Appendix 1

Aspect ratio dependence of SEM-CSDs

CSDCorrections (Higgins 2000) addresses two stereological problems of the conversion from 2D to 3D: the cut-section effect and the intersection-probability effect. The stereological corrections are applied sequentially from the largest size interval (i.e., the first interval) to smaller intervals. The number of crystals per unit volume in the *i*th interval, n_{Vi}, is converted stereologically from the number of crystal cross sections per unit area, n_{Ai}, as (modified from Equations 5 and 8 of Higgins 2000 and Equation 7 of Sahagian and Proussevitch 1998):

$$n_{Vi} = \left(n_{Ai} - \sum_{j=1}^{i-1} n_{Vj} P_{ji} \overline{H}_j \right) \cdot \frac{1}{P_{ii} \overline{H}_i} = n_{Ai} \cdot \frac{1 - \sum_{j=1}^{i-1} \frac{n_{Vj} P_{ji} \overline{H}_j}{n_{Ai}}}{P_{ii}} \cdot \frac{1}{\overline{H}_i}$$

$$n_{Vi} = n_{Ai} \cdot \frac{1 - CF_i}{P_{ii}} \cdot \frac{\ln(y_i/x_i)}{y_i(L) - x_i(L)} \qquad \text{(A1)}$$

where $x_i(L)$ and $y_i(L)$ are the lower and upper limits of the *i*th interval of *L*, P_{ji} is the probability that a crystal with a true (i.e., determined in 3D) size in the *j*th interval will have a cross-sectional length in the *i*th interval, and CF_i is a correction factor representing the proportion of crystals with true sizes larger than the *i*th interval among the cross sections within that interval. $–H_i$ is the Mean Projected Height defined by Tuffen (1998) and the Equation 8 of Higgins (2000). Regarding the

Equation (A1), the first term, n_{Ai}, is obtained by dividing the number of crystal cross sections by the area analyzed. The second and third terms are correction terms for the effects of sectioning and the probability of intersection, respectively. Note that the interval width as a function of *L* is calculated from Equation (1). More specifically, in the case of conversion from *w*, the interval width is:

$$y_i(L) - x_i(L) = A \cdot \left(y_i(S) - x_i(S)\right) \qquad \text{(A2a)}$$

and in the case of conversion from *l*, it is:

$$y_i(L) - x_i(L) = B \cdot \left(y_i(I) - x_i(I)\right). \qquad \text{(A2b)}$$

The population density in the *i*th interval, N_i, is obtained by dividing n_{Vi} (cf. Equation A1) by the interval width (Equation 10 of Higgins 2000). Consequently, *CSDCorrections* calculates the *L*-plot population density as:

$$N_i(L) = \frac{n_{Vi}}{y_i(L) - x_i(L)} = n_{Ai} \cdot \frac{1 - CF_i}{P_{ii}} \cdot \frac{\ln(y_i/x_i)}{\left(y_i(L) - x_i(L)\right)^2}$$

$$N_i(L) = \frac{1}{A^2} \cdot \frac{1 - CF_i}{P_{ii}} \cdot \frac{n_{Ai} \ln(y_i/x_i)}{\left(y_i(S) - x_i(S)\right)^2} \qquad \text{(A3a)}$$

or

$$N_i(L) = \frac{1}{B^2} \cdot \frac{1-CF_i}{P_{ii}} \cdot \frac{n_{Ai} \ln(y_i/x_i)}{\left(y_i(I)-x_i(I)\right)^2}. \tag{A3b}$$

To obtain *S*-plot and *I*-plot CSDs, we converted the *L*-plot population densities output by *CSDCorrections*, $N_i(L)$, as:

$$N_i(S) = \frac{n_{Vi}}{y_i(S)-x_i(S)} = \frac{y_i(L)-x_i(L)}{y_i(S)-x_i(S)} \cdot N_i(L) = A \cdot N_i(L) \tag{A4a}$$

$$N_i(I) = \frac{n_{Vi}}{y_i(I)-x_i(I)} = \frac{y_i(L)-x_i(L)}{y_i(I)-x_i(I)} \cdot N_i(L) = B \cdot N_i(L). \tag{A4b}$$

Substituting Equation (A3) into Equation (A4) gives:

$$N_i(S) = \frac{1}{A} \cdot \frac{1-CF_i}{P_{ii}} \cdot \frac{n_{Ai} \ln(y_i/x_i)}{\left(y_i(S)-x_i(S)\right)^2} \tag{A5a}$$

$$N_i(I) = \frac{1}{B} \cdot \frac{1-CF_i}{P_{ii}} \cdot \frac{n_{Ai} \ln(y_i/x_i)}{\left(y_i(I)-x_i(I)\right)^2}. \tag{A5b}$$

The third terms of Equations (A3) and (A5) are independent from the 3D aspect ratio used for the 2D–3D conversion. As shown by the first right-hand terms of the equations, $N_i(L)$ depends more strongly on the aspect ratio than do $N_i(S)$ and $N_i(I)$. In addition to the aspect ratio dependence of the population densities, the *L*-plot SEM-CSDs require an additional correction in which their horizontal axes are enlarged by *A* or *B* times. This procedure softens the slopes of *L*-plot CSDs by *A* or *B* times those in *S*- or *I*-plot CSDs, respectively. Therefore, *L*-plot CSDs are more strongly changed by the aspect ratio (*A* or *B*) than are *S*- and *I*-plot CSDs (Fig. S1).

Appendix 2

Simulation of CSDs based on Marsh (1998)

Marsh (1998) formulated CSDs in the non-steady closed systems by employing the Johnson-Mehl-Avrami equation for crystallinity related to time-variant nucleation and growth rates. Considering the exponential variations in time of nucleation (J) and growth (G) rates, their functions are

$$J(x) = J_o \exp(ax) \tag{A6}$$

$$G(x) = G_o \exp(bx) \tag{A7}$$

where a and b are constants, and x is the dimensionless time (0–1). The dimensionless time x is normalized by the crystallization duration, τ (i.e., $x = t/\tau$). The subscript o for the parameters indicates the initial values (i.e., at time $x = 0$). The final size (i.e., at $x = 1$) of a crystal which nucleated at a certain time x, $R(x)$, is expressed as follows (cf. Equation 5 in Marsh 1998).

$$R(x) = \frac{G_o \tau}{b}\{\exp(b) - \exp(bx)\} \tag{A8}$$

The natural logarithm of population density of crystals which nucleated at the certain time x, Ln $N(x)$, is expressed as (modified from Equations 3, 12, and 26 in Marsh 1998):

$$\mathrm{Ln}\,N(x) = \mathrm{Ln}(J_o/G_o) - \frac{4\pi}{3}{G_o}^3 J_o \tau^4 f(x,a,b) + (a-b)x \tag{A9}$$

where the function $f(x, a, b)$ is defined as the Equation 10 in Marsh (1998):

$$f(x,a,b) = \int_0^x \exp(ax')\left\{\int_{x'}^x \exp(bx)\,\mathrm{d}x\right\}^3 \mathrm{d}x'. \tag{A10}$$

From the Equations (A8) and (A9), we obtain the simulated CSDs for specified kinetic conditions (*a*, *b*, J_o, G_o, and τ) by plotting Ln $N(x)$ against $R(x)$ for x = [0, 1]. To investigate the effect of increasing growth rate on CSDs under different ascent paths, we simulated four sets of conditions as shown in Table S1. The ranges of the kinetic parameters (Figs 2c and 2d) are realistic (cf. Marsh 1998; Shea and Hammer 2013).

Appendix 3

SEM-CSDs converted using XCT average aspect ratios

For the stereological conversions, we used two distinct 3D aspect ratios to assess the effect of the estimation error on the CSD shapes: the value estimated from the 2D data by *CSDslice* (Morgan and Jerram 2006) and the average value determined by SR-XCT (Table 4). Here, we refer to the SEM-CSDs converted from the datasets of *w* and *l* and corrected with the ratio estimated from the 2D data as SEM(*w*-2D)-CSDs and SEM(*l*-2D)-CSDs, respectively, and those corrected with the average ratio from the SR-XCT data as SEM(*w*-3D)-CSDs and SEM(*l*-3D)-CSDs, respectively. In addition to the seven types of CSDs presented in the main text, we thus obtained 11 types of CSDs.

Figure S2 compares CT-CSDs, SEM(2D)-CSDs, and SEM(3D)-CSDs. We observed obvious discrepancies between the SEM(2D)-CSDs and the SEM(3D)-CSDs for the sub-Plinian pumice that resulted from the 3D aspect ratio used. The *S*- and *I*-plot SEM-CSDs are vertically displaced but have similar shapes, and the SEM(3D)-CSDs were closer to (i.e., less vertically displaced from) the CT-CSDs (Figs. S2a and S2b; Table 4). Although the SEM(3D)-CSDs were almost consistent with the *L*-plot CT-CSD, the SEM(2D)-CSDs were considerably distorted (Fig. S2c). In the Vulcanian *L* plot (Fig. S2f), the slopes of the SEM(*w*)-CSDs differed slightly from that of the CT-CSD, irrespective of the 3D aspect ratio used, whereas the SEM(*l*)-CSDs were similar to the CT-CSD. This discrepancy may reflect the non-equivalence between the *w* and *l* datasets (Higgins 1994; Muir et al. 2012) or may indicate that both of the *L*/*S* ratios (*A*) differed from an appropriate value, which is possibly associated with the large variation in the ratios (Castro et al. 2003).

Comparing the sub-Plinian and Vulcanian pumice samples, the SEM(3D)-CSDs were similar in the *L* plot (Fig. S3f), but different in the *S* and *I* plots, especially in the size range of nanolites (Figs. S3d and S3e). Consequently, the SEM(3D)-CSDs more clearly reflected the difference in magma ascent conditions in the *S* and *I* plots than in the *L* plot (Table 4), consistent with the CT-CSDs (Fig. 8).

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