-LE	Porphyritic obsidian	73.56	0.318	9.78	5.59	0.216	<0.004	0.22	5.8	4.353	0.009	<0.002	-0.4	99.45
13-05-04	Welded ignimbrite	71.13	0.34	11.46	5.8	0.219	0.1	0.39	5.79	4.514	0.025	-0.001	0.22	100
15-01-07A	Silicic enclave within scoria cone	72.85	0.341	10.84	5.79	0.213	0.12	0.32	5.41	4.187	0.026	<0.002	0.19	100.27
Following data is from Di Paola (1972)														
DP4	Scoriaceous boulder	42.38	4.15	14.59	14.8	0.21	6.24	10.65	2.88	0.64	0.44	2.24	-	99.22
DP34	Aphyric obsidian	70.45	0.44	9.32	6.97	0.36	0.05	0.33	6.18	4.48	0.01	3.74	-	102.33
DP35	Aphyric obsidian	72.23	0.34	8.51	7.01	0.38	0.04	0.23	6.13	3.94	0	0.91	-	99.72
Following data is from Teklemariam (1996) and Teklemariam et al. (1996)														
S-1	Ignimbrite (Munesa escarpment)	70.21	0.5	12.23	5.85	0.17	0.3	0.13	4.06	4.09	0.03	1.91	-	99.48
S-2	Coarsely porphyritic basaltic lava	49.76	2.86	16.79	11.56	0.17	4.15	7.83	3.28	1.1	0.62	1.49	-	99.61
S-3	Grey welded ignimbrite	69.92	0.44	10.37	6.74	0.27	0.21	0.54	5.98	4.38	0.02	0.68	-	99.55
Following data is from Yimer (1984) and Mamo (1985)														
ETH-A12	Coarsely porphyritic basaltic lava	50.45	2.84	18.4	10.87	0.17	2.03	7.48	4.43	1.35	0.66	0.56	-	99.24
ETH-A13	Aphyric basaltic lava	50.08	2.78	14.44	13.93	0.26	3.43	7.07	3.87	1.79	1.43	0.01	-	99.09
Mamo-1494	Porphyritic basaltic lava	48.04	2.81	15.18	11.84	0.2	4.24	7.52	2.95	1.5	0.65	3.43	-	98.36

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967 **Caption/footnote of Table 1:** Samples 17-01-01G and 17-01-01K have low Na₂O and high LOI values, which may reflect post-emplacement alteration of otherwise pristine
968 pumice (Hutchison et al., 2016c), as identified in studies of other volcanic systems (Fontijn et al., 2013; Peccerillo et al., 2003). Uncertainty on the whole-rock analyses of
969 Hutchison et al. (2016c) are reported as counting statistical errors (cse) and analysis of a USGS standard was used to assess the data quality (see Hutchison et al., 2016c for
970 details). Average cse for samples used in this study are: SiO₂ 0.032%; TiO₂ 0.002%; Al₂O₃ 0.014%; Fe₂O₃ 0.003%; MnO 0.001%; MgO 0.003%; CaO 0.002%; Na₂O 0.010%; K₂O
971 0.005%; P₂O₅ 0.000%; SO₃ 0.001%.

972 **Table 2** – Magma storage conditions investigated in Rhyolite-MELTS models, the range of values used and the
973 intervals over which they were varied. The range of model input variables are constrained based on previous
974 studies of peralkaline magma systems as cited in the table.

Parameter	Range	Interval	Previous constraints
Pressure (MPa)	50 to 300	50	Experimental work of Caricchi et al. (2006) has shown that peralkaline rhyolites cannot be generated from fractional crystallisation of a transitional basaltic parent at pressures above 0.5 GPa. Hence, lower pressure conditions are investigated.
Oxygen Fugacity (log units relative to the Quartz-Fayalite-Magnetite buffer)	-2 to +1	1	Macdonald (2012) suggests that peralkaline magmas commonly have an fO_2 between QFM-1 and QFM. This is supported by the results of White et al. (2005) which suggested the oxygen fugacity of peralkaline rhyolites from Pantelleria lie near the QFM buffer.
Water Content (wt%)	0.5, 1 to 3	1	White et al. (2009) found that initial water contents in transitional basaltic parental magmas are likely to be around 1.5wt% H ₂ O, resulting in peralkaline rhyolite with 4-6wt% H ₂ O pre-eruption. This is supported by measured concentrations of H ₂ O in basalts (0.9-1.6 wt%) and pantellerites (up to 5 wt%) from pantelleria (Giocada and Landi, 2010; Neave et al., 2012). Field et al. (2012) indicated that lower initial water contents (<1 wt%) may be present in parental magmas for peralkaline rhyolites.

976 **Table 3** – Summary of magma storage conditions identified previously in peralkaline volcanic systems and
 977 comparison with the storage conditions at Aluto identified in this study.

978 Volcano	Region	Relevant study	Method	Storage conditions and comments
Aluto	CMER	This study	MELTS	'Best fit' storage conditions are identified to be: $fO_2 = QFM$; $P = 150$ MPa; initial $H_2O = 0.5$ wt%.
Boseti	NMER	Ronga et al. (2010)	MELTS	Composition of eruptive products are reproduced by thermodynamic modelling at 100 MPa, 1 wt% H_2O , $fO_2 = QFM$.
Gedemsa	NMER	Peccerillo et al. (2003)	MELTS	Composition of eruptive products are reproduced by thermodynamic modelling at 50 MPa, 1 wt% H_2O , $fO_2 = QFM$.
Dabbahu	Afar	Field et al. (2012; 2013)	Melt inclusions; InSAR; seismics; Fe-oxide oxybarometry	Shallow storage (stacked sills) at 1-5km b.s.l. hypothesised deeper storage (14-20 km depth) where bulk of magmatic differentiation occurs. Low water content <1wt% ($MgO = 9$ wt%) and $fO_2 = QFM$.
Pantelleria	Straits of Sicily	White et al. (2005; 2009); Di Carlo et al. (2010)	MELTS; QUILF; Fe-oxide oxybarometry	Composition of trachytic magmas can be generated by fractional crystallisation models at 100 MPa, 1 – 1.5 wt% H_2O ($MgO = 6$ wt%), $fO_2 = QFM-1$. Additionally, QUILF equilibria and Fe oxide pairs indicate crystallisation near the QFM buffer for highly peralkaline magmas. Finally, experimental results are consistent with crystallisation at 5 ± 1 km depth.
Eburru	Kenyan peralkaline province	Ren et al. (2006)	QUILF	Calculated temperatures of 700-708°C and $fO_2 = QFM - 1.6$ to +0.5.
Olkaria	Kenyan peralkaline province	Scaillet and Macdonald (2001)	Experimental analysis on comenditic samples	Experiments indicate that phenocryst assemblages crystallised at $fO_2 \sim QFM$, $T = 660 - 740^\circ C$, and $P = 50 - 150$ MPa.
Bursey; Andrus; Kosciusko	Marie Byrd Land, Antarctica	LeMasurier et al. (2011)	Comparison of phonolitic and pantelleritic lavas and consideration of phase stability	Polybaric processes may be important, but the final stages of pantellerite evolution is likely to take place at <5 km depth, which favours fractionating a high plagioclase/clinopyroxene ratio increasing the peralkalinity of a magma.
Furnas	São Miguel, Azores	Jeffery et al. (2016)	MELTS	$P = 150$ MPa, $fO_2 = QFM$, initial $H_2O = 1.5$ wt% ($MgO = 10$ wt%).























