

# Relating the physical properties of volcanic rocks to the characteristics of ash generated by experimental abrasion

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| 2  | Relating the physical properties of volcanic rocks to the characteristics of ash generated by                                      |
| 3  | experimental abrasion                                                                                                              |
| 4  |                                                                                                                                    |
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| 12 |                                                                                                                                    |

#### 13 Abstract

Interactions between clasts in pyroclastic density currents (PDCs) generate volcanic ash that can be dispersed to the atmosphere in co-PDC plumes, and due to its small size, is far-travelled. We designed a series of experiments to determine the effects of pyroclast vesicularity and crystal content on the efficiency and type of ash generated by abrasion. Two different pyroclastic materials were used: (1) basaltic-andesite pyroclasts from Fuego volcano (Guatemala) with ~26-46% vesicularity and high groundmass crystallinity and (2) tephri-phonolite Avellino pumice (Vesuvius, Italy) with ~55-75% vesicularity and low groundmass crystallinity.

21 When milled, both clast types produced bimodal grain size distributions with fine ash modes between 22 4 and  $5\varphi$  (32-63µm). Although the vesicular Avellino pumice typically generated more ash than the 23 denser Fuego pyroclasts, the ash-generating potential of a single pyroclast was independent of density, 24 and instead governed by heterogeneous crystal and vesicle textures. One consequence of these 25 heterogeneities was to cause the vesicular Avellino clasts to split in addition to abrading, which further 26 enhanced abrasion efficiency. The matrix characteristics also affected ash shape and componentry, 27 which will influence the elutriation and transport properties of ash in the atmosphere. The experimental abrasion successfully replicated some of the characteristics of natural co-PDC ash samples, as shown 28 29 by similarities in the Adherence Factor, which measures the proportion of attached matrix on 30 phenocrysts, of both the experimentally generated ash and natural co-PDC ash samples. Our results 31 support previous studies, which have shown that abrasion is an effective mechanism for generating fine 32 ash that is similar in size ( $\sim 5\varphi$ ; 30µm) to that found in co-PDC deposits. We further show that both the 33 abundance and nature (shape, density, components, size distribution) of those ash particles are strongly 34 controlled by the matrix properties of the abraded pyroclasts.

#### 35 Keywords

36 Volcanic ash, abrasion, milling, shape, vesicularity, crystallinity

37

38

#### 39 **1. Introduction**

Efficient fragmentation of bubbly melt into a dispersion of gas and pyroclasts can generate a large proportion of volcanic ash (Walker, 1981). Most commonly, the process is fuelled by magma overpressure, which generates the explosivity that transforms magma into pyroclasts, often with fractal size distributions (Rust and Cashman, 2011; Turcotte, 1986). Ash can also form, however, by 'secondary fragmentation' (Gonnermann, 2015) within the volcanic conduit (Dufek et al., 2012; Jones et al., 2016), in the thrust region of a volcanic plume (Cioni et al., 2014) or during transport in pyroclastic density currents (PDCs; Dufek and Manga, 2008; Eychenne et al., 2012).

47 Here we focus on secondary fragmentation in PDCs, where the energetic flow of gas and rock facilitates 48 abrasion and comminution between clasts, as evidenced by the abundance of rounded pumice clasts in 49 PDC deposits (e.g. Manga et al., 2011). These processes generate fine ash, which is important because 50 it affects PDC runout (Dufek and Manga, 2008) and can become buoyant and form a co-PDC plume 51 that disperses the fine fraction into the atmosphere (Engwell et al., 2016; Sparks et al., 1997). These 52 ash particles are hazardous for human health and aviation (Gudmundsson et al., 2012; Horwell, 2007), 53 and can add substantially to the fine ash fraction of the grain size distribution created by primary fragmentation; for this reason, it is important to anticipate the contribution of ash generated in PDCs to 54 55 the total ash budget produced by an eruption. Previous studies show that the amount of ash generated 56 by abrasion in PDCs will depend on a variety of factors such as the material in the PDC, the flow conditions and background eruptive behaviour (e.g., Gravina et al., 2004; Kueppers et al., 2012; Manga 57 58 et al., 2011). Here we analyse the effects of pyroclast vesicularity and crystal content on the efficiency 59 and type of ash generated by abrasion. We take an experimental approach, following previous studies 60 that imitate abrasion in PDCs (Kueppers et al., 2012; Manga et al., 2011; Mueller et al., 2015). We 61 extend these studies by using variably crystalline starting material and by quantifying the amount of ash 62 generated, as well as the ash size, componentry and shape, as these parameters influence the particle 63 settling behaviour and therefore both elutriation and transport potential in the atmosphere (Coltelli et 64 al., 2008).

#### 65 2. Background

66 Secondary fragmentation in PDCs has the potential to alter the total grain size distribution (TGSD) of 67 an eruption (Rose and Durant, 2009). Specifically, the small mean grain size of co-PDC ash (typically 68 <63 µm; Engwell and Eychenne, 2016; Marti et al., 2016) increases the power law exponent of the grain 69 size distribution (Jones et al., 2016). These small ash particles can travel long distances in the 70 atmosphere (Engwell et al., 2014), such that co-PDC ash is often the furthest travelled ash from large ignimbrite-forming eruptions (Engwell and Eychenne, 2016). Consequently, understanding ash 71 72 formation by secondary processes, such as abrasion in PDCs, is key for predicting the ash size 73 distribution and ash dispersion potential of future PDC-generating eruptions.

74 The typically smooth shapes of pumice clasts in PDCs, regardless of distance of transport, indicate that 75 clast rounding and, by inference, peak ash production, is rapid and takes place close to the vent (Manga 76 et al., 2011). Experiments have replicated the rounding of pumice clasts observed in PDC deposits and 77 the change in clast shape during abrasion has been related to the amount of ash generated in the flow 78 (Dufek and Manga, 2008; Manga et al., 2011). Importantly, these experiments found that the maximum 79 extent of clast rounding is not simply a function of the mass removed by abrasion. In fact, peak clast 80 rounding can occur when clasts have lost between 15% and 60% of their initial mass. The broad range 81 in the mass loss indicates that the peak ash production must be related, to some extent, to the properties 82 of the abrading material.

83 The effect of clast vesicularity on the efficiency of ash generation has also been examined 84 experimentally (Kueppers et al., 2012; Mueller et al., 2015), with low-density (vesicular) clasts 85 generating more ash than denser (less vesicular) clasts when subjected to the same duration of milling 86 in a rock tumbler. Here we suggest, however, that the differences in ash production are controlled not 87 only by differences in vesicularity, but also by the crystal content (size, shape and abundance of phenocrysts and microlites). As PDC components from different volcanoes are extremely varied in 88 89 both vesicularity and crystal content, both properties must be assessed to anticipate the ash-generating 90 potential of a specific PDC.

Experiments have shown that the shape of ash is also altered by abrasion (Jones et al., 2016). In an
experimental study using a ball mill, Jones et al. (2016) found that increased milling duration produced

93 ash particles with higher axial ratios (more equant shapes), which indicates the progressive rounding of 94 ash grains, as well as increased removal of matrix from phenocrysts and phenocryst fragments (Freundt 95 and Schmincke, 1992; Jones et al., 2016). From this perspective, it is possible that the shape and internal 96 texture of ash generated by abrasion could be used to distinguish it from ash generated by the primary 97 fragmentation of the magma. Field studies of the Latera Volcanic Complex, Italy, have shown that 98 sanidine crystals in the PDC deposit have fewer boundary irregularities than crystals of the same grain size in the fall deposit (Taddeucci and Palladino, 2002). Similarly, a preliminary assessment by Liu et 99 100 al. (2015a) of Tambora ash suggests that co-PDC ash particles have grain boundaries with reduced 101 roughness compared to fall-deposit ash of the same grain size.

102 The phenocryst content should also affect the efficiency of ash generation by abrasion, as crystals can 103 influence fracture and breakage patterns in pyroclasts (Wawersik and Fairhurst, 1970). However, the 104 effect of phenocrysts is not clear. Crystals may either resist erosional processes such as abrasion in mass 105 flows, or conversely, may introduce planes of weakness, especially if they have platy textures or have 106 been fractured by the initial brittle fragmentation (Bindeman, 2005; Thomson et al., 2013). For 107 instance, experiments show that crystal-rich pyroclasts from Mount Unzen are more resistant to 108 abrasion than less crystalline samples (Kueppers et al., 2012). Similarly, crystal-bearing Medicine Lake 109 pumice abraded more slowly than crystal-free, texturally homogenous Taupo pumice of similar density 110 (Manga et al., 2011). What still is unclear, however, is the effect of a heterogenous distribution of 111 phenocrysts and microlites, such as localised dense zones within a pumice clast, on the characteristics 112 of ash generated by abrasion.

Also unexplored is the effect of groundmass crystallinity (i.e. microlite content) on the resistance of pyroclasts to abrasion. Hydrous mafic (basalt, basaltic andesite) magmas, in particular, experience extensive groundmass crystallization during rapid magma ascent in the conduit (e.g., Shea and Hammer, 2013). The products of these eruptions also have total grain size distributions (TGSDs) with less fine ash than their microlite-free counterparts (Durant and Rose, 2009; Jones et al., 2016; Rust and Cashman, 2011; Turcotte, 1986), which suggests that microlites affect primary fragmentation. We expect the role of groundmass crystallinity in secondary fragmentation processes to be four-fold. First, microlite-rich samples also have low vesicularities, which would suggest that they should resist abrasion. Second, these samples have thick bubble walls, so that even if they abrade, the ash particles should be larger than in low crystallinity, high vesicularity samples. Third, analogue and numerical experiments (Oppenheimer et al., 2015; Parmigiani et al., 2016, 2011) show that groundmass crystals aid formation of gas pathways, which could control patterns of breakage. Finally, the crystal content of the resultant ash influences ash shape, as crystalline ash is typically blocky and lacks the perimeter concavities caused by abundant vesicles (Liu et al., 2015).

127 In this study, we test the efficiency of secondary ash generation by abrasion as a function of both 128 vesicularity and crystallinity (phenocrysts and groundmass) of starting pyroclasts. We use samples 129 from a violent Strombolian eruption of basaltic andesite Volcan Fuego, Guatemala, and from the Plinian Avellino eruption of tephra-phonolite from Vesuvius volcano, Italy. Informed by previous studies 130 131 (Kueppers et al., 2012; Manga et al., 2011) we expected that the low density (high vesicularity) Avellino 132 pumice would generate more ash than the high density (low vesicularity) Fuego pyroclasts. To test this hypothesis, we characterised the size, shape, vesicularity and crystallinity of the products both prior to 133 134 and following abrasion experiments to dissect the links between the crystallinity, ash generation and 135 ash shape. We then compare experimental and natural ash particles to assess the reliability of ash shape 136 as an indicator of fragmentation process.

137 2.1

## 2.1 Volcan de Fuego (2012 eruption)

138 Volcan de Fuego is a stratovolcano located along the Central American volcanic arc in Guatemala. It 139 produces frequent discrete violent Strombolian blasts with small ash plumes (<10km) and lava flows. 140 Occasional 'paroxysms' of heightened activity generate Vulcanian to sub Plinian eruption columns and 141 PDCs (Rose et al., 2008). The material used in our experiments is from a paroxysmal eruption in 142 September 2012, which was more explosive than typical Strombolian activity (Chigna et al., 2012). 143 The eruption produced a block and ash flow that we sampled ~6km from the vent on a field campaign in March 2016. The deposit is massive, poorly sorted and contains large blocks of basaltic andesite. 144 145 We assume that the material is geochemically similar to basaltic-andesite explosive products from the 146 1974 eruption of Fuego (Chesner and Rose, 1984) as no geochemical analyses have been carried out on 147 the 2012 eruptive products. This assumption is supported by the similarity of the phase assemblage to 148 the 1974 material, and the lack of compositional differences between magma erupted in 1974, 1999 and 149 2003 (Berlo et al., 2012). Major element analyses of the 1974 pyroclasts show an average silica content 150 of 56.5 wt% and a total alkali content (Na<sub>2</sub>O + K<sub>2</sub>O) of ~5.6 wt% (Chesner and Rose, 1984).

Additional material from Volcan de Fuego used in this study includes 2012 ash that was sampled from the top of a small block and ash flow. The ash represents the dilute portion of the PDC that settled out after the emplacement of the dense basal layer and is termed "PDC-surge" ash henceforth. The  $3\varphi$ (177-125µm) fraction of this PDC-surge ash was imaged by E. Liu (*personal comm*). As a reference, we also analyse fall deposit ash generated during a paroxysm of Fuego in 2016 (sampled by E. Liu).

# 156 2.2 Pomici de Avellino (3.9ka B.P.)

157 An eruption of Somma-Vesuvius, Italy in 3.9ka B.P produced the Pomici de Avellino (PdA) deposit 158 (Sulpizio et al., 2008). It was a large explosive eruption characterised by three distinct eruptive phases: 159 an opening Plinian phase followed by a column collapse phase and a final phreatomagmatic phase 160 (Sulpizio et al., 2010a). The stratigraphy is split into eruption units EU1 to 5 (Cioni et al., 1999). The 161 products used in our experiments are from a PDC from EU3 that was generated during the column 162 collapse phase. The PDC from this phase (EU3pf) was directed to the north, had a short runout (<10km) 163 compared to the later EU5 PDCs (>20km, Sulpizio et al., 2010b) and is described as 'poorly sorted 164 massive to stratified lapilli and ash' (Sulpizio et al., 2010b). Analyses of Avellino EU3 glasses by 165 Sulpizio et al. (2010b) yield an average silica content of 55.6 wt% and a total alkali content of 12.8 166 wt%, classifying the products as tephri-phonolite.

No distinct co-PDC or surge ash layer is found in the Avellino EU3 deposit (Sulpizio et al., 2010a).
Instead, ash from the matrix of the dense part of the EU3pf deposit was sampled to compare to our
experimentally generated ash and to examine the results of natural abrasion during transport in the flow.

170

#### 171 **3. Methods**

#### 172 **3.1.** Characterisation of starting materials

173 3.1.1. Density

174 Juvenile clast densities were measured using the methodology of Houghton and Wilson (1989). Removable paraffin film of known density was used to seal the clasts for the density measurements 175 176 which enabled clasts of known density to be later used in the abrasion experiments. The density 177 distributions of the Fuego and Avellino clasts in the  $-3\varphi$  (8-16mm) size range that we used as starting material are shown in Figure 1. The mean densities of the Fuego and Avellino pyroclasts are 1.92 gcm<sup>-</sup> 178 <sup>3</sup> and 0.97 gcm<sup>-3</sup>, respectively. Each sample was divided into three sub-samples for the abrasion 179 180 experiments (Fig. 1): one with a density close to the average of the bulk juveniles (AD), one with lower 181 than mean density (LD) and the third with a higher than mean density (HD). This was done to test 182 whether LD clasts (high vesicularity) generate more ash than the HD (low vesicularity) clasts. To track the change in shape of the large clasts (e.g., Manga et al., 2011), the clasts in each sub-sample were 183 placed on a light board in order of ascending density and photographed before and after each abrasion 184 185 experiment.

Dense rock equivalent (DRE) densities are required for calculating vesicularity. The Avellino DRE
density used was 2.42 gcm<sup>-3</sup> (Balcone-Boissard et al., 2012). The DRE density for Fuego was
approximated from a dense juvenile, measured at 2.79 gcm<sup>-3</sup> (Houghton and Wilson, 1989).

189 3.1.2. Vesicularity

190 The vesicularity ' $V_D$ ' was calculated using the DRE densities of Avellino and Fuego lavas and the 191 measured juvenile densities (e.g., Houghton and Wilson, 1989; Fig. 1). The average  $V_D$  calculated for 192 Fuego was 31.2% and 59.6% for Avellino.

193 We also measured the vesicularity from polished thin sections of juvenile clasts  $(-3\varphi)$ , as imaged using 194 a Hitachi S-3500N Scanning Electron Microscope (SEM) at the University of Bristol in backscattered

195 electron (BSE) mode. Imaging the clasts at several magnifications ensured the optimum resolution for 196 subsequent image analysis of both vesicles and crystals (Shea et al., 2010). Vesicularity and bubble 197 number density (BND) were calculated from BSE images using FOAMS, a MATLAB<sup>TM</sup> based program 198 (Shea et al., 2010) that converts 2D BSE images into a 3D representation of the vesicularity, ' $V_{\rm F}$ '. The 199 average vesicularity of four Fuego juvenile clasts was 32.4%, which is similar to the average measured 200  $V_{\rm D}$ . It was not possible to determine an accurate  $V_{\rm F}$  for the EU3pf pumice using FOAMs because of the 201 heterogeneity of the vesicle population. The measured  $V_F$  of pumice from the EU3 fall deposit is 56.1% 202 (Voarino, 2016), which is similar to the V<sub>D</sub> value of 59.6%. This, together with the close agreement of 203  $V_{\rm D}$  and  $V_{\rm F}$  for Fuego, gives us confidence in the robustness of the density measurements.

204 3.1.3. Crystal Content

The phenocryst content of the starting materials was quantified using the BSE images acquired for vesicularity. Additional high magnification (1500x) BSE images were used to quantify the microlite content of the samples. Crystal identities were checked by energy dispersive spectroscopy (EDS) spot analyses on the SEM, after which the BSE grey scale value of each phase was used to map the phase distribution. The abundance of each phenocryst phase and the ratio of microlites to glass in the groundmass on a vesicle-free basis were determined with ImageJ.

# 211 **3.2 Abrasion Experiments**

212 The abrasion experiments were carried out using a planetary ball mill manufactured by Retsch Ltd. The 213 Retsch Ltd PM 100 model has a vertical axis of rotation. This differs from studies in which a rock 214 tumbler was used (Kueppers et al., 2012; Manga et al., 2011), where the rotation axis was horizontal. 215 Typically, the Retsch planetary ball mill is used to generate very fine powders due to the milling 216 efficiency that results from high centrifugal forces generated at high milling speeds (Mio et al., 2002). 217 We did not want complete pulverisation of the samples, therefore short milling times were used (4-5 218 mins). Additionally, the agate milling balls that encourage high energy impacts across the milling jar 219 (Lu et al., 2012) were excluded, which means that the milling produces only clast-to-clast and clast-to-220 jar-wall interactions. We assume that friction at the jar walls is minimal due to the small mass of a single clast and the smooth nature of the agate lined jar (Kueppers et al., 2012) and that the abrasion isdriven mainly by clast interactions (both frictional sliding and impacts).

Sub-samples with an initial grain size of  $-3\varphi$  (8-16mm) were used in the abrasion experiments. This ensured that all post-milling material smaller than the starting grain size was generated in the experiment. The number of clasts in each experiment was limited by the size of the grinding jar and the availability of material in the  $-3\varphi$  fraction (Table 1). The grinding jar was sealed, placed on the rotating base and milled for 4 minutes with a rotation rate of 450 rpm (Fig. 2). Two additional Avellino abrasion experiments were carried out to investigate the effect of varying the starting grain size and the addition of a lithic component (sample Av31b and Av3AG, respectively; Table 1).

Our aim is to simulate processes occurring as clasts are transported in a PDC. The equivalent transport
distance (x) of the milling,

$$x = l \cdot t \cdot v, \tag{1}$$

is ~425m for our experiments, with *l* being the inner milling jar circumference, *t* is the milling time in mins and *v* is the rotation rate in rpm (Kueppers et al., 2012). The speed at the wall of the rotating jar (= x/t) was ~6.5km/hr which is an order of magnitude lower than typical PDC ground velocities (e.g., Cole et al., 2002). However, the velocities of the clasts relative to each other during milling, are much higher due to the planetary ball mill configuration (>20km/h, Lu et al., 2012) and more comparable to the internal velocities in a natural PDC.

### **3.3 Characterisation of experimental products**

#### 240 3.3.1. Grain size distributions (GSDs)

The milled samples were manually dry-sieved from -3 to 3 phi ( $\varphi$ ), with half phi sieving performed where possible. The size distribution of the fine material (>2 $\varphi$ , <250 $\mu$ m) was quantified using a Mastersizer 3000<sup>TM</sup> by Malvern Instruments Ltd in the School of Geographical Sciences at the 244 University of Bristol. The fine and coarse fractions were combined to produce a GSD for each 245 experiment using the overlap in the  $3\varphi$  size fraction (e.g., Jones et al., 2016).

246

# 3.3.2. Sample preparation and analysis conditions

After milling, grains from each size fraction were mounted and polished separately to ensure that all 247 ash particles could be imaged at the same magnification (Liu et al., 2015a). When imaging the ash 248 249 mounts, the objective was to balance the resolution of the images and the number of grains imaged. 250 Typically, we found that at a working distance of  $\sim 15$  mm, an accelerating voltage of 15 or 20kV and 251 200x magnification gave an optimum resolution ( $\sim 1560$  pixels/mm). Automated grid images, typically 252 made up of 20-60 individual images, were stitched together using the ImageJ stitching plugin. The 253 collated images contain 400-1000 grains with 30,000-60,000 pixels per grain. The ash componentry 254 was then assessed from the BSE grid images with grains classified as free crystals, crystals with attached 255 melt or matrix, groundmass grains, vesicular glass or pure glass.

#### 256 3.3.3. Post-abrasion analysis of large clasts

The largest clasts (-3 and -2 $\varphi$ ) remaining after the abrasion experiments were photographed and their size and shape compared with the original clasts of the same sub-sample. It was not possible to match each individual clast pre- and post-abrasion, therefore we amassed the shape data for all the sub-samples of each starting material and compared the average change in shape. We used the metric form factor 'FF' (Liu et al., 2015; termed 'R' in Manga et al., 2011) to measure shape, where FF = 1 represents a perfect circle (Table 2):

263

$$FF = 4\pi^* area of particle/ (perimeter of particle)^2$$
. (2)

The results of the shape analysis, particularly whether the FF increased (rounded) or decreased, informed our choice of which remaining clasts to mount for vesicularity and crystallinity characterization. As both starting materials were sampled from PDC deposits, the clasts had already experienced some degree of natural abrasion. The change in FF of the larger clasts from pre- to postabrasion could therefore represent the effect of further transport in a natural flow.

# 269 3.3.4. Ash shape analysis

270 The BSE grid images gathered for componentry analysis were also used to measure particle shape. 271 Images were first cleaned and converted to a binary form, then ash shape was measured using an ImageJ 272 macro from Liu et al. (2015a). The natural ash samples from Fuego and Avellino (Table 1) were sieved, 273 the  $3\varphi$  fraction imaged, and the cross-sectional shape measured using the same approach used for the 274 experimentally generated ash. For comparison purposes, the resolution of the BSE images of the experimental ash was reduced to match the Fuego PDC-surge ash images (~390 pixels/mm, E. Liu, 275 276 personal comm.), as convexity is sensitive to perimeter resolution. This was done by reducing the image 277 size in ImageJ, which resamples the original image using bilinear interpolation (Ferreira and Rasband, 278 2012). The image analysis and shape measurements were then repeated using the lower resolution 279 images. Table 2 outlines the key shape parameters used in this study. Grains classified as 'crystals with 280 attached matrix' in the componentry analysis were further analysed to determine the 'Adherence Factor' 281 (AF; Jones et al., 2016):

282 
$$AF = (area of matrix) / (area of matrix + area of crystal).$$
 (3)

Values of AF close to 1 reflect a large proportion of attached matrix. This measurement required
further manipulation of the BSE grid images to enhance the grey scale contrast between attached
matrix and crystal.

286 The projected area of the experimental ash particles was measured using Morphologi G3SE<sup>TM</sup> by 287 Malvern Instruments Ltd, an Optical Particle Analyser (OPA) located in the Laboratoire Magmas et 288 Volcans, Clermont-Ferrand, France. Morphologi homogeneously disperses dry particles onto a glass 289 plate using compressed air. The ash is then imaged and basic size and shape parameters of individual 290 grains are measured using the same metrics in Table 2 (Leibrandt and Le Pennec, 2015). We analysed 291 the 3 $\varphi$  fraction of Fuego sample F3a and Avellino sample Av31a (Table 1) to compare with the results 292 of the 2D shape analysis. The  $4\varphi$  (74-63 µm) fraction of these samples was also analysed to compare 293 the shape measurements made across the two grain size fractions. The  $4\phi$  fractions were sieved by quarter phi increments, cleaned with ethanol and dried prior to analysis to ensure that very fine materialwas removed.

For each starting material and grain size, a standard operating procedure (SOP) was designed. The light source (diascopic or episcopic), magnification, focus, threshold and the area of the dispersion to scan were set manually (Leibrandt and Le Pennec, 2015). We concluded that the best resolution was provided by using a 5x optic with diascopic light (light from below) to obtain the 2D projected area of the ash shape. Once the SOP was defined, each analysis took 30-40 minutes depending on the scanning area.

Following the automated analysis, the Morphologi software (Malvern) generates an output file of the shape and grain size data. The  $3\varphi$  size fraction produced grain counts of 3000-7000, and the  $4\varphi$  analyses contained over 10 000 grains per sample. The output files required cleaning due to the imperfect nature of the imaging and automated dispersion (see Fig. S1). Grain counts in the  $4\varphi$  fraction were reduced to a sub-sample of 6000 randomly selected grains before the anomalous grains were removed. This ensured the same degree of manual error in the datasets of each grain size fraction.

308 <u>4. Results</u>

# 309 4.1. Sample characterisation

On a vesicle-free basis, Fuego pyroclasts have ~35% phenocrysts, but the phenocryst content ranges from 41% in a HD clast to 30% in a LD clast. The phenocrysts are dominantly plagioclase and olivine with sizes of 0.3-4mm. The average microlite content is ~40% in the groundmass (on vesicle-free basis) and comprises plagioclase (dominant), clinopyroxene, oxides and olivine. There is minor variability, but no significant difference in the microlite crystallinity between the high- and low-density subsamples.

The vesicle size distribution (VSD) varies with density in the Fuego pyroclastic starting material. We prepared and imaged LD, AD (average density) and HD Fuego pyroclasts (Fig. 3) to analyse with FOAMS (Shea et al., 2010). There is a general trend of larger vesicle sizes and thicker bubble walls with increasing density. The modal vesicle size of the HD clasts (Table 1) is 1-0 $\varphi$  (0.5-1mm), with no vesicles  $<5\varphi$  (0.031mm) (Fig. 3d, h). LD clasts have a substantial fraction of bubbles (by area and by number)  $<2\varphi$ mm (0.25mm) and a modal vesicle size between 3-2 $\varphi$  (Fig. 3a,b). Some LD clasts have a small number of large vesicles ( $>0\varphi$ ) which account for a large area fraction (Fig. 3b, f). These clasts with large vesicles (Fig. 3b) also have more glomerocrysts and higher groundmass crystallinity than other LD clasts (Fig. 3a). From examining a small number of LD clasts (6), it appears the two types of LD textures (Fig. 3e, f) occur in equal proportion in the Fuego pyroclasts.

326 The average (vesicle-free) phenocryst crystallinity of the Avellino pyroclasts is  $\sim 12\%$ , ranging from 327 8% in an LD pumice clast to a maximum of 20% in an HD pumice clast. The phenocrysts in the Avellino pumices are 0.2-1.5mm in size and dominantly sanidine and nepheline with minor amphibole, 328 329 vesuvianite and clinopyroxene. The microlite content on a phenocryst and vesicle-free basis is 7-13%, 330 and comprises sanidine, nepheline and minor skeletal leucites (Voarino, 2016). Spatial variations in 331 microlite content include microlite-rich areas that are most frequent in the HD clasts (Fig. 4a and b). 332 There are also fragments of thermally recrystallised limestone embedded in the pumice associated with 333 large vesicles, which likely result from volatile release due to thermal decomposition of carbonate 334 minerals (Fig. 4c, e.g. Jolis et al., 2015; Sottili et al., 2010). Leucite microlites and the dense microlite-335 rich areas appear to be unique to the dense mode of the EU3pf deposit (Voarino, 2016).

## **4.2. Grain size distributions**

337 All post-abrasion grain size distributions (GSDs) are bimodal, with most of the resultant  $ash \le 125 \mu m$ 338  $(\geq 3\varphi)$ ; Fig. 5). Details of the GSDs differ, however, in that the fine mode lies at  $4\varphi$  (63-90µm) in the 339 Fuego experiments and  $5\varphi$  (32-45µm) in the Avellino experiments. Moreover, the Avellino clasts 340 generally produce more ash than the Fuego clasts. Cumulative grainsize distributions for all experiments 341 are shown in Figure 6, where the y-axis represents the starting grain size (- $3\varphi$ ). These distribution 342 profiles highlight another consistent difference between Fuego and Avellino samples. The relatively 343 flat profiles of the Fuego GSDs across the intermediate grain size fractions (-1 to  $3\varphi$ ) shows negligible formation of ash of intermediate size. In contrast, the Avellino GSDs do not have flat profiles between 344 345 -1 and 3 $\varphi$ , meaning that milling generated particles across a wide range of grain sizes.

346 Overall, the Fuego milling experiments generated less ash than the Avellino experiments. In detail however, the response of individual Fuego pyroclasts to abrasion varies. The expected result was that 347 348 the high density (low  $V_D$ ) sub-samples would generate the least ash and the low density (high  $V_D$ ) the most. For example, experiment F3a behaved as expected: >60% of the material generated in the LD 349 350 experiment was smaller than  $-3\varphi$ , whereas <16% of the material in the HD experiment was smaller than 351 -3 $\varphi$ . However, most experiments did not show this behaviour: there are other Fuego experiments where LD sub-samples (highest V<sub>D</sub>), do not generate the highest proportion of ash (e.g., F2a; Fig. 5c). 352 353 Similarly, the HD sub-samples, which contain clasts with high crystal contents (both phenocrysts and microlites) and low V<sub>D</sub>, are not always the most resistant to abrasion. 354

355 Crystal-poor, high vesicularity Avellino experiments consistently have a smaller proportion of the 356 material remaining in the  $-3\varphi$  fraction than the experiments with crystal-rich, vesicle poor Fuego 357 material. This indicates more efficient abrasion and ash generation for pyroclasts with low crystallinity 358 and high vesicularity. We still see variability in the amount of ash generated, however. In one extreme, 359 LD sub-sample Av31a yields <10% of mass remaining in the starting grain size, while milling of the 360 AD (average density) sub-sample generated less ash (>40% of the material remaining in the starting 361 size) than the HD sub-sample (Fig. 5d).

362 **4.3. Abrasion products** 

#### 363 4.3.1. Shapes of large clasts

364 The median form factor (FF) of all large (-3 and  $-2\varphi$ ) clasts increased after milling (Fig. 7), reflecting 365 the expected clast rounding due to abrasion (Manga et al., 2011). Although the median FF values 366 increase, post-abrasion clast shapes are variable. At the same time, some experiments show an increase 367 in the number of grains in the -3 and - $2\varphi$  grainsize fractions (see Table 1; Fig. 7). For example, milling 368 of Av33a HD caused the number of large clasts to increase from 7 to 9. Both the range in FF values and 369 the increase in particle number can be explained by splitting, rather than solely abrasion, of clasts. 370 Splitting of clasts also explains the low (outlier) FF measurements in the Avellino data (Fig. 7) that 371 indicate an increase in clast angularity.

372 4.3.2. Projected area ash analysis from Morphologi

373 Ash shape measurements of the  $3\varphi$  AD sub-samples are shown in convexity (CVX) - solidity (SLD)-374 plots in Figure 8a, b (see Fig. S2 for  $4\varphi$  shape data). The shape data based on Morphologi images are 375 concentrated at high-CVX and high-SLD values, as illustrated by the 20% contour of Fuego data, which plots between SLD values of 0.94-0.98 and CVX values of 0.95-0.98 for all the sub-samples. The same 376 377 contour of the Avellino shape data plots between SLD and CVX values of ~0.95-0.98. In the extreme, however, the Avellino contour plots show a somewhat greater range, particularly in convexity. For 378 379 example, the Avellino 90% contour extends to convexities of  $\sim 0.87$ , whereas the Fuego 90% contour 380 does not reach below CVX = 0.90.

381 Ash componentry was also evaluated using the Morphologi images, with categories designated as free 382 crystal, vesicular, opaque (likely matrix dominated), crystal with attached melt or, in some cases, 383 'unknown'. The categorisation was done on a random selection of 500 grains per sample. The 384 convexity and solidity values of these 500 grains are plotted in Figure 8c, d. The Fuego ash comprises 385 mostly free crystals and opaque grains. The free crystals (red diamonds) typically record the highest 386 SLD and CVX; opaque grains have a broader range in SLD and CVX. In the Avellino samples, vesicular grains (green asterisks) are numerous and plot with lower SLD and CVX values than the rest 387 388 of the grains, reflecting the irregular outlines of these particles.

#### 389

## 4.3.3. Cross-sectional area (BSE) ash analysis

390 BSE images of polished 2D sections provide a more nuanced measure of particle shape as well as details 391 of the internal textures of individual grains (Liu et al., 2015a). Again, we plot CVX vs. SLD of 3 $\phi$  grains 392 from post-milling samples F3a and Av31a (Fig. 9). As anticipated, SLD and CVX measured on 2D 393 images of cross-sections are much lower and more varied than the projected area (Morphologi) results. 394 Note the more limited number of grains analysed (lower n values) compared to Figure 8 are because 395 sample preparation and BSE image acquisition are more time consuming than Morphologi analyses. 396 The Fuego ash ranges from 0.39-0.96 SLD and 0.50-0.95 CVX. The Avellino ash covers an even wider 397 spectrum, with SLD ranging from 0.22 to 0.97 and CVX from 0.21-0.97.

Componentry analysis of the same particles (Fig. 10, Fig. S3) helps to explain the shape parameter range. Fuego ash is dominated by variably crystalline matrix fragments, with >50% of the grains being groundmass grains that have CVX and SLD values of  $\sim 0.75 - 0.9$ . Conversely, Avellino ash comprises predominantly free crystals and vesicular grains, the former having very high CVX and SLD values, and the latter marking the low CVX and SLD extremes.

403 The differences between the shape parameters measured by ImageJ and Morphologi reflect the 404 differences between cross-sectional area and projected area (Liu et al., 2015a). Lower and more varied 405 values of convexity and solidity are measured using BSE images and ImageJ due to the ability to 406 measure small-scale roughness. The most extreme ash shapes measured by both methods (lowest values 407 of CVX and SLD) are the vesicular grains that characterize the microlite-poor Avellino samples. The free crystals, in contrast, yield the highest SLD and CVX values (>0.95 in Fig. 8 and >0.8 in Fig. 9) 408 409 using both methods, and thus provide an approximate measure of the free crystal content of individual 410 ash samples.

# 411 4.3.4 Natural ash samples

Two natural Fuego ash samples were analysed for their shape (Fig. 11a, b). The Fuego 2016 fall ash contains more grains with lower SLD and CVX values than the experimental or natural PDC-surge ash (Fig. 11). Similar to the experimental Fuego ash, the natural fall and surge ash show a range in microlite textures with an average microlite content of ~41%, comparable to the crystallinity measured from Fuego pyroclasts used in the experiments.

The CVX-SLD density plot of the natural PDC-surge and experimental Avellino ash (Fig. 11c) shows a slight increase in the number of low SLD and CVX values in the experimental ash compared to the natural ash. An important difference between the two samples is the presence of holocrystalline ash grains in the natural ash, which comprise ~29% of the components (Fig. 11f). They are the remnants of lithics that were incorporated into the Avellino PDC.

422 4.3.5 Adherence Factor

423 Experimental ash from both Fuego and Avellino contained numerous ash grains that were classified as 424 'crystals with attached matrix' (Fig. 10). We quantified the amount of matrix attached to each imaged 425 crystal in the 3 $\varphi$  size class using the 'Adherence Factor' (AF; Eq. 3). As shown in Figure 12, the 426 Avellino and Fuego samples show contrasting AF distributions. Avellino ash samples yield the lowest AF values, whereas the Fuego samples, show a wide range of AF values. For comparison, we also 427 428 include the results of milling experiments on Soufriere Hills Volcano (SHV) pumice (Jones et al., 2016) 429 which has intermediate values of vesicularity and crystallinity, and correspondingly intermediate AF 430 trends.

# 431 <u>5. Discussion</u>

432 The abrasion experiments show that, for the same duration of milling, the Avellino pumice generated 433 more ash than the denser Fuego juvenile material (Figs. 5 and 6). This supports the results of Kueppers 434 et al. (2012), who found that material of lower density and higher vesicularity generates more ash when 435 abraded. However, the results are less straightforward when the starting material is examined in the 436 context of density-designated sub-samples from the same eruption. This means that density alone 437 cannot be used to predict the behaviour of material subjected to abrasion. Moreover, our experiments show that ash shape, size and componentry vary both within and between samples. As these parameters 438 439 determine the transport properties of secondary ash particles, understanding controls on these properties 440 is critical for predicting the contributions of co-PDC ash to distal ash clouds.

# 441 **5.1 Effects of pyroclast textural heterogeneity on abrasion**

#### 442 5.1.1 Fuego pyroclasts

The vesicle size distribution (VSD) of pyroclasts influences the TGSD generated by primary fragmentation (Liu et al., 2015b; Rust and Cashman, 2011). The VSD of pyroclasts may also affect the extent of secondary fragmentation because vesicles affect brittle rock strength (Heap et al., 2014) and form zones of weakness that may control patterns of breakage. Sub-samples in the Fuego experiments, in particular, display a wide range in the amount of ash generated, which likely reflects VSD variations in the Fuego starting material (Fig. 3). If clasts abrade by fracturing at the weakest point, which typically will be the thinnest part of a vesicle wall, then the low vesicle number density and increased thickness of the bubble walls in the HD samples explains the resistance to abrasion of the HD clasts. The average bubble wall thickness may also control the size of the abraded ash particles (typically  $4\varphi$ , or ~63 µm, in Fuego LD samples).

453 Not all Fuego LD clasts generated a large proportion of ash when abraded (Fig. 5c), however. To 454 address this discrepancy, we imaged two post-milling LD clasts of similar density: one that recorded a 455 25% decrease in projected area with visible rounding (Fig. 3a) and one that recorded only a 2% decrease 456 in projected area and showed a decrease in FF post-milling, rather than the expected increase (Fig. 3b). 457 The clast that did not round (Fig. 3b) has a smaller modal vesicle size, elevated microlite crystallinity 458 and a greater number of phenocrysts, glomerocrysts and large vesicles compared to the LD clast that 459 did round (Fig. 3a). Here the few large vesicles (~0.5mm; Fig. 3b) allow the non-rounded clast to retain 460 a low density, whilst the high crystallinity (both glomerocrysts and microlites) controls its resistance to 461 abrasion. It seems likely that the relative proportions of these two clast types in the LD sub-samples 462 explains the range in ash production rates. The importance of heterogeneities in ash generation means 463 that our individual experiments with a small number of clasts are not representative; however, the 464 limited set of clasts allows us to explore the differences in sub-samples by isolating individual clasts, 465 monitoring their behaviour post-milling and attributing differences in behaviour to differences in 466 vesicularity and crystal content.

# 467 5.1.2 Avellino pyroclasts

The Avellino experiments consistently generated a higher proportion of ash than the experiments with denser Fuego pyroclasts. Interestingly, however, the HD sub-samples generated more ash than the AD sub-samples in two of the Avellino experiments, contrary to the expectation that high density (low vesicularity) clasts would be most resistant to abrasion (Kueppers et al., 2012). Sectioning of selected clasts shows that the EU3pf pumice is highly heterogeneous in both vesicularity and crystal content, particularly in the HD clasts. For example, HD clasts can contain microlite-rich zones that have a high number density of microlites and low vesicularity relative to the surrounding melt (Fig. 4a, b; Voarino, 475 2016), textural features that should make these zones relatively resistant to abrasion, leading us to476 further question the high proportion of ash generated by two of the HD sub-samples.

The  $-2\varphi$  fraction of the EU3pf deposit has a bimodal density distribution, unlike the  $-3\varphi$  fraction, with 477 a secondary dense mode created by clasts with relatively high microlite content (Voarino, 2016). To 478 479 test the response to abrasion of microlite-rich clasts, we carried out an additional abrasion experiment 480 using the dense Avellino pumices from the -2\phi fraction (sample Av31b in Table 1). The resulting GSD 481 (Fig. 6c) shows that the clasts were less abraded than the  $-3\varphi$  clasts even when milled for an extra 482 minute, confirming that the increased microlite content of the  $-2\varphi$  strongly enhances their resistance to 483 abrasion. However, the extremely microlite-rich zones (Fig. 4), were only present in two out of five HD clasts imaged post-abrasion. Therefore, rather than limiting abrasion, the HD clasts which have 484 485 elevated microlite content or contain dense zones could increase abrasion by acting as resistant grains 486 that abrade the more vesicular clasts. This is analogous to the scenario where a lithic component (i.e. 487 more resistant lithology) incorporated in PDCs encourages efficient abrasion (Bernard and Le Pennec, 488 2016). This may be one explanation for the unexpected behaviour of some HD subsamples (Fig. 5,6).

Additional heterogeneities in Avellino clasts include xenoliths of altered limestone, which generate cavities in the surrounding melt by decarbonation (Fig. 4c). These cavities introduce internal weaknesses that may promote clast splitting rather than abrasion (see section 5.2).

The heterogeneities observed within the Fuego and Avellino pyroclasts are typical of the textural heterogeneity found in natural PDC material (e.g. Bernard and Le Pennec, 2016; Gardner et al., 1998). Importantly we have shown that textural heterogeneity within a density class from a single eruption can influence pyroclast resistance to abrasion. The demonstrated sensitivity of clast abrasion to internal heterogeneities means that density alone cannot be used to predict the efficiency of ash production by abrasion within a PDC.

# 498 **5.2 Enhanced ash production due to clast splitting**

499 The increase in the post-milling clast count in the  $-2\varphi$  and  $-3\varphi$  fractions (Table 1) indicates that some 500 clasts split during the abrasion experiments. Clast splitting can facilitate further abrasion, as it increases

501 the total surface area and number of sharp corners that control abrasion (Manga et al., 2011). The 502 Avellino experiments showed the most evidence for splitting during milling, with the clast number 503 almost doubling (from 7 to 12 clasts) in one sub-sample (Av32a AD sample; Table 1). In these 504 experiments, the large proportion of material in the  $-2\varphi$  grain size (expected for splitting of  $-3\varphi$  grains), 505 as well as the abundance of ash in the intermediate grainsizes ( $-1\phi$  to  $2\phi$ ; Fig. 5), suggest that splitting 506 was a dominant, rather than occasional, process. The post-abrasion range in FF values and the 507 frequency of outlying FF values (Fig. 7) also provide evidence of splitting. The HD sub-sample of 508 Av33a provides a particularly striking example of the hypothesis that splitting of Avellino clasts aids 509 efficient ash generation. Here the largest clast completely disintegrated (extensive splitting), which 510 increased ash generation across the intermediate  $(-1\phi \text{ to } 2\phi)$  grain sizes compared to the other Avellino HD subsamples (Fig. 6b). 511

512 Our data further suggest that internal weaknesses that encourage splitting (such as large vesicles or 513 cavities formed by decarbonation) are exploited rapidly, so that over time, splitting becomes far less 514 common. This means that after long milling times (as in Manga et al., 2011), rounding of all fragments 515 makes the products of splitting indistinguishable from clasts that experienced only abrasion. If we had 516 used longer milling times for our experiments, the effects of splitting may have been masked by 517 continual abrasion of the split clasts. In a natural setting, initial splitting of clasts may add to the early 518 maximum in ash production by comminution between clasts as the splitting exposes rough surfaces that 519 are readily available for progressive abrasion (Dufek and Manga, 2008).

# 520 5.3 Enhanced ash production due to the interaction of pyroclasts with variable resistance

A recent study of PDC deposits from Tungurahua volcano in Ecuador (Bernard and Le Pennec, 2016) found that old lava clasts incorporated within the flow caused early splitting of pristine juvenile components (termed 'grinding'), due to the increased strength of the lava. This interpretation is supported by our supplementary Avellino experiment, Av3AG (Table 1). Here adding an agate ball to the grinding jar (analogous to adding a resistant lithology) enhanced ash production, particularly in the intermediate grain size fractions (Fig. 6c). The study of the Tungurahua PDC deposits further indicates that initial splitting (or 'grinding') of the juvenile clasts generates a 'milling factory' that produces small juvenile fragments that are available to abrade (Bernard and Le Pennec, 2016). This is consistent with our results showing that initial breakage of clasts (-3 or  $-2\varphi$ ) will enhance further ash production by abrasion (see section 5.2).

Taken together, these observations suggest that the presence (in PDCs) of mixed lithologies with different strengths encourages a productive milling factory, making block-and-ash flows prone to ash production. Large volume PDCs (ignimbrites), in contrast, tend to be pumice-dominated, with minor contributions of lithic fragments (e.g., Sparks et al., 1973). Therefore, enhanced splitting and abrasion due to grinding by a resistant lithic component could be less important in pumice-dominated flows than in flows with a higher lithic content. Our experiments also suggest, however, that even a small proportion of lithics will enhance ash production.

# 538 5.4 Controls of the starting materials texture on the characteristics of the ash produced

539 5.4.1 Ash grainsize

540 The fine modes of GSDs produced by abrasion experiments are  $5\phi$  for Avellino and  $4\phi$  for Fuego. The 541 difference in the fine modes reflects the different starting materials used. Thin vesicle walls provide 542 preferential places for rocks to fracture (e.g., Heap et al., 2014). If abrasion is caused by breakage of 543 juvenile clasts at vesicle walls, the size of the ash generated will be limited by the bubble wall thickness, 544 which explains the coarser mode  $(4\varphi)$  of the ash generated by the (thick walled) Fuego experiments. 545 Fracturing and breakage of volcanic rocks can also occur in the absence of vesicles. In this case it is 546 likely that the high crystallinity (~40%) of the Fuego groundmass will prevent extensive breakage of 547 particles into smaller grainsizes as microlites likely increase the strength of the groundmass (Heap et 548 al., 2016).

549 5.4.2 Ash componentry

The experimental GSDs show that the crystallinity and vesicularity of the starting material exert strong controls on the amount and size of ash generated by abrasion. The starting material properties also strongly influence the physical attributes of the resulting ash. For example, the abundance of vesicular ash grains is controlled by the minimum vesicle size in the starting material (Liu et al., 2015b). For this reason, the Fuego  $3\varphi$  (175-125 µm) ash lacks vesicular grains because the dominant vesicle size in Fuego clasts is >100µm in diameter (Figs. 3 and 10). Avellino ash of the same grain size, in contrast, contains a high proportion of vesicular grains because the average vesicle size in the pumice is smaller than the ash size (Fig. 10).

558 The crystal content of the ash reflects the crystallinity of the starting materials. The Fuego  $3\varphi$  ash 559 comprises >90% crystalline particles (free crystals and crystal-rich groundmass) compared to ~70% 560 crystalline particles in the Avellino 3\u03c6 ash (mostly phenocrysts with or without attached matrix). 561 However, the proportion of free crystals in the Avellino ash is higher than in the Fuego ash. This could 562 be the result of efficient liberation of crystals from the surrounding matrix during abrasion (Fig. 12) and/or breakage of larger crystals along cleavage planes (Jones et al., 2016). Without analysing ash 563 564 componentry in other grain size fractions, however, it is not possible to determine the primary origin of the free crystal fragments or the abundance of free crystals in the whole ash sample. 565

566 Experimental milling (abrasion) can also reduce the amount of matrix adhered to crystals (the 567 Adherence Factor, AF) of ash particles (Jones et al., 2016). The Adherence Factor trends shown in Figure 12 clearly distinguish the Fuego ash (higher proportion of high AF) from the Avellino ash 568 (mostly low AF values). The different AF trends reflect different textures in the starting material, most 569 570 importantly, the vesicularity of the matrix surrounding the crystals. For example, the low vesicularity 571 and thick bubble walls in the Fuego clasts (Fig. 3) means the maximum amount of matrix that can be attached to a crystalline ash particle is greater than for the Avellino pumice, thus explaining the higher 572 Another factor that could influence the AF is the variable thermal contraction of 573 AF values. 574 components during cooling which can create curvilinear fractures separating crystals from the glass 575 (Andronico and Cioni, 2002). These fractures would help liberate crystals from the melt increasing the occurrence of crystals with a minor amount of attached matrix and therefore a low AF. 576

577 5.4.3 Ash Shape

578 It has been suggested that co-PDC ash might have more uniform shapes than fall ash due to the 579 smoothing of grain boundaries by abrasion (Liu et al., 2015a; Taddeucci and Palladino, 2002). This was illustrated by Tambora co-PDC ash, which had a higher convexity (CVX) than fall ash with the same solidity (SLD), consistent with an abrasion-induced decrease of the roughness of grain perimeters during transport within PDCs (Liu et al., 2015a). The analysis was based, however, on a limited number of  $2\varphi$  (350-500µm) ash particles (Maria and Carey, 2007). Here we assess the extent to which finegrained ash generated by experimental abrasion records similarly high convexity values as the result of rounding.

586 The highest density of data points for both Avellino and Fuego ash (the 50% contour encloses 587 approximately half of the data) plot with CVX and SLD >0.9 (Fig. 8). However, the Avellino projected 588 area shape analysis extends to lower CVX and SLD values (~0.87) than for Fuego ash (~0.9). This 589 difference reflects the componentry, with the vesicular grains in the Avellino ash having low CVX and 590 SLD. The different SLD and CVX ranges for Avellino and Fuego ash are more pronounced in the 591 cross-sectional shape analysis, shown in Figure 9. This demonstrates the ability of this method to 592 distinguish particles characterised by contrasting internal structures rather than using the projected 593 shape (Morphologi). We therefore conclude that not all ash formed by abrasion will record high 594 convexity values, particularly when grains are vesicular. These data also show that the morphological 595 changes undergone by lapilli and ash can be decoupled during milling in PDCs, with larger clasts 596 rounding but newly generated ash having highly irregular shapes. This is particularly apparent in the 597 Avellino experiments where lapilli clasts exhibit pronounced rounding (Fig. 7) but the ash generated 598 preserves the initial textures of the starting material (high vesicularity) resulting in extremely irregular 599 ash shapes (Fig. 9).

Shape and componentry have been linked using BSE images (Liu et al., 2015a); here we extend this to projected area (Morphologi) images using simple componentry classifications that allowed us to relate CVX and SLD shape parameters to ash componentry in the  $3\varphi$  grainsize (Fig. 8). We can also use the shape data to infer componentry from CVX-SLD plots (Fig. S2). From these data, it appears that the vesicular clast content of the  $4\varphi$  Avellino ash is higher than for the  $3\varphi$  grainsize. This is contrary to expectation, as the proportion of vesicular fragments typically decreases with grain size when the bubble diameter approaches the average particle diameter. It is possible that this effect is not apparent between 3 and  $4\varphi$  due to the small modal vesicle size in the Avellino pumice and/or there may be fewer free crystal grains in the  $4\varphi$  size fraction. This comparison, and our interpretation of the results, demonstrates the dependency of componentry on grain size and builds on the conclusions of Liu et al. (2015b) that multiple grain sizes should be characterised for shape and componentry where feasible. Furthermore, the differences between results from the two methods of shape analysis - BSE and Morphologi - demonstrate that caution is required when comparing datasets collected by different methods and protocols (e.g. Liu et al., 2015a).

## 614 5.5 Comparison between experimental results and natural settings

615 The shapes of ash particles produced by experimental and natural processes are not identical. Ash from 616 a Fuego pyroclastic surge deposit, for example, has higher values of solidity and convexity than the 617 experimentally generated ash (Fig. 11). This is likely explained by the short duration of our experiments, 618 the experimental conditions of room (rather than high) temperature and the uniformity (in size and 619 density) of the starting material. Interestingly, although the experimental ash does not exactly replicate 620 the natural PDC-surge ash properties (Fig. 11b), the morphologies of the two ash samples are more 621 similar to each other than to ash produced by primary fragmentation (fall ash from Fuego 2016), which records a much larger range of CVX and SLD values (Fig. 11a). This suggests that the abrasion 622 experiments have successfully produced ash with shapes that replicate the shape characteristics of 623 624 naturally abraded ash.

Ash from the PDC matrix of the Avellino EU3pf deposit also has higher values of SLD and CVX than the experimental products (Fig. 11c), consistent with more extensive milling in a PDC compared to the short experimental milling times. An additional difference between the natural and experimental Avellino ash samples is the large lithic component of the natural PDC-matrix ash (Fig. 11f). These fragments of lavas, skarn and wall rocks provide evidence for the presence and abrasion of resistant lithologies during PDC emplacement.

The most compelling similarities between the natural and experimental samples, are the AdherenceFactor trends (Fig. 12), which closely match each other for both Fuego and Avellino. Moreover, the

633 AF trend of the intermediate Soufriere Hills pumice (intermediate in both vesicularity and crystallinity) 634 demonstrates that AF can be used to distinguish abrasion products that have formed from starting 635 materials with different physical properties, as the efficiency of matrix stripping is dependent on the 636 vesicularity and microlite content of the matrix. Finally, experiments on Avellino pumice show that 637 extensive abrasion within a PDC can produce abundant fine ash (mean size ~30-60µm). In natural 638 mixed fallout deposits (deposits containing both fall and material generated in the PDC), the GSDs can 639 be either uni- or bimodal depending on factors such as the distance from source and the positions relative 640 to the primary dispersion axis (Engwell and Eychenne, 2016). With increasing distance from the vent, 641 the GSD becomes unimodal converging on the fine mode as the size of the primary fall ash approaches 642 the grain size of the ash generated by secondary processes, such as abrasion in PDCs. The fine mode 643 of the GSDs produced by the abrasion experiments are  $5\phi$  for the Avellino and  $4\phi$  for the Fuego. This 644 size range not only overlaps with natural co-PDC ash (mean of the fine subpopulations of bimodal 645 samples), but may also explain the variations in co-PDC ash size from different source volcanoes. For 646 example co-PDC (blast) ash from Mount St Helens has a mean size of 5-6 $\phi$ , which contrasts with the 4φ mean of the highly crystalline Fuego samples (Engwell and Eychenne, 2016). 647

The amount and size distribution of fine ash will be a key factor in the development of a co-PDC plume, which can transport ash to the atmosphere (Woods and Wohletz, 1991). Numerous co-PDC layers associated with large pumice-rich PDCs are found in the stratigraphic record, including eruptions from Somma Vesuvius and the nearby Phlegrean fields (e.g., Engwell et al., 2014; Sulpizio et al., 2008). These deposits support our finding that crystal-poor pumice clasts, such as those that comprise the Avellino deposit, are susceptible to extensive secondary fragmentation and co-PDC plume formation, which can greatly enhance ultra-distal transport of fine ash.

In contrast, discrete co-PDC ash layers from block and ash flows, such as from Fuego, are much less common. Evidence of ash generation in such flows comes mainly from observation of the bimodality of the fallout deposit grainsize (e.g., Engwell and Eychenne, 2016; Eychenne et al., 2012). Nonetheless, abrasion within block and ash flows can generate sufficient fine ash to increase their mobility, thus elevating their risk factor (Cole et al., 1998; Fujii and Nakada, 1999). However, we suggest that the 660 elevated groundmass crystallinity of constituent clasts will ultimately limit the smallest grain size that 661 can be produced by abrasion, and the efficiency of ash generation. Therefore, crystal-rich material in 662 block and ash flows produced by microlite-rich magma is less likely to produce a co-PDC plume with 663 a large dispersal extent than low density, low crystallinity pumice flows.

664 6. Conclusions

665 Previous abrasion experiments have found that low density (high vesicularity) pyroclasts generate more ash than high density (low vesicularity) pyroclasts. Although generally true in our experiments, we 666 667 found that the pyroclast density (vesicularity) alone is insufficient to predict how pyroclastic material 668 will react to abrasion. The ash-generating potential of pyroclastic material is also controlled by the 669 crystal content, as well as heterogeneities in bubble and crystal textures. Two key relationships were 670 observed between the amount of ash generated and the crystal content. Firstly, experiments on 671 microlite-rich pyroclasts from Fuego volcano showed that increasing the heterogeneity of the crystal 672 population of a (relatively) low-density clast renders it more resistant to abrasion than a high-density 673 clast with more uniform phenocryst and groundmass characteristics. Secondly, details of both the 674 vesicle and crystal content of pyroclasts can encourage clasts to split rather than abrade. Increasing the 675 frequency of clast splitting enhances the ash-generating potential because splitting creates new irregular 676 surfaces that are susceptible to abrasion.

677 The vesicularity and crystal content of the starting material strongly influences the properties of the ash 678 generated by abrasion, in addition to the amount of ash generated. Critically, high microlite crystallinity 679 limits the generation of very fine ash, as shown by the lower abundance and larger size of the ash 680 generated in the crystal-rich Fuego experiments ( $4\phi$ ) compared to the crystal-poor Avellino experiments 681  $(5\phi)$ . The vesicularity and crystal content of pyroclasts also controls the componentry of ash generated 682 by abrasion, which in turn influences ash shape and, by extension, elutriation and transport (settling) 683 properties. Co-PDC ash with mixed componentry, (e.g. mainly free crystals and highly vesicular ash), 684 poses a challenge to dispersion modelling as typically a single density measurement is required for dispersion models (e.g., Mastin et al., 2010; Witham et al., 2007). Moreover, the vesicular glass 685 686 particles will be preferentially elutriated compared to crystals of the same size, due to their low density,

altering the componentry of the ash that is dispersed from the PDC to the atmosphere as evidenced bycrystal-enriched ignimbrite deposits (Sparks and Walker, 1977).

689 Abrasion can also alter ash shape. Ash produced in a PDC-surge deposit from Fuego preserves grains 690 with reduced roughness compared to natural fall Fuego ash (e.g., Liu et al., 2015a). However, we did 691 not detect reduced roughness of ash generated by abrasion of the highly vesicular Avellino ash. The 692 Adherence Factor, which quantifies the amount of matrix adhered to (micro)phenocrysts, is most 693 effective for distinguishing ash that has undergone different degrees of abrasion (Jones et al., 2016) We 694 also have shown that AF can clearly distinguish ash from different starting materials due to the 695 maximum amount of matrix that can be attached to a crystal which is determined by the vesicularity of 696 the matrix. Efficient abrasion strips the matrix from ash grains and this process is more efficient in high vesicularity materials with low-crystallinity vesicular groundmass such as the Avellino pumice. 697

When compared to natural samples, the experimentally generated ash differed somewhat from the natural PDC-surge and PDC-matrix ash samples due to differences in abrasion conditions and the presence of xenoliths. However, the fine modes of the grainsize distributions produced by the milling are similar to the fine modal grainsizes measured in natural fallout deposits where co-PDC ash has been recorded (Engwell and Eychenne, 2016). This suggests the experimental abrasion is effectively replicating the natural process.

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#### 713 Figures, captions and tables (for high quality figures please see separately uploaded pdfs):

Figure 1: Density histograms of the -3φ juvenile populations of both Fuego (a) and Avellino (b). The bins are 0.1gcm-3 intervals and the number of clasts measured is denoted by 'n'. The diamond, circle and triangle symbols indicate the mean density of the low density (LD), average density (AD) and high density (HD) sub-samples, respectively. a.) FUE2a: filled black symbols, FUE3a: white symbols and FUE5a: open black symbols. b.) Av31a: filled black symbols, Av32a: white symbols and Av33a: open black symbols. The upper x-axis is the vesicularity (%) calculated using the DRE densities (Houghton and Wilson, 1989). See Table 1 for the list of samples and experiments.

Figure 2: Summary of abrasion experiment apparatus. The Retsch Planetary Ball Mill PM100 consists of a rotating basal plate and agate-lined 7.5cm diameter milling jar which contains the sample. The rotation rate and milling duration are input by the user. The sample is retrieved after milling using a clean fine brush and stored in a tightly sealed sample bag. Between all samples and sub-samples, the milling jar is cleaned with clean silicon sand, milled for 4 minutes.

Figure 3: Textural differences in vesicle populations between Fuego sub-samples from BSE-SEM images. a.) Low density clast, b.) low density clast with large glomercrysts and increased microlite content, c.) average density clast and d.) high density clast. The boxes below contain the density of individual clasts and the results of the two methods of quantifying vesicularity. The VF values are lower than the VD values for the LD and HD clasts as the 2D imaging tends to exclude the largest vesicles. e.) to h.) are the vesicle size distributions of the e.), f.) low Density, g.) average density and h.) high density clasts. Bin sizes are in phi increments.

Figure 4: Irregularities in vesicularity and crystal content within Avellino pumice. a.) An example of a localised area of dense microlite rich melt. b.) A close-up view of the boundary between the two zones of crystal and vesicle content. c.) An image of an embedded fragment of thermally recrystallised limestone with large surrounding vesicle likely due to thermal decomposition of the carbonate minerals. Figure 5: Grain size histograms after 4 minutes of milling. The grey bar represents the starting grain
size. 'Expected trend' meaning, low density (high vesicularity) sub-samples generate more ash than
high density (low vesicularity) sub-samples.

Figure 6: Total grainsize distributions of all samples following 4 minutes of abrasion in a planetary ball mill. The starting grainsize is represented by the y-axis  $(-3\varphi)$ . a.) TGSDs generated following three abrasion experiments using Fuego juveniles. Different lines styles correspond to different density subsamples. b.) The GSDs generated following the abrasion experiments using the Avellino juveniles. The colours correspond to the histograms in Figure 5. c.) TSGDs of supplementary Avellino experiments. \*Av31b used a -2 $\varphi$  starting grainsize fraction.

Figure 7: Box plots of form factor (FF) measurements for *all* milling experiments of Fuego and Avellino material. Numbered boxes indicate clast count for all three experiments before  $(-3\varphi)$  and after milling (-3 and -2 $\varphi$ ). Black forms at top are examples of binary images used to measure the FF using ImageJ. The examples are from the F2a AD and Av33a HD sub-samples. Small black boxes indicate outliers, which only occur in the Avellino data.

Figure 8: Morphologi shape analysis of F3a and Av31a AD  $3\varphi$  ash using shape factors solidity and convexity. a.) Fuego ash analysis. b.) Avellino ash analysis. 'n' denotes the grain count. The points are translucent, so the stronger colour indicates more points. The contours are of the kernel density and are labelled at with the proportion of points they enclose. c.) and d.) are plots of randomly selected 500 grains from the Fuego (c) and Avellino (d) ash that have been split into different components.

Figure 9: Results of ImageJ and BSE image analysis of a.) F3a and b.) Av31a 3φ AD sample ash at
high image resolution (~ 1560 pixels/mm). Scatter plots of solidity and convexity shown. The contours
and dashed boxes are where the Morphologi results of the same sample plot (Figure 8). Binary images
of ash grains are shown along axes to demonstrate range in shape parameters.

Figure 10: Componentry analysis from BSE images of a.) F3a and b.) Av31a 3φ (125-180 μm) ash.
The glass category refers to particles that are almost entirely vesicle-free and microlite-free glass.
Images of each component type can be found in Figure S3.

763 Figure 11: Cross-sectional shape analysis of natural and experimental ash samples. a.) to c.) Kernel density contours of solidity and convexity of a.) natural fall ash from a paroxysm of Volcan de Fuego 764 in 2016; b.) natural Fuego PDC-surge ash from 2012 (E. Liu, personal comm.); c.) natural PDC-matrix 765 ash from the EU3pf deposit. The contour intervals correspond to those in Figure 8 and are labelled with 766 the proportion of data they enclose. The shaded contours at the same intervals represent the Fuego F3a 767 (red) and Avellino Av31a (blue) SLD and CVX data of experimental ash at reduced resolution (~390 768 769 pixels/mm). d.) and e). are cumulative frequency plots of convexity and solidity. The dashed lines 770 correspond to the natural samples which record a higher proportion of high CVX and SLD values than the equivalent experimental ash sample. f.) BSE image of the natural Avellino PDC-matrix ash, 771 772 highlighting the large lithic component.

Figure 12: Adherence factor plot of the Avellino and Fuego milling experiments, the natural PDCsurge and matrix ash samples and the data from two milling experiments carried out by Jones et al., (2016) using Soufriere Hills Volcano (SHV) pumice; the latter were shorter duration experiments but six agate milling balls used so we assume similar milling efficiencies. Adherence Factor is plotted cumulatively so that each point indicates the percentage of crystals with that AF value or less. AF values close to 1 represent a large amount of attached matrix.

| * There w                                                     | AV-16-01               | Av3AG    |     |     | Av31b |                        |                          | Av33a                    |     |              | Av32a |                        |                        | Av31a                  | FUE003       |              |     | FUE5a        |                        |              | FUE3a        |                          |                          | FUE2a                    | Sample<br>Name                         |
|---------------------------------------------------------------|------------------------|----------|-----|-----|-------|------------------------|--------------------------|--------------------------|-----|--------------|-------|------------------------|------------------------|------------------------|--------------|--------------|-----|--------------|------------------------|--------------|--------------|--------------------------|--------------------------|--------------------------|----------------------------------------|
| were 7 clast                                                  | Avellino f             | AD       | HD  | AD  | LD    | HD                     | AD                       | LD                       | HD  | AD           | LD    | HD                     | AD                     | LD                     |              | HD           | AD  | LD           | HD                     | AD           | LD           | HD                       | AD                       | LD                       | Sub-<br>Samples                        |
| but one bro                                                   | all pumice no          | 7        | 40  | 40  | 40    | 7                      | 7                        | 7                        | 7   | 7            | 8*    | 7                      | 7                      | 7                      |              | 10           | 10  | 10           | 10                     | 10           | 10           | 10                       | 10                       | 10                       | No. of<br>clasts BM                    |
| roke during loading prior to milling                          | t used in abrasion exp | 5        |     |     |       | -                      | 2                        | 1                        | 3   | 2            | 1     | 1                      | 3                      | 1                      |              | 4            | 5   | 7            | 8                      | 6            | 4            | 9                        | 8                        | 8                        | No. of clasts AM<br>(remaining in -3φ) |
|                                                               | periments, only vesicu | 9        | 42  | 38  | 15    | 9                      | 7                        | 7                        | 7   | 10           | 12    | 10                     | 7                      | 6                      |              | 13           | 13  | 11           | 10                     | 12           | 9            | 12                       | 11                       | 10                       | No. of clasts AM<br>(in -3φ and -2φ)   |
|                                                               | larity chara           | 4        | 5   | S   | S     | 4                      | 4                        | 4                        | 4   | 4            | 4     | 4                      | 4                      | 4                      |              | 4            | 4   | 4            | 4                      | 4            | 4            | 4                        | 4                        | 4                        | Milling<br>time<br>(mins)              |
| ** Imaging and FOAMS analysis carried out by Voarino, (2016). | cterisation **         | 450      | 450 | 450 | 450   | 450                    | 450                      | 450                      | 450 | 450          | 450   | 450                    | 450                    | 450                    |              | 450          | 450 | 450          | 450                    | 450          | 450          | 450                      | 450                      | 450                      | Rotation<br>Rate<br>(rpm)              |
|                                                               | 2 x clast              |          |     |     |       | 1 x clast AM           |                          |                          |     | 1 x clast AM |       | 4 x clasts AM          | 2 x clasts AM          | 3 x clasts AM          | 2 x clast BM | 1 x clast AM |     | 1 x clast AM |                        | 2 x clast AM | 1 x clast AM | 1 x clast AM             |                          | 2 x clasts AM            | Sample<br>Characterisation             |
|                                                               | 1 x clast              |          |     |     |       |                        |                          |                          |     |              |       |                        |                        |                        | 1 x clast    | 1 x clast    |     | 1 x clast    | 1 x clast              | 2 x clast    | 1 x clast    |                          |                          | 2 x clast                | FOAMS                                  |
|                                                               |                        | <i>۲</i> |     |     |       | <i>۲</i>               | ٩                        | ۲                        | <   | ٩            | <     | <i>۲</i>               | ۲                      | ۲                      |              | <            | ۲   | ۲            | <i>د</i>               | ٩            | ۲            | <                        | ٩                        | ۲                        | Clast<br>Shape                         |
|                                                               |                        |          |     |     |       |                        |                          |                          |     |              |       | 🗸 3φ, 4φ               | 🗸 3φ, 4φ               | 🗸 3φ, 4φ               |              |              |     |              | 🗸 3φ, 4φ               | 🗸 3φ, 4φ     | 🗸 3φ, 4φ     |                          |                          |                          | Morphologi<br>Analysis                 |
|                                                               |                        | ✓ 3φ     |     |     |       | <ul><li>✓ 3φ</li></ul> | <ul> <li>✓ 3φ</li> </ul> | <ul> <li>✓ 3φ</li> </ul> |     |              |       | <ul><li>✓ 3φ</li></ul> | <b>√</b> 3φ            | ✓ 3φ                   |              |              |     |              | <ul><li>✓ 3φ</li></ul> | <b>√</b> 3φ  | ✓ 3φ         | <ul> <li>✓ 3φ</li> </ul> | <ul> <li>✓ 3φ</li> </ul> | <ul> <li>✓ 3φ</li> </ul> | BSE-SEM<br>and ImageJ<br>Analysis      |
|                                                               |                        |          |     |     |       |                        |                          |                          |     |              |       | <ul><li>✓ 3φ</li></ul> | <ul><li>✓ 3φ</li></ul> | <ul><li>✓ 3φ</li></ul> |              |              |     |              |                        |              |              | ✓ 3φ                     | <ul><li>✓ 3φ</li></ul>   | <ul><li>✓ 3φ</li></ul>   | Adherence<br>Factor                    |

> mounted for vesicularity and crystal content assessment. Some of the sample characterisation was before milling (BM clasts) and some was done after milling (AM clasts). Table 1 – Abrasion experiments and summary: sample names, experiment conditions and subsequent analysis. The 'sample characterisation' column lists the clasts that were

The clast shape analysis column signifies the samples where the larger clasts were photographed, and their shape analysed. All experiments started with -3 p clasts solely and

| Table 2 – Shape parameters and definitio   | is. This table only highlights key parameters used in this study, see Liu et |
|--------------------------------------------|------------------------------------------------------------------------------|
| al., (2015) for a comprehensive summary of | f shape factors and uses.                                                    |

| Shape<br>Parameter  | Abbreviation | Formula                            | Symbols                                                                      | Sensitivity                  | Alternative<br>nomenclature                                                                | References                                               |
|---------------------|--------------|------------------------------------|------------------------------------------------------------------------------|------------------------------|--------------------------------------------------------------------------------------------|----------------------------------------------------------|
| Form<br>Factor      | FF           | $4\pi A_{\rm P}/{\rm P_{\rm P}}^2$ | $A_P$ = area of<br>particle<br>$P_P$ = perimeter of<br>particle              | Form and roughness           | Sphericity, Roundness,<br>Circularity, HS<br>Circularity, Shape<br>Factor, Cox circularity | Liu et al.,<br>2016 and<br>references<br>within          |
| Solidity            | SLD          | A <sub>P</sub> /A <sub>CH</sub>    | $A_P$ = area of<br>particle<br>$A_{CH}$ = area of<br>convex hull             | Roughness<br>(morphological) |                                                                                            | Liu et al.,<br>2016                                      |
| Convexity           | CVX          | P <sub>CH</sub> /P <sub>P</sub>    | $P_{CH}$ = perimeter of<br>convex hull<br>$P_{P}$ = perimeter of<br>particle | Roughness<br>(textural)      | Roughness                                                                                  | Liu et al.,<br>2016                                      |
| Adherence<br>Factor | AF           | $A_M/A_M+A_C$                      | $A_M$ = area of<br>attached matrix<br>$A_C$ = area of<br>crystal             | Amount of<br>attached matrix | Abrasion index $(A_P/A_M)$                                                                 | Jones et al.,<br>2016,<br>Freundt &<br>Schminke,<br>1992 |

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# a.) Fuego



b.) Avellino











# AVELLINO

b.) Expected trend (Av33a)







12



















F3a AD 3p







Adherence Factor