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How to fragment peralkaline rhyolites: Observations on pumice using combined multi-scale 2D and 3D imaging

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Abstract

Peralkaline rhyolites are volatile-rich magmas that typically erupt in continental rift settings. The high alkali and halogen content of these magmas results in viscosities two to three orders of magnitude lower than in calc-alkaline rhyolites. Unless extensive microlite crystallisation occurs, the calculated strain rates required for fragmentation are unrealistically high, yet peralkaline pumices from explosive eruptions of varying scales are commonly microlite-free. Here we present a combined 2D scanning electron microscopy and 3D X-ray microtomography study of peralkaline rhyolite vesicle textures designed to investigate fragmentation processes. Microlite-free peralkaline pumice textures from Pantelleria, Italy, strongly resemble those from calc-alkaline rhyolites on both macro and micro scales. These textures imply that the pumices fragmented in a brittle fashion and that their peralkaline chemistry had little direct effect on textural evolution during bubble nucleation and growth. We suggest that the observed pumice textures evolved in response to high decompression rates and that peralkaline rhyolite magmas can fragment when strain localisation and high bubble overpressures develop during rapid ascent.

Keywords: peralkaline rhyolite, fragmentation, textural analysis, X-ray microtomography

1. Introduction

Peralkaline rhyolites, although less common than their calc-alkaline counterparts, are nonetheless found in many settings including continental rifts, ocean islands and back-arc basins. During the Holocene, central volcanoes along the East African Rift, from Afar to Tanzania, have produced explosive ignimbrite-forming eruptions of peralkaline magma (Macdonald et al. 1987). Today, these volcanic centres threaten many hundreds of thousands of people, yet the dynamics of peralkaline eruptions are poorly understood and have never been observed directly. Despite their high silica contents, peralkaline melts have a relatively low viscosity (equivalent to calc-alkaline andesite for similar water contents) as a result of their alkali-rich nature (molar $(\text{Na}_2\text{O}+\text{K}_2\text{O})/\text{Al}_2\text{O}_3 > 1$, e.g., Dingwell et al. 1998; Di Genova et al. 2013). Their volatile-free viscosity is two to three orders of magnitude lower than that of calc-alkaline rhyolites: $\sim 10^8$ Pa.s for calc-alkaline rhyolite using the model of Giordano et al. (2008) versus $\sim 10^{5.5}$ Pa.s for peralkaline rhyolite using the model of Di Genova et al. (2013), both at 1223 K. Peralkaline rhyolite viscosities are so low that the fragmentation threshold for brittle failure (10^8 to 10^9 Pa.s; Papale 1999) should never be reached during magma ascent and degassing unless significant microlite crystallisation takes place (Di Genova et al. 2013), though recent numerical modelling has suggested that initial temperature may also exert a strong control on the depth of brittle fragmentation and whether it can occur at all (Campagnola et al. 2016).

Peralkaline magmas are associated with a large range of eruption styles (Houghton et al. 1985a, 1985b, 1987, 1992; Mahood and Hildreth 1986; Stevenson and Wilson 1997). For example, on the island of Pantelleria, Italy, magmas with near-identical major element compositions have produced domes, lava flows (including fountain-fed agglutinates), pumice cones, thick tephra fall deposits and pyroclastic flow deposits (Villari 1974; Mahood and Hildreth 1986; Civetta et al. 1988, 1998; Stevenson and Wilson 1997; White et al. 2009; Neave et al. 2012; Williams et al. 2013). The widespread welding and rheomorphism of the ignimbrites and fall deposits (Schmincke 1974; Wolff and Wright 1981; Mahood 1984) are a consequence of the low viscosity and correspondingly low glass transition temperature (T_g) of peralkaline melts, which can allow deformation to continue for many days after emplacement (Di Genova et al. 2013).

In this study, we use textural observations made on pumices from Pantelleria, Italy, to investigate the mechanisms of peralkaline rhyolite fragmentation. Our aim is to unravel the vesiculation and crystallisation processes in operation during magma ascent and hence understand magma properties to the point of fragmentation. Vesicle textures preserve information about bubble nucleation and growth, but are also modified by deformation, coalescence and outgassing (e.g., Sparks 1978; Klug and Cashman 1994; Sable et al. 2006). A crucial assumption made when interpreting pyroclast vesicle textures is that they represent the magma at the moment of fragmentation; that they have experienced no post-fragmentation modification (e.g., Houghton and Wilson 1989). This assumption is valid when samples are rapidly quenched, as is the case for many pumices from Pantelleria, but the timescale over which textural modification occurs depends on magma viscosity, magma composition and the depth of fragmentation (Gurioli et al. 2015).

In order to examine vesicle and crystal textures, as well as their interrelationships, in detail, we combined the complementary methods of multiscale 3D X-ray microtomography (XMT) and high resolution 2D scanning electron microscopy (SEM) (e.g., Gurioli et al. 2008; Giachetti et al. 2011). By integrating these techniques, we obtained high spatial resolution information about the geometry of objects in three dimensions, which is critical for understanding eruption processes (Baker et al. 2012). We compare our data to published textural studies of explosive eruptions, and assess similarities and differences in textures, bulk porosities, vesicle population characteristics and strain localisation features. By integrating textural and geochemical data, we reconstruct the peralkaline fragmentation process that accompanies the eruptions of these magmas, and test the limits of existing models to explain magma fragmentation. Finally, we use a fragmentation model to explore the role of overpressure inside rapidly growing bubbles as a driver for strain rate-driven fragmentation during rapid ascent.

2. Geological setting

The Quaternary volcano of Pantelleria (Figure 1) lies on the thinned continental crust of the E-W extending Sicily Channel (Civile et al. 2008, 2010), and has been active for at least 324 ka (Mahood and Hildreth 1986). The mafic northwest portion of the island is separated from the caldera-dominated, felsic southwest portion by N-S striking regional faults (Catalano et al. 2009). The volcanic history of Pantelleria has been punctuated by ignimbrite-forming eruptions (Jordan et al. 2013; Rotolo et al. 2013), of which the ~45.7 ka Green Tuff eruption

was the most recent (Villari 1974; Mahood and Hildreth 1986; Scaillet et al. 2013). Continuous geochemical zonation in the Green Tuff deposit, from pantellerite (Fe-rich peralkaline rhyolite) at its base to trachyte at its top, may represent the evacuation of a stratified reservoir of cogenetic magmas (Civetta et al. 1988; Williams et al. 2013). Indeed, pantellerites are most likely formed by 70–80 % fractional crystallisation of trachytic liquids (White et al. 2009; Neave et al. 2012; Landi and Rotolo 2015). Small eruptions generating non-welded fall deposits have been most common over the last 20 ka on Pantelleria (Mahood and Hildreth 1986; Orsi et al. 1991; Scaillet et al. 2013). Deposits from these eruptions have been classed Strombolian from the limited, circular extent of their tephra dispersal (Orsi et al. 1991, 1989; Stevenson and Wilson, 1997; Rotolo et al. 2007), in line with similar observations from Mayor Island, New Zealand (Houghton et al. 1985a).

Cuddia di Mida is the site of one such Strombolian eruption, which produced a small pumice cone around the eruptive vent (Figure 1; Orsi et al. 1991). Deposits from the Cuddia di Mida eruption are characteristic of the numerous small explosive eruptions that have taken place since the ~45.7 ka Green Tuff eruption, making it well suited to a study of the eruption dynamics and fragmentation of peralkaline magmas. The lowermost layer of the sequence is an explosion breccia (1 m thick) and is overlain by a poorly-sorted fallout layer, which has an increasing ash content towards the top (0.3 m). Above this is an ashy bed (0.08 m) overlain by a much thicker, massive, poorly-sorted fall deposit (1.2 m) (Orsi et al. 1991). The Cuddia di Mida deposits have not been dated, but the eruption probably occurred at a similar time to the Cuddia del Gallo eruption (7.1(±0.8) ka; Scaillet et al. 2013): a likely eruption window of 9.7(±0.6)–7.1(±0.8) ka can be inferred from the ages of the nearby Serra Fastuca and Cuddia del Gallo eruptions (Rotolo et al. 2007; Scaillet et al. 2013). A bulk sample of pumice clasts was collected from a single horizon in the middle of the upper massive layer (Orsi et al. 1991) on the Cuddia di Mida cone (36.781°N, 11.993°E). The unit consists of juvenile clasts ~1–10 cm in diameter (Figure 2). Grey clasts make up ~95 vol.%, with the remainder made up of black and mixed clasts and non-juvenile clasts which are <10 cm in diameter (obsidians, lithics and occasional enclaves). This is sample number 09PNL001 from Neave et al. (2012).

3. Methods

Density measurements of juvenile material were carried out using the method of Houghton and Wilson (1989), with the type of material (black, mixed or grey pumice) being noted.

Bulk densities were converted to porosities using a glass density of 2520 kg.m^{-3} , calculated from the Cuddia di Mida glass composition of Neave et al. (2012) at room temperature and pressure (Bottinga and Weill 1970; Lange and Carmichael 1990; Lange 1997; Toplis et al. 1994; Ochs and Lange 1999). The grey and black pumices have indistinguishable major element glass compositions and the same glass density was therefore used for both pumice types (Table 1). Grey pumices exhibit the lowest density of any juvenile material from the Cuddia di Mida eruption. In Strombolian eruptions, grey pumices are thought to represent the films that encase gas slugs and are therefore most likely to capture the moment of fragmentation (Lautze and Houghton 2005). The black and mixed pumices appear to be collapsed grey pumices and therefore were not considered further as they are unlikely to capture the moment of fragmentation. Cylinders $\sim 10 \text{ mm}$ in diameter and $\sim 10\text{--}20 \text{ mm}$ in height were cut from four clasts (A-D) of the grey pumices for qualitative textural analysis by SEM (Back Scatter Electron mode) and XMT imaging (e.g., A₁₀ in Table 2). Two additional cylinders $\sim 5 \text{ mm}$ in diameter were cut from clasts A and C (e.g., A₅) in order to acquire high quality XMT images at a range of resolutions. These two cylinders were also imaged by SEM. Full details of SEM and XMT image acquisition and processing, including the calculation of vesicle size distributions which followed the principles employed in the FOAMS software (Shea et al. 2010), are included in the supplementary material. All images, both SEM and XMT, are available from the authors upon request.

4. Results

4.1 Porosity

As the histogram of porosities (Figure 3) shows a bimodal distribution, a robust estimate of the average density of the whole population cannot be made owing to insufficient measurements (67 measurements of juvenile material, of which 48 were grey pumices and 19 were black/mixed pumices) (Bernard et al., 2015). The broad, low porosity mode (mean $36.9 \pm 12.2 \text{ vol.}\%$, equivalent density of $1.59 \pm 0.31 \text{ g.cm}^{-3}$) consists of black and mixed pumices whereas the narrow, high porosity mode (mean $78.5 \pm 2.7 \text{ vol.}\%$, equivalent density $0.54 \pm 0.07 \text{ g.cm}^{-3}$) consists exclusively of grey pumices. Sufficient measurements of the high porosity mode were made to obtain a robust estimate of its average (Bernard et al. 2015). The clasts used for textural analysis are all grey pumices from the high porosity mode. The average porosity estimated from the bulk density of A and C is $76.2 \text{ vol.}\%$, which compares well with the vesicularity calculated from 2D SEM images ($78.2 \text{ vol.}\%$).

4.2 Crystals

Crystal phases are dominantly anorthoclase and aegirine augite (tabular euhedral to angular and broken), alongside subordinate Fe-Ti oxides (generally equant) and aenigmatite (elongate bladed) (Neave et al. 2012). The average crystal content estimated using XMT images (A and C) is 3.24 vol.% (13.7 vol.% when recalculated on a vesicle-free basis) and the average aspect ratio of the crystals is 2.41. Crystal size distributions were not calculated from SEM or XMT images due to the low number of crystals present, i.e., crystal populations are not statistically robust. Therefore, only crystal area contents were measured in SEM images for calculation of crystal-free vesicle number densities. No microlites were observed, even in the highest resolution SEM images (Figure 4). The uniform BSE intensity of the pumice glasses implies that any nanolites present must be $< 0.02 \mu\text{m}^2$ (< 1 pixel on the highest resolution SEM images).

4.3 Qualitative textural analysis of vesicles

Grey pumices (A-D) show a variety of vesicle textures in both SEM and XMT images (Figures 5–7). In some regions, there is a sub-spherical, unimodal, isotropic vesicle population connected by thin melt films ($\sim 10^{-3}$ mm) that have an overall appearance resembling a polyhedral foam (Figure 5a). Some regions contain elongate vesicles which have thicker vesicle walls ($\sim 10^{-2}$ mm) than the surrounding regions and therefore appear denser (Figure 5b). Whilst vesicles within these regions are strongly aligned, nearby regions have different alignments and there is no overall bulk preferred orientation. Medium-sized vesicles ($L \sim 10^{-1}$ mm, where L is the equivalent diameter of a sphere with the same volume as the vesicle) associated with crystals are often somewhat elongated perpendicular to crystal faces and are connected to the crystals by thin melt films; the crystals themselves are often mantled by melt films (Figure 5c). The largest vesicles ($L \sim 10^0$ mm) are distributed randomly throughout the samples and have highly convoluted surfaces that are often, but not always, associated with crystals or regions of small vesicles. The films separating these large vesicles are very thin and often pinch out in the middle to widths thinner than the resolution of the SEM images ($0.15 \mu\text{m}$; Figure 5d). In SEM and XMT images, all samples display all these textures in approximately similar amounts (Figures 6 and 7) with two exceptions: in SEM images, A₅ only displays the polyhedral foam texture with occasional larger vesicles (Figure 6); and in XMT images, C₅ displays more of the elongate and orientated deformation vesicles (Figure 7).

4.4 Quantitative textural analysis of vesicles

Vesicle size varies by three orders of magnitude in clasts A and C with L ranging from 1.69×10^{-3} mm (SEM; Figure 6) to 4×10^0 mm (XMT; Figure 7). Vesicle wall thicknesses vary from below the resolution of SEM images ($< 0.15 \mu\text{m}$) to $\sim 30 \mu\text{m}$ (Figures 6 and 7). A_{10} and C_{10} contain equal proportions of circular and elongate vesicles (where elongate vesicles are defined as having long axis to short axis ratios > 2) whereas A_5 contains 33 % elongate vesicles and C_5 62 %, as observed qualitatively. Relationships between the number of vesicles per unit volume (N_V) and L from the SEM data are similar for both clasts in the range $L = 0.15\text{--}4000 \mu\text{m}$, with greater variation found at the upper and lower limits of L (Figure 8a). Stereological correction procedures from Sahagian and Proussevitch (1998) and Mangan et al. (1993) (abbreviated to SP98 and MCN93 respectively throughout), produced similar results (e.g., for A_{10} , $N_{V,\text{tot}}$ is $7.26 \times 10^5 \text{ mm}^{-3}$ using MCN93 and $6.14 \times 10^5 \text{ mm}^{-3}$ using SP98). Vesicle properties calculated with the more widely used SP98 procedure were carried forward into further calculations (Table 2; Figure 8a). The XMT data show very similar trends for clasts A and C (Figure 8b), with greater inter-sample variation for large vesicles ($L > 1 \text{ mm}$). In these samples, the XMT data extend the range of L to values half an order of magnitude greater than those recovered by SEM, and the higher number of vesicles observed at larger L means less scattered data at larger vesicle sizes (Figure 8c). At intermediate values of L ($6 \times 10^{-2} < L < 4 \times 10^{-1} \text{ mm}$), XMT and SEM data have very similar N_V distributions (Figure 8c).

For cumulative vesicle number density ($N_V > L$), changes in slope at $\sim 2 \times 10^{-2} \text{ mm}$ (from SEM data) and $\sim 5 \times 10^{-1} \text{ mm}$ (from XMT data) define three segments, which can be fitted with power-law curves (e.g., Blower et al. 2001) (Table 3; Figure 8d). At small values of L (SEM; $L < 2 \times 10^{-2} \text{ mm}$), the curve can be fitted with a power law exponent (d) of 1.96. For intermediate values of L (SEM and XMT; $2 \times 10^{-2} < L < 5 \times 10^{-1} \text{ mm}$), d increases to 3.24 and 3.28 respectively. For large values of L (XMT; $L > 5 \times 10^{-1} \text{ mm}$), d has a lower of 2.06.

The average melt corrected total vesicle number density ($N_{V,\text{tot}}^{\text{melt}}$) from SEM images is $2.52 \times 10^6 \text{ mm}^{-3}$, which is two orders of magnitude larger than the value of $4.23 \times 10^4 \text{ mm}^{-3}$ from XMT images (Table 2). $N_{V,\text{tot}}^{\text{melt}}$ values are dominated by the smallest vesicles, which can be artificially combined by XMT when image resolution is insufficient to capture the finest of melt films or artificially separated by SEM when complicated vesicles are counted

multiple times on a 2D surface. When $N_{V,tot}^{melt}$ is calculated using vesicles of $2 \times 10^{-2} < L < 2 \times 10^{-1}$ mm (the resolution range covered well by both techniques), the XMT and SEM datasets show close agreement.

The spatial correlation between crystals and moderately large vesicles identified qualitatively (Figure 5c) was tested further in A_{10} and C_{10} as they contain the most crystals and were imaged with a resolution appropriate for capturing larger vesicles. The N_V versus L relationship of all vesicles was compared to that of the 100 vesicles closest to each crystal quantified using 3D nearest neighbour analysis implemented in the SpatStat package in R (Baddeley and Turner 2005). Due to small instabilities during repeated iterations of nearest neighbour calculations, N_V versus L systematics of near-crystal vesicles are presented as a field rather than a single line (Figure 9). Vesicles near crystals have larger modal equivalent diameters by $\sim 1.5 \times 10^{-1}$ mm, verifying previous qualitative assessments.

5. Discussion

5.1 Comparison of results from SEM and XMT

By combining SEM and XMT imaging, we were able to obtain high spatial resolution images (SEM) as well as quantifying 3D relationships between objects (XMT). When applying any method with a finite spatial resolution, a population of small features may always be beyond the limits of imaging resolution. The resolution (and contrast) of the XMT data was insufficient to determine the finest of vesicle walls and the presence, or in this case absence, of microlites. Region of interest scanning, or higher resolution XMT laboratory systems, can yield 3D datasets with voxel resolutions down to 50 nm which would allow SEM-comparable imaging of thin vesicle walls, albeit within much smaller 3D volumes. However, the large, heterogeneously distributed high density crystals (aegirine augite, Fe-Ti oxides and aenigmatite) increased image noise and thus prevented observation of fine scale structures in these samples. In highly porous samples, like those investigated here, XMT image analysis generally underestimates vesicle number densities, primarily by the over-coalescence of neighbouring vesicles. Direct comparison of volcanological interpretations from SEM and XMT multiscale data should therefore be made with caution. For example, multiscale imaging studies of basaltic scoria and bombs from Villarrica observed discrepancies between SEM- and XMT-derived $N_{V,tot}$ values of a similar magnitude to those we observe at Pantelleria (Gurioli et al. 2008). In contrast, in datasets where vesicles are large with respect to the XMT voxel resolution, SEM and XMT datasets may agree well with each other, as

reported in pumices from Montserrat (Giachetti et al. 2011). Imaging using any method (optical, SEM, XMT, etc.) where the smallest feature (vesicle or vesicle wall) is less than three pixels/voxels in diameter will be subject to significant uncertainty (Lin et al. 2015).

Segmentation and separation of the vesicles in the 3D dataset was performed by automated methods (20–60 mins per step, per sample), and was entirely parameterised from the data. The processing of XMT data therefore avoided the time-consuming manual rectification required for SEM data (>16 hours per sample) and eliminates user-induced bias for feature recognition. The good agreement between the VSDs from both methods (Figure 8c) indicates that our SEM and XMT datasets can be combined to extend the range of L. Using XMT scans at two resolutions, it is theoretically possible to constrain VSDs over at least five orders of magnitude of equivalent diameter (beyond the 10^3 range observed in our sample). XMT is able to accurately define the volume of all vesicles (within the image resolution) without using stereological corrections. This is particularly important for non-spherical elongate or coalesced vesicles, which are treated poorly by standard stereological conversions applied to 2D data. For ellipsoidal vesicles, vesicle volume calculated assuming sphericity using the 2D cross-section can significantly over or underestimate volume depending on orientation relative to the 2D section plane. Vesicles with highly complex morphologies can be counted multiple times depending on their intersection with the plane of the 2D section, affecting size distributions and number densities (e.g., Sahagian and Proussevitch 1998). The limited sample area of 2D analyses impacts on the structural information extracted, and 3D imaging is critical for textural studies (Giachetti et al. 2011; Baker et al. 2012). This is highlighted by sample C₅, where the strong, localised and variably oriented fabric visible in the XMT images is entirely missed by the SEM data acquired in a single plane through the same sample volume. 3D imaging also allowed us to quantify spatial correlations between vesicles and crystals, which was not possible from 2D data due to the limited number of crystals intersected in single slices.

5.2 Bubble nucleation, growth and deformation recorded in pumice textures

Grey pumices exhibit a narrow range of porosities (78.9 ± 2.4 vol.%) and are texturally similar to one another – they have VSDs that are within error over the full range of L. The modal density of the grey pumices ($0.5\text{--}0.6 \text{ g.cm}^{-3}$) is similar to the Oira pumice cone ($0.5\text{--}0.6 \text{ g.cm}^{-3}$) and Ruru Pass Tephra ($0.4\text{--}0.5 \text{ g.cm}^{-3}$) of Mayor Island, NZ, both magmatic peralkaline eruptions of Strombolian-to-Hawaiian intensity (Houghton et al. 1987). The power-law

relationships in the cumulative VSD data imply non-equilibrium, continuous and/or accelerating nucleation and growth of bubbles; conditions common during explosive eruptions of silica-rich magmas (e.g., Mangan and Cashman 1996; Blower et al. 2001, 2003). Power law exponents, d , of < 2 have been shown experimentally to represent continuous nucleation and free growth of bubbles (Blower et al. 2001, 2003); we suggest that the smallest vesicles ($L < 2 \times 10^{-2}$ mm; $d = 1.96$) originated in this way. This value of d is comparable to those reported for vesicles of a similar size from Askja 1875 (Carey et al. 2009) and Chaitén 2008 (Alfano et al. 2012) (Table 4), where bubble development is thought to reflect a final stage of rapid decompression that occurred shortly before fragmentation at a high degree of vapour supersaturation. For intermediate vesicle sizes ($2 \times 10^{-2} < L < 5 \times 10^{-1}$ mm), our peralkaline samples have a power law exponent of ~ 3.25 , a change in slope which may have been caused by bubble coalescence overprinting continuous nucleation (Gaonac'h et al. 1996), a process that has been reported for Askja 1875 (Carey et al. 2009), Chaitén 2008 (Alfano et al. 2012), Mount Mazama 7700 BP (Klug et al. 2002) and Taupo 1.8 ka (Houghton et al. 2010) (Table 4). This intermediate-sized population of vesicles includes heterogeneously distributed bubbles that we interpret as having nucleated early on phenocrysts at low degrees of supersaturation (Figures 4c and 9). Our largest vesicle population ($L > 5 \times 10^{-1}$ mm) returns to a power law exponent typical of continuous nucleation and free growth ($d = 2.06$), which we suggest could be related to dynamic processes such as tearing and deformation during fragmentation, but has not been noted in previous studies.

There is a high degree of spatial heterogeneity in vesicle deformation over small length scales (< 1 mm), suggesting that strain was localised (Wright and Weinberg 2009). This is especially noticeable in C₅ (Figure 7). The presence of deformed, elongated vesicles (with elongation factors often > 10) suggests that maximum strain rates during the eruption were locally much higher than those that would be calculated using bulk parameters (e.g., conduit radius and volume flux). However, the larger, near-crystal vesicle population shows little or no deformation, which suggests the possible formation of strain shadows around crystals. The spatial relations between crystals and deformation require further investigation before this can be quantified.

To compare vesicle textures of the Cuddia di Mida eruption with those from other eruptions, literature data from a variety of magmatic (i.e., not phreatomagmatic) eruptions is shown in Figure 10. Figure 10a displays N_V versus melt SiO₂ content for a wide range of magma

compositions (basalt to rhyolite) and eruption styles (Strombolian to Plinian). In general, rhyolitic eruptions have higher N_V than basaltic eruptions, although some basaltic Plinian eruptions reach values similar to rhyolitic eruptions. Within basaltic eruptions, Plinian eruptions tend to have higher N_V than Strombolian events but the values do overlap. Conversely, N_V for rhyolitic eruptions does not correlate with eruption style as the small cone-forming events have N_V values similar to those from sub-Plinian and Plinian events. For example, the Cuddia di Mida eruption has N_V values similar to those from a small cone-forming rhyolitic eruption on Raoul (Rotella et al. 2014) and from sub-Plinian to Plinian rhyolitic eruptions. These values are one-to-four orders of magnitude larger than basaltic Strombolian eruptions and at the maximum values for basaltic Plinian eruptions. However, the total vesicle number densities we report for the Cuddia di Mida eruption are an order of magnitude larger than those reported from member A of the peralkaline Green Tuff eruption by a recent study (Campagnola et al. 2016).

Figures 10b and 10c only include a sub-set of the eruptions used in Figure 10a selected to represent data from two end-member fragmentation mechanisms (Gonnermann 2015): inertia-driven break-up of low viscosity melt (e.g., basaltic Strombolian) and strain-induced brittle failure (e.g., crystal-free rhyolitic Plinian). Crystal-free rhyolitic eruptions were chosen as the Cuddia di Mida eruption contains only a minor phenocryst component and no microlites, implying that a high crystal content did not lead to fragmentation. As expected, comparing N_V to melt viscosity (Figure 10b) shows a very similar trend to comparing to melt SiO_2 content.

Small peralkaline eruptions have been compared to basaltic Strombolian eruptions in previous work due to their low viscosities (e.g., Houghton et al. 1985a). However, the viscosity and N_V of the Cuddia di Mida eruption are much more similar to rhyolitic eruptions than basaltic Strombolian eruptions. This may be due to the lower diffusivities of volatile species through cooler rhyolitic melts influencing bubble nucleation and growth: with slower diffusion it is easier to nucleate new bubbles than to diffuse volatiles into existing bubbles, which results in higher N_V (Sparks 1978).

Figure 10c shows vesicle size distributions (VSDs) for rhyolitic sub-Plinian to Plinian and basaltic Strombolian eruptions as well as our data from the Cuddia di Mida eruption. VSDs from single eruptions are similar to each other, but VSDs do not appear to correlate with

eruption style or magma composition in general. Basaltic Strombolian eruptions tend to have few large vesicles compared to rhyolitic eruptions but rhyolitic eruptions also span wide ranges of vesicle sizes. However, our samples from Cuddia di Mida are more similar to those from rhyolitic eruptions than from basaltic Strombolian eruptions because they contain many small vesicles that are absent in the basaltic eruptions.

The low viscosity of the peralkaline Cuddia di Mida melt does not appear to have exerted a major control on the final vesicle textures of the pumices (Figures 5 and 10). That is, the peralkaline rhyolites studied here resemble deposits from silica-rich, calc-alkaline eruptions with much higher melt viscosities, particularly with respect to minimum vesicles sizes and strain localisation features (see studies on Chaitén 2008 and the Campanian Ignimbrite from Alfano et al. (2012) and Polacci et al. (2003) respectively). The pumice textures do not resemble those of scoria from basaltic, Strombolian eruptions at Stromboli or Villarrica, which are characterised by much larger vesicles (Gurioli et al. 2008; Lautze and Houghton 2005, 2006, 2008; Polacci et al. 2009; Leduc et al. 2015). Furthermore, the $N_{V,tot}^{melt}$ values and VSDs calculated are similar to those from the products of high-silica calc-alkaline eruptions of varying size (Table 4, Figures 10a and 10c).

5.3 The fragmentation mechanism of peralkaline magmas

Interaction with external water is not considered to be a viable fragmentation mechanism for the Cuddia di Mida eruption due to the lack of field evidence for magma-water interaction (Mahood and Hildreth 1986). Furthermore, pumice clasts from Cuddia di Mida lack the fluidal shapes associated with inertia-driven fragmentation of the type observed in Hawaiian eruptions (Namiki and Manga 2008); and the total vesicle number density is one to four orders of magnitude larger than those found in the products of basaltic Strombolian eruptions. Therefore tearing apart of melt by bubble bursting is also not a viable fragmentation mechanism (Figure 10; Gonnermann 2015). Textural similarities between peralkaline and calc-alkaline pumices thus suggest similar brittle fragmentation mechanisms, despite differences in chemistry and physical properties.

Magmas fragment in a brittle fashion when a critical, viscosity-dependent strain-rate is exceeded (Papale 1999). Bulk magma viscosity depends on melt composition and on magma crystallinity and vesicularity (e.g., Rust and Manga 2002; Giordano et al. 2008; Vona et al. 2011; Mader et al. 2013). Magma water content decreases dramatically during decompression

and degassing, increasing the bulk viscosity (Giordano et al. 2008) and bringing the magma closer to fragmentation. Assuming that the melt was largely degassed at the point of fragmentation, we use the PS-GM viscosity model of Di Genova et al. (2013) to calculate a melt viscosity range of $10^{4.28}$ to $10^{7.11}$ Pa.s (at 0.0–1.0 wt.% water) at a temperature of 1075 K (Neave et al. 2012). The PS-GM viscosity model is based on a modified Vogel-Fulcher-Tammann equation and is specifically calibrated for peralkaline compositions (Di Genova et al. 2013). Including crystals (13.8 vol.%, average aspect ratio of 2.4) has a negligible effect on the bulk viscosity ($10^{4.65}$ to $10^{7.48}$ Pa.s at 0.0–1.0 wt.% water; Mader et al. 2013).

Samples contain elongate vesicles (33 – 62 % of total vesicle populations) which implies that melt capillary numbers were high and that the bulk viscosity decreased with increasing bubble content (Rust and Manga 2002). At the high vesicle volume fractions observed here (~76 vol.%), the standard models that relate viscosity to porosity are not applicable (they remain robust up to a maximum porosity of 50 vol.%; Mader et al. 2013). It is therefore not possible to calculate the bulk viscosity at the moment of fragmentation precisely. However, assuming that the melt had an initial water content of 5 wt.% (Neave et al. 2012), contained 13.7 vol.% crystals when resident in the magma chamber at 1.5 kbar (Neave et al. 2012) and carried only a negligible volume of pre-existing bubbles, we calculate a bulk viscosity of $10^{1.54}$ Pa.s prior to decompression (1075 K, Neave et al. 2012). If there was no melt-bubble separation during the initial ascent, the viscosity, bubble content and pressure-dependent melt water content up to the 50 vol.% porosity threshold can be estimated (the porosity threshold is estimated to occur at ~25 bars; Papale et al. 2006). Beyond this threshold we cannot assess the effect of bubbles on viscosity and therefore a maximum estimate for the viscosity of a bulk magma at containing 50 vol.% bubbles at fragmentation is $10^{4.15}$ to $10^{6.61}$ Pa.s (assuming 0.0–1.0 wt. % water at 1075 K; Mader et al. 2013).

The minimum bulk viscosity (μ) required for strain-induced fragmentation is defined as $\mu \geq (CG_\infty\pi r^3/Q)^{(1/0.9)}$, where r is the conduit radius (m), Q is the volume flux ($\text{m}^3.\text{s}^{-1}$), G_∞ is the elastic modulus at infinite frequency (10 GPa) and C is a fitting parameter ($0.01 (\text{Pa.s})^{-0.1}$) (Gonnermann and Manga 2003). For a realistic conduit radius of 10 m (e.g., Campagnola et al. 2016) a mass flux of 2.4×10^8 to $3.5 \times 10^{10} \text{ kg.s}^{-1}$ (equivalent to a volume flux of 3.5×10^5 to $5.8 \times 10^7 \text{ m}^3.\text{s}^{-1}$) is required to achieve the minimum strain rate required for fragmentation when considering the viscosities calculated above ($10^{4.15}$ to $10^{6.61}$ Pa.s). These should be considered as minimum mass flux estimates as bulk viscosity will likely be reduced further at

higher vesicle contents (~25 vol.% of measured porosity beyond the model limits, Mader et al. 2013). The much larger Green Tuff eruption had a comparable viscosity to the Cuddia di Mida eruption during the earliest explosive and crystal-poor part of the eruption (Campagnola et al. 2016), yet the mass fluxes we calculate to be necessary for fragmentation are much larger than those estimated for both the entire Green Tuff eruption ($\sim 2 \times 10^8$ kg.s⁻¹; Williams et al. 2013), and member A of the Green Tuff (9.3×10^5 kg.s⁻¹; Campagnola et al. 2016) and are therefore unfeasible. Conversely, achieving fragmentation using the lower bound of the published mass fluxes for these eruptions would require a conduit radius of < 1 m. Assuming strain-induced fragmentation, the calculated minimum mass fluxes and conduit radii required for fragmentation in both small (Cuddia di Mida) and large (Green Tuff) eruptions of peralkaline rhyolite respectively are thus geologically unrealistic.

An alternative mechanism invokes bubble overpressure causing strain-induced fragmentation when gas is unable to expand over the timescale of decompression due to the tensile strength of the surrounding melt (Zhang 1999; Spieler et al. 2004; Mueller et al. 2008). Although there is no permeability data available for the Cuddia di Mida pumice, the overpressure required for fragmentation (ΔP_{fr} ; MPa) can be calculated from $\Delta P_{fr} = \sigma_m / \phi$ using the known porosity (ϕ) and magma tensile strength ($\sigma_m = 0.995$ MPa; Spieler et al. 2004). With a porosity of 76 vol.%, the Cuddia di Mida pumices require a bubble overpressure of 1.3 MPa to cause fragmentation. Bubble overpressure is a function of decompression rate and melt viscosity (Barclay et al. 1995). An $N_{v,tot}^{melt}$ of 2.5×10^6 mm⁻³ implies decompression rates of the order 10^7 Pa.s⁻¹ (Toramaru 2006), and the melt viscosity gives relaxation times (τ_s) of 1.9×10^{-6} to 1.3×10^{-3} s for 1.0–0.0 wt.% water using the expression $\tau_s = \mu_s / G_\infty$ (Dingwell and Webb 1989). The onset of non-Newtonian, unrelaxed, viscoelastic behaviour at 1.9×10^{-4} to 1.3×10^{-1} s, thus implies that average decompression rates of 1.0×10^7 to 6.9×10^9 Pa.s⁻¹ are required for fragmentation. Even the lower of these estimates (for the most viscous melt) is extreme, and significantly larger than the value estimated for member A of the Green Tuff eruption (3.82×10^6 Pa.s⁻¹; Campagnola et al. 2016).

Rapid decompression following edifice collapse has been suggested to explain the explosive behaviour of other magmas with seemingly insufficient viscosity to fragment (e.g., $\sim 10^6$ to 10^8 Pa.s for Chaitén 2008; Castro and Dingwell 2009; Alfano et al. 2012). However, edifice collapse is not a viable mechanism for driving rapid decompression on Pantelleria, where cone-forming events have defined recent silicic volcanism. Instead, the high volatile content

and low viscosity of peralkaline magmas may play a crucial role in promoting rapid decompression during the initial stages of eruption.

Our 3D XMT data show significant, localised bubble deformation, implying that substantial partitioning of strain across heterogeneous samples took place prior to fragmentation. Strain localisation entails a complex interaction of shear heating (decreasing viscosity) and volatile solubility modification (increasing viscosity) that can drive gas exsolution (increasing or decreasing viscosity depending on strain rate), elastic stress unloading and changes in the rheological behaviour of vesicles (Wright and Weinberg 2009). These shear bands have been observed in low viscosity magmas, such as phonolites from Vesuvius, and are thought to develop in the conduit due to lateral velocity gradients and cause outgassing (Shea et al. 2012; 2014). These processes result in a variable and highly heterogeneous rheology on a range of spatial and temporal scales, and a consequently variable fragmentation criterion at the bubble-wall scale. Therefore, strain localisation could have permitted fragmentation to have occurred at a lower *bulk* viscosity than calculated above, but requires further empirical and theoretical investigation.

6. Conclusions

By investigating the textures of pumices erupted from the Cuddia di Mida vent on Pantelleria, Italy, we have inferred that, despite having bulk magma viscosities seemingly far too low, peralkaline magmas fragment by brittle failure. Integrating multiscale 2D and 3D analysis techniques on pumice samples allowed vesicle size and shape distribution characteristics to be defined across a wide range of equivalent vesicle diameters. The textures, bulk porosity, VSDs and $N_{V,tot}^{melt}$ values of pumices from Cuddia di Mida are comparable with those from calc-alkaline rhyolite deposits, and imply that, despite the difference in viscosity between calc-alkaline and peralkaline rhyolites, both magma types fragment by strain-induced brittle fragmentation. We show that initial nucleation occurred on large crystals at low degrees of volatile supersaturation. This was followed by some degree of coalescence and textural maturation before homogeneous, continuous nucleation occurred during rapid ascent at higher degrees of volatile supersaturation. Our data also show a possible third regime for the largest vesicles. We show that microlite-free peralkaline pumices cannot reach classically defined fragmentation conditions under even the most extreme of permitted geological conditions, and mechanisms such as bubble overpressure driven by rapid decompression and strain localisation around crystals are suggested instead. The very high decompression rates

suggested by our analysis may be aided by the high volatile content and low viscosity of peralkaline magmas.

7. Acknowledgements

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Author Contributions

The project was conceived by ME, following the work of DAN. The manuscript arose from the M.Sci. (Cambridge) thesis of ECH. DAN collected the samples and processed the SEM dataset. ECH acquired the XMT data and performed the analysis under the supervision of KJD. PJW provided access to the MXIF. ECH led manuscript production with further contribution from all authors.

Tables

Table 1 Major element glass composition in wt.% from Neave et al. (2012) for grey and black pumices from Cuddia di Mida. 1σ errors are shown.

	SiO ₂	TiO ₂	Al ₂ O ₃	FeO	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅
Grey	70.74±0.35	0.25±0.02	7.18±0.06	8.73±0.14	0.36±0.01	0.04±0.02	0.34±0.03	6.83±0.07	4.44±0.07	0.01±0.00
Black	70.92±0.27	0.26±0.03	7.14±0.09	8.69±0.20	0.37±0.02	0.04±0.02	0.34±0.03	6.88±0.09	4.49±0.08	0.01±0.00

Table 2 Vesicle and crystal data from SEM and XMT.

Sample	Density g.cm ⁻³	Porosity vol.%	Vesicularity vol.%	Proportion elongate vesicles	Crystal (vesicle free) vol.%	Crystal aspect ratio	Microlite vol.%	N _{A,tot} (mm ⁻²)	N _{v,tot} MCN93 (mm ⁻³)	N _{v,tot} ^{melt} MCN93 (mm ⁻³)	N _{v,tot} SP98 (mm ⁻³)	N _{v,tot} ^{melt} SP98 (mm ⁻³)	N _{v,tot} XMT (mm ⁻³)	N _{v,tot} ^{melt} XMT (mm ⁻³)
A ₁₀	0.58	76.9	79.2	0.49	2.62 (11.4)	2.48	0	2.51×10 ³	7.26×10 ⁵	3.14×10 ⁶	6.14×10 ⁵	2.66×10 ⁶		
A ₅	0.58	76.9	77.6	0.50	3.99 (17.3)	2.36	0	2.41×10 ³	7.00×10 ⁵	3.03×10 ⁶	6.03×10 ⁵	2.61×10 ⁶		
C ₁₀	0.60	76.0	83.0	0.33	3.81 (16.1)	2.44	0	1.98×10 ³	6.49×10 ⁵	2.70×10 ⁶	5.80×10 ⁵	2.42×10 ⁶		
C ₅	0.67	73.8	73.5	0.62	2.82 (10.8)	2.02	0	2.29×10 ³	7.29×10 ⁵	2.78×10 ⁶	6.16×10 ⁵	2.35×10 ⁶		
Average	0.60	76.2	78.2	0.44	3.24 (13.7)	2.41	0	2.34×10 ³	7.05 ×10 ⁵	2.93×10 ⁶	6.08×10 ⁵	2.52×10 ⁶	1.02×10 ⁴	4.23×10 ⁴

Porosity is calculated using the bulk density measurements with volumes measured from low resolution XMT images. Vesicularity and microlite content are calculated from the highest resolution SEM images. Crystal content and aspect ratios are calculated from XMT images.

Table 3 Power law exponents (d) and vesicle equivalent diameter (L) break in slope values for small (s), medium (m) and large (l) vesicle populations using SEM and XMT data.

	L _{s-m} (mm)	L _{m-l} (mm)	d _s	d _m	d _l
SEM	2.5×10 ⁻²		1.96	3.24	
XMT		5.0×10 ¹		3.28	2.06

Table 4 Summary table for crystal-poor rhyolitic, basaltic Strombolian and peralkaline eruptions.

Volcano	Melt SiO ₂ (wt.%)	AI	Log [Anhydrous melt viscosity, (Pa.s)]	Vesicle content (vol.%)	Crystal content (vol.%)	Microlite content (vol.%)	$N_{v,tot}^{melt}$ ($\times 10^6 \text{ mm}^{-3}$)	d_1	d_2
Small cone-forming									
Cuddia di Mida, Pantelleria	70.7	2.23	6.80	78.5	3.2	0	2.5	2.0	3.3
Raoul, KA	69.3	0.51	8.12	82.3	<5	<1	3.0	n.d.	3.9
Strombolian									
Stromboli, Italy	52.2–52.5	0.59–0.64	3.10–3.54	24–78	12–35	<1	0.000096–0.030	n.d.	0.7–1.3
Villarica, Chile	53.9–54.4	0.43–0.45	2.80–2.85	47.9–88.8	1.12–19.8	<1	0.00074–0.014	n.d.	n.d.
Violent Strombolian									
Vesuvius, Italy	46.7	0.67	2.66	43.2–46.3	28.7–39.1	Low	0.018–0.12	n.d.	n.d.
Sub-Plinian to Plinian									
Askja, USA	71.0–72.4	0.72–0.80	6.50–6.68	77.6–88.5	<0.5	0	0.71–2.4	1.6–2.1	3.0–5.1
Chaitén, Chile	74.2	0.73	10.61	43–80	<1	Rare	0.064–0.23	1.0–1.7	3.5–4.2
Mount Mazama, USA	70.4	0.76	8.28	78.5–85.0	10	0	0.36–6.0	n.d.	3.3
Mount St. Helens, USA	72.7–79.6	0.67–0.93	8.15–9.35	55.6–80.7	6–15	0–7	0.82–2.0	n.d.	n.d.
Pantelleria, Italy	62.7–69.4	1.0–1.8	6.52–7.28	78–81	8–22	0–11	0.026–0.35	n.d.	n.d.
Raoul, KA	68.0–69.0	0.44–0.48	7.53–8.06	34.7–88.6	<5	<22	0.98–19	n.d.	3.6–4.0
Taupo, NZ	73.7	0.76	9.85	44–89	2–3.5	Sparse	0.019–4.8	n.d.	3.2

AI is agpaite index $(\text{Na}_2\text{O}+\text{K}_2\text{O})/\text{Al}_2\text{O}_3$ in mol.%; viscosity is for the melt phase, excluding the effects of bubbles, crystals and microlites using Giordano et al. (2008) and Di Genova et al. (2013); vesicle, crystal and microlite content are relative to total volume; $N_{v,tot}^{melt}$ is the total vesicle number density corrected for vesicularity; and power law exponents (d) are for the smaller (1) and larger (2) vesicle populations. *References:* Cuddia di Mida, Pantelleria: this study, Neave et al. (2012); Stromboli, Italy: Metrich et al. (2001), Lautze and Houghton (2005, 2007), Polacci

et al. (2009), Leduc et al. (2015); Villarica, Chile: Gurioli et al. (2008); Vesuvius, Italy: Cioni et al. (2011); Raoul, KA: Barker et al. (2012), Rotella et al. (2014); Askja, USA: Sigurdsson and Sparks (1981), Carey et al. (2009); Chaitén, Chile: Castro and Dingwell (2009), Alfano et al. (2012); Mount Mazama, USA: Bacon and Druitt (1988), Klug et al. (2002); Mount St. Helens, USA: Rutherford et al. (1985), Klug and Cashman (1994); Pantelleria, Italy: Campagnola et al. (2016); Taupo, NZ: Sutton et al. (1995), Houghton et al. (2010). A more complete dataset is available in the Supplementary Material.

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Figure captions

Figure 1 Geological map of Pantelleria, Italy, with the location of the sample from Cuddia di Mida (09PNL001) indicated by the red diamond (after Mahood and Hildreth 1986).

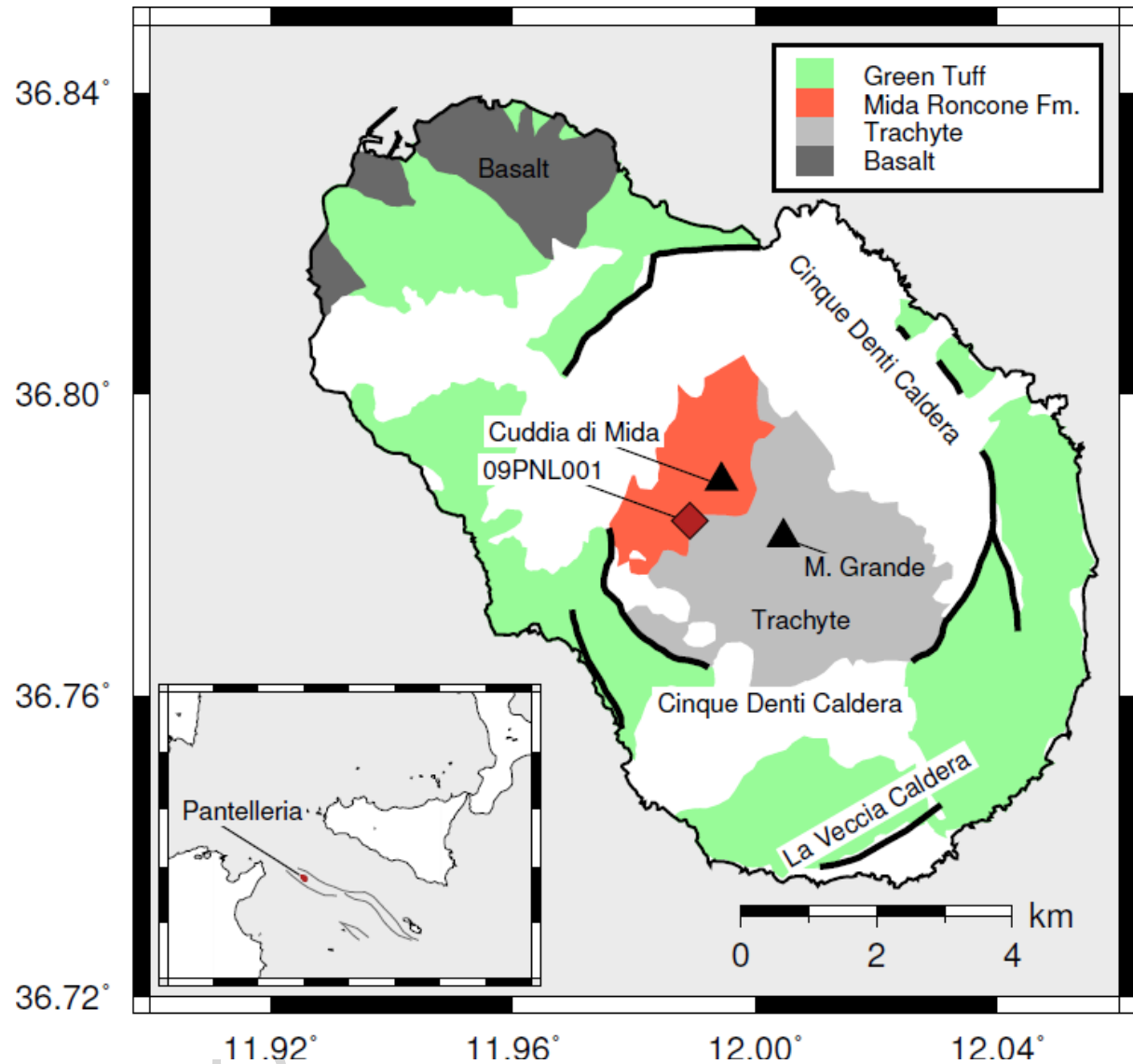


Figure 2 Photograph of the Cuddia di Mida deposit (a) (lower contact of the explosion-breccia is not visible), where * indicates the layer sampled which is shown in detail (b).

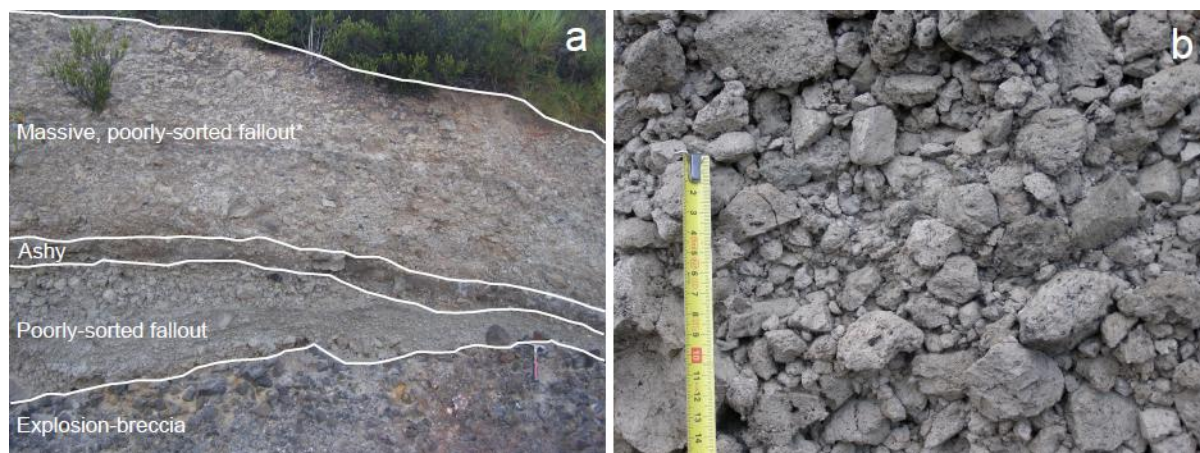
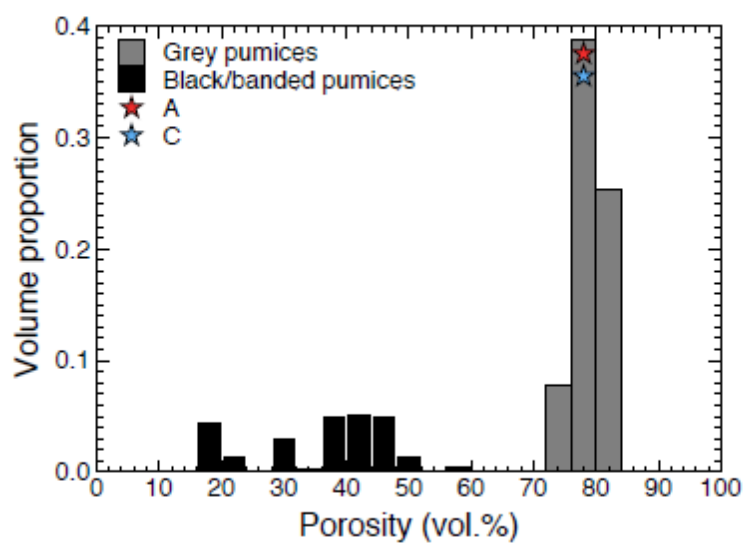


Figure 3 Porosity distribution of juvenile material from the Cuddia di Mida second airfall deposit (09PNL001) coloured for grey and black/mixed clasts. Porosity of clasts A (red) and C (blue) highlighted.



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Figure 4 SEM image at the highest resolution showing the absence of any microlites. Vesicles are black and melt is grey.

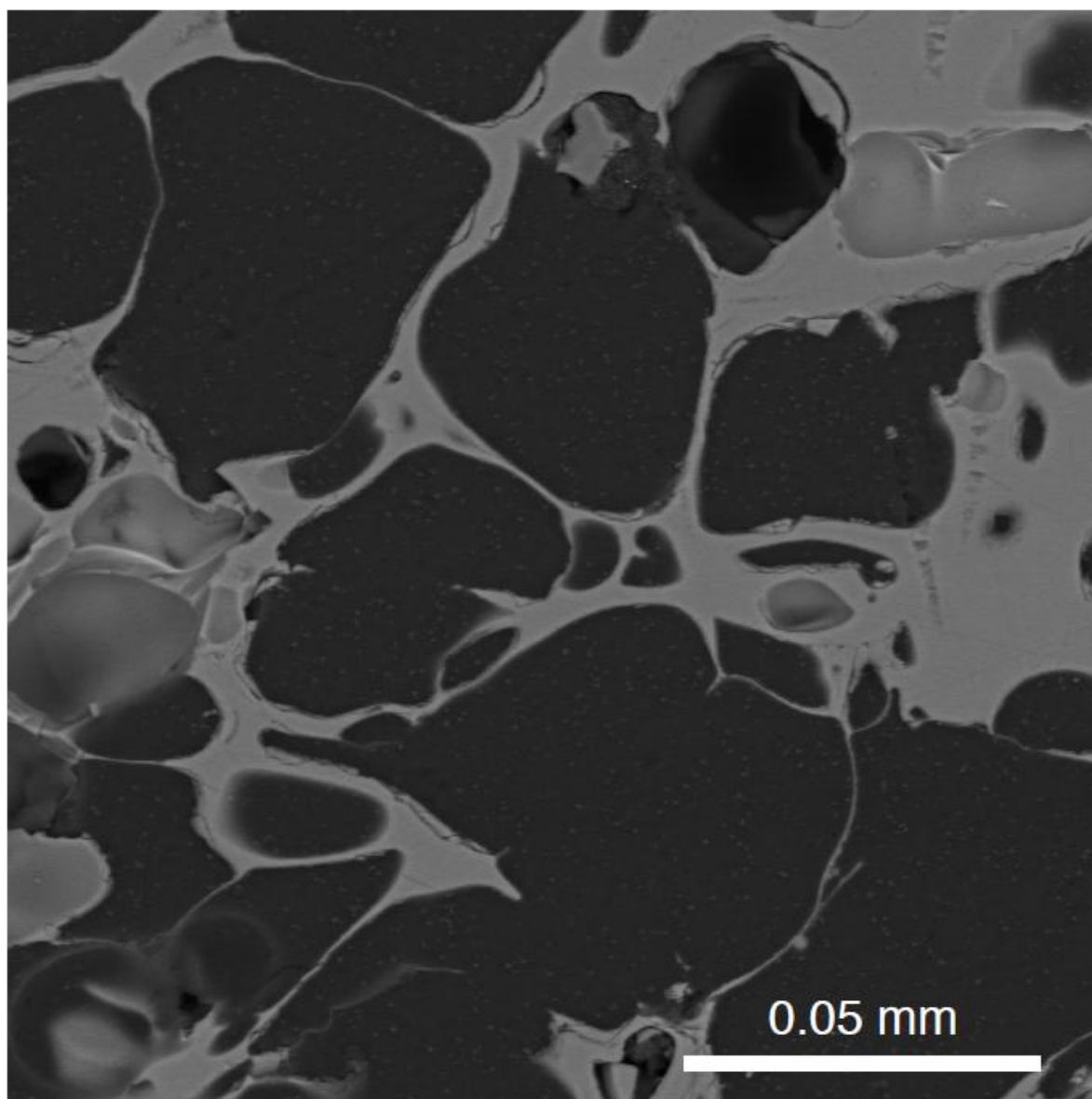


Figure 5 SEM images highlighting the different vesicle textures found in both the SEM and XMT images: a) polyhedral foam; b) sub-spherical, thicker walled vesicles; c) vesicles attached to crystal faces and; d) large vesicles with convoluted faces. Vesicles are black and melt/crystals is grey. Images shown are not necessarily stacked in order to represent typical textures at equivalent resolutions.

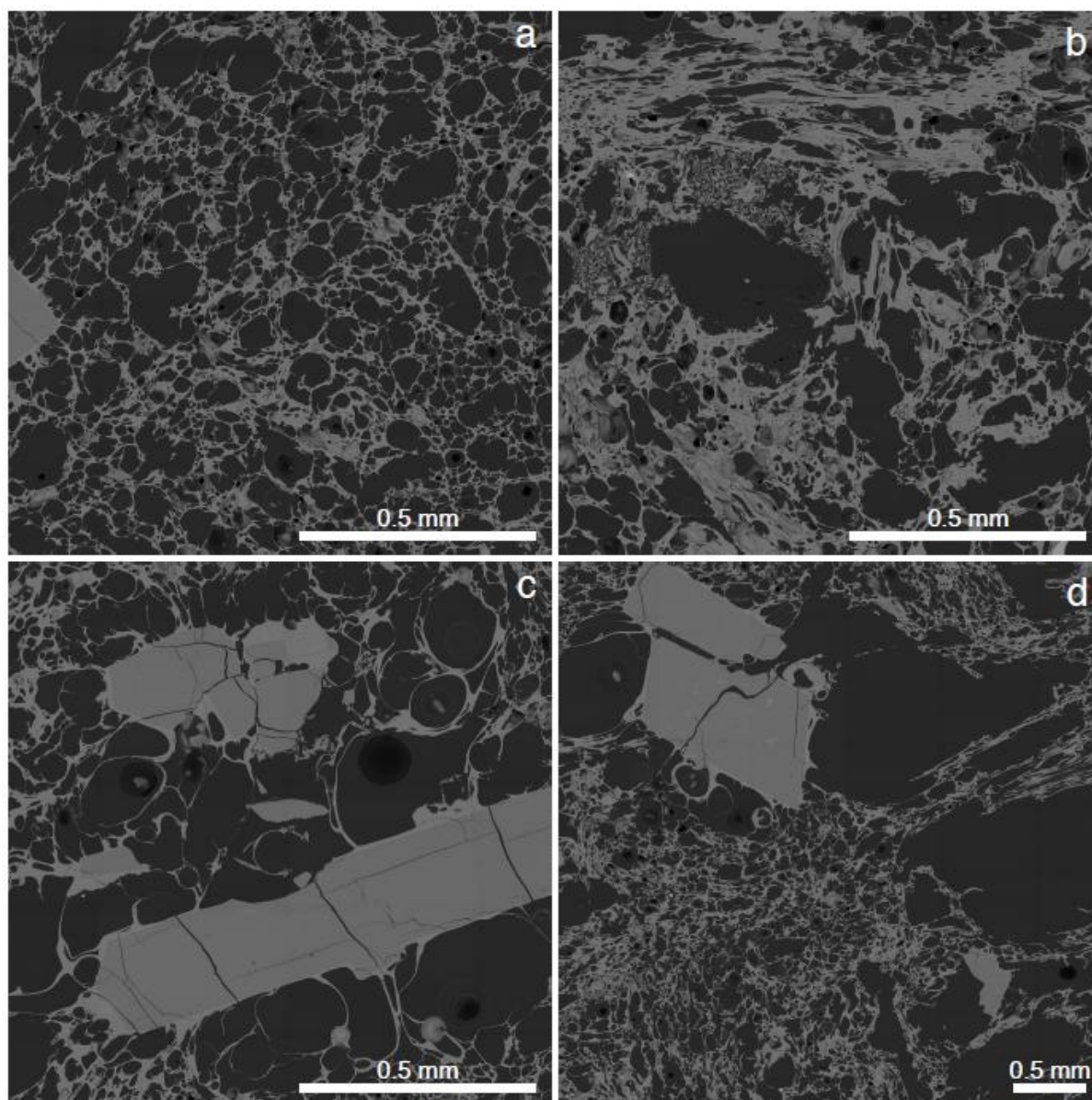


Figure 6 Selected SEM images with increasing resolution from top to bottom (field of view shown along each side). Vesicles are black and melt/crystals are grey. Sample letter shown along the top. All slices are in the XY plane of the XMT data.

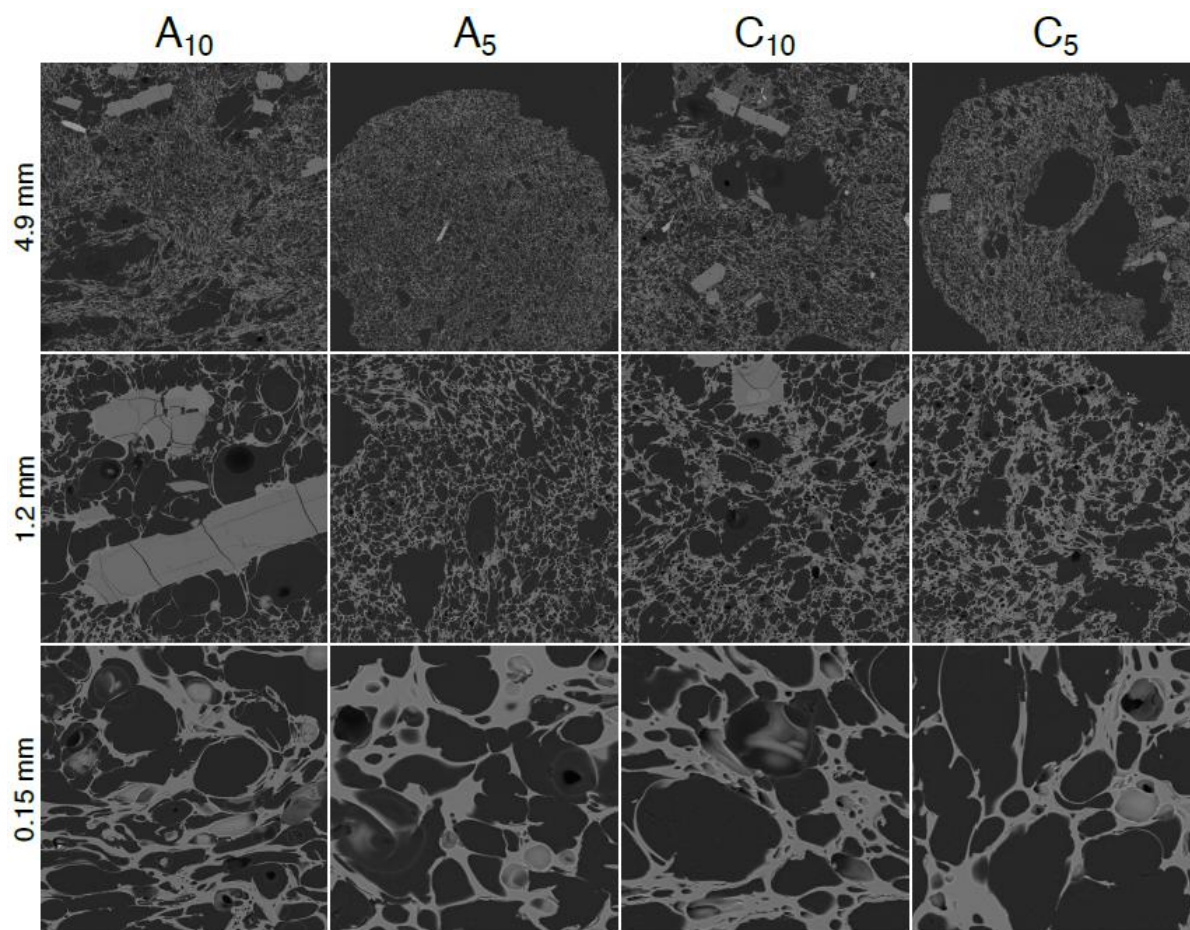


Figure 7 Selected orthogonal 2D slices through the 3D XMT images with field of view shown on the bottom. Vesicles are black, melt/feldspars/pyroxenes are grey and oxides are white. Sample letter shown along the top. Arbitrary slice orientations shown along each side.

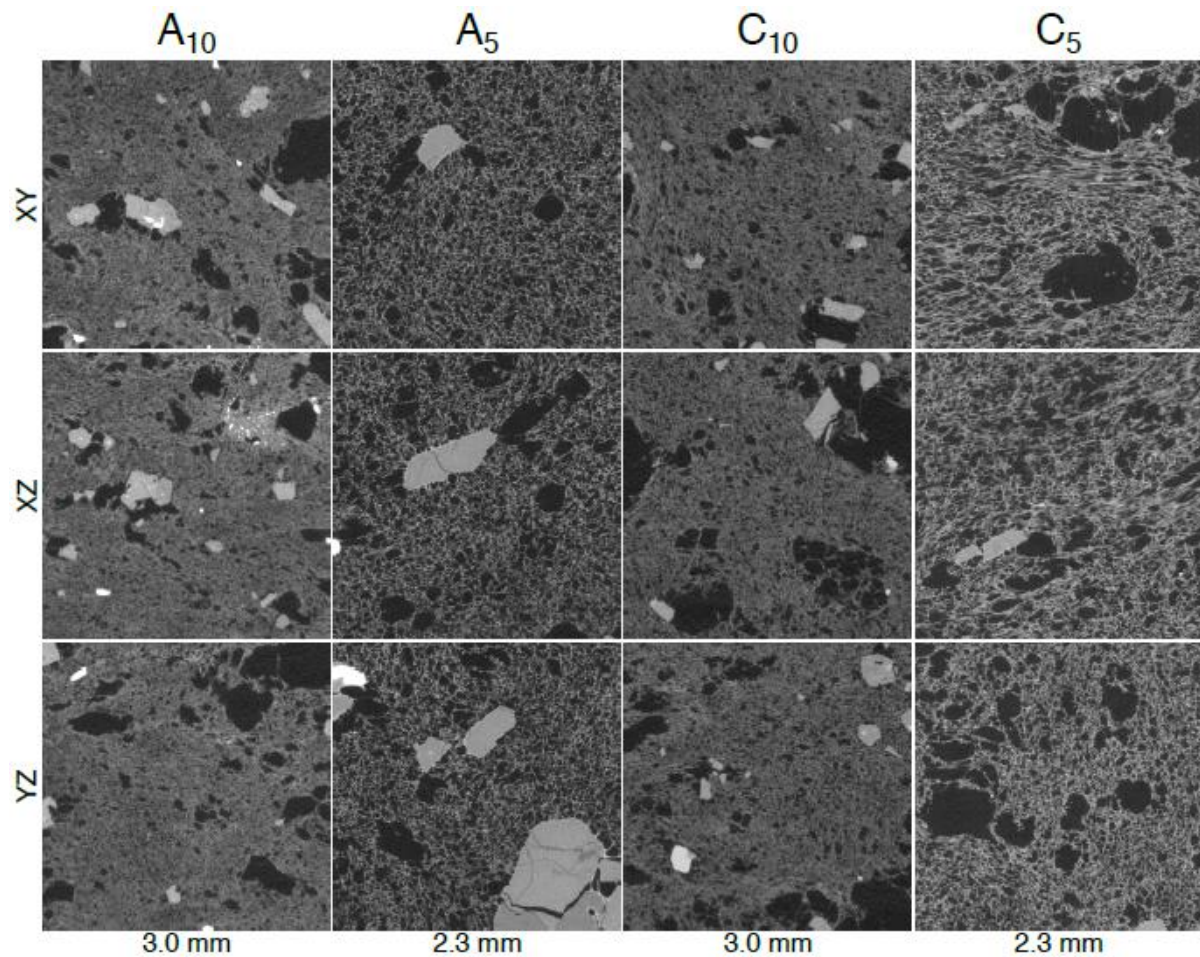


Figure 8 Vesicle size distributions (VSDs) with respect to equivalent diameter (L) for SEM and XMT data: a) SEM generated VSDs (N_V) stereologically corrected using Mangan et al. (1993) (MCN93, dashed line) and Sahagian and Proussevitch (1998) (SP98, solid line); b) XMT generated VSDs (N_V); c) comparison of VSDs generated by SEM and XMT and; d) comparison of cumulative VSDs ($N_V > L$) for SEM and XMT showing exponential and power law fits. Small, medium and large in the legend refer to the vesicle sizes.

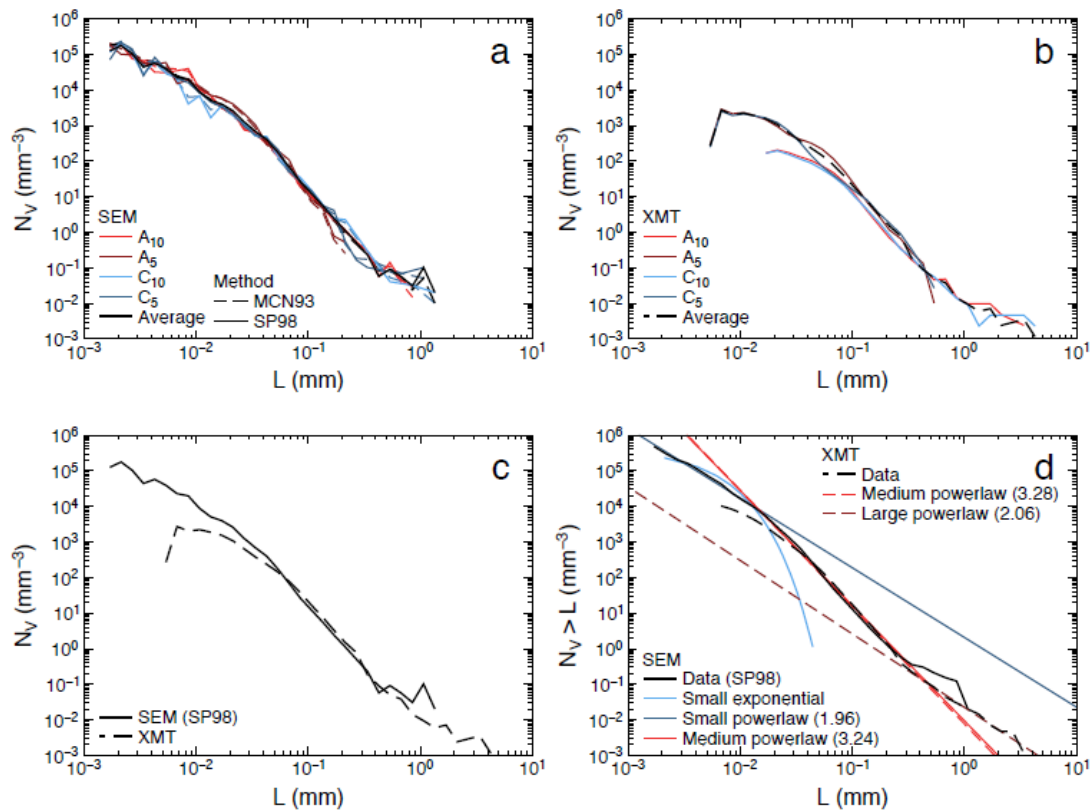
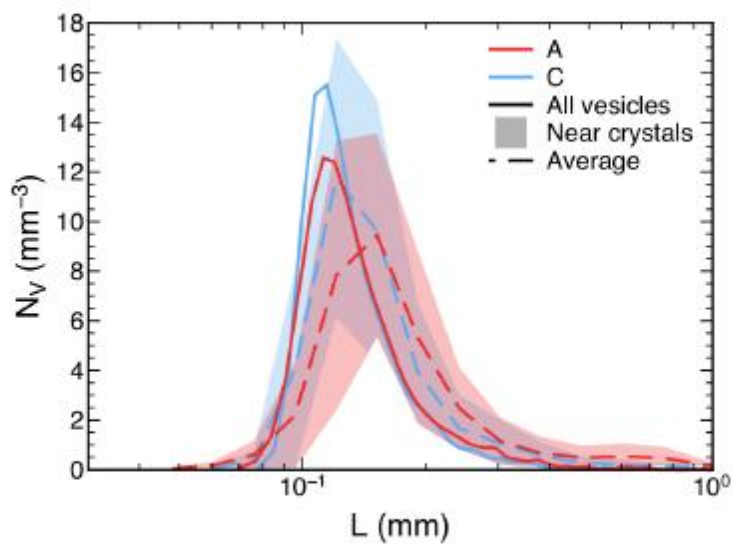
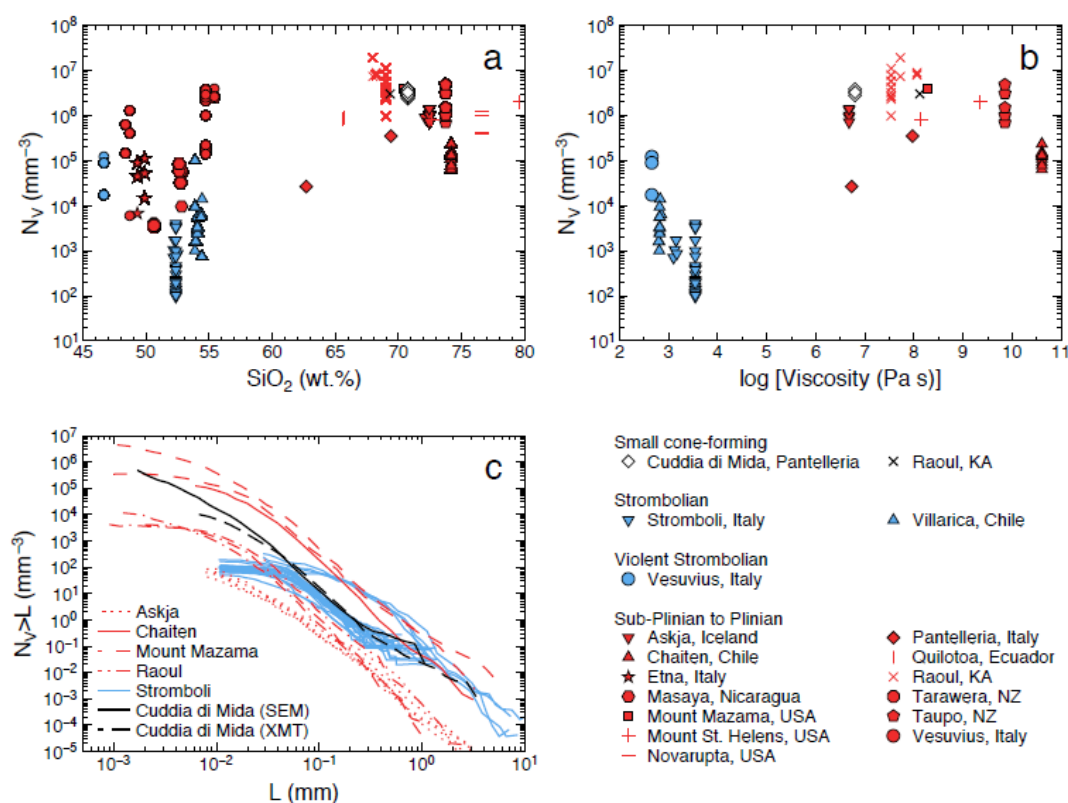


Figure 9 Vesicle size distribution (VSDs) for all vesicles (solid line) and vesicles next to crystals (average indicated by the dashed line and range indicated by the filled region) for A (red) and C (blue).



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Figure 10 a) Effect of melt composition (silica content) on total melt corrected vesicle number density ($N_{V,tot}^{melt}$) for various eruption styles; comparison of crystal-poor rhyolitic, basaltic Strombolian and peralkaline eruptions for b) $N_{V,tot}^{melt}$ variation with anhydrous melt viscosity; c) comparison of cumulative melt corrected VSD ($N_V^{melt} > L$). Viscosities calculated using Giordano et al. (2008), except in the case of Pantelleria where Di Genova et al. (2013) was used. *References:* Sigurdsson and Sparks (1981), Rutherford et al. (1985), Bacon and Druitt (1988), Klug and Cashman (1994), Sutton et al. (1995), Metrich et al. (2001), Klug et al. (2002), Landi et al. (2004), Lautze and Houghton (2005, 2007), Adams et al. (2006), Sable et al. (2006, 2009), Gurioli et al. (2008), Carey et al. (2009), Castro and Dingwell (2009), Polacci et al. (2009), Costantini et al. (2010), Houghton et al. (2010), Cioni et al. (2011), Rotella et al. (2014), Alfano et al. (2012), Barker et al. (2012), Neave et al. (2012), Leduc et al. (2015), Campagnola et al. (2016) and this study.



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Highlights

- First detailed vesicle textural study of a peralkaline magma.
- Combines 3D X-ray microtomography with high resolution 2D scanning electron microscopy.
- Peralkaline magmas fragment in a similar manner to calc-alkaline rhyolites.
- Viscosity does not appear to impact bubble nucleation and growth processes in peralkaline magmas.
- Bubble overpressure and strain localisation are important for peralkaline magma fragmentation.

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