I. Estimation of Particle Size Distribution

This appendix summarizes the procedures used to obtain the particle size distribution PSD shown in Figure 3. Figure 1 shows a FESEM microscopic photo of part of a particle laden 0.02 μm Anodisc filter taken with 500X magnification. To measure the sizes of the particles observed in this photo, we first use the microscopic image processing software ImageJ to draw the outline of each distinguishable particle and measure the area enclosed in the outline (Figure 1b). We then determine the effective radius of each particle as the radius of the circle having an identical area.

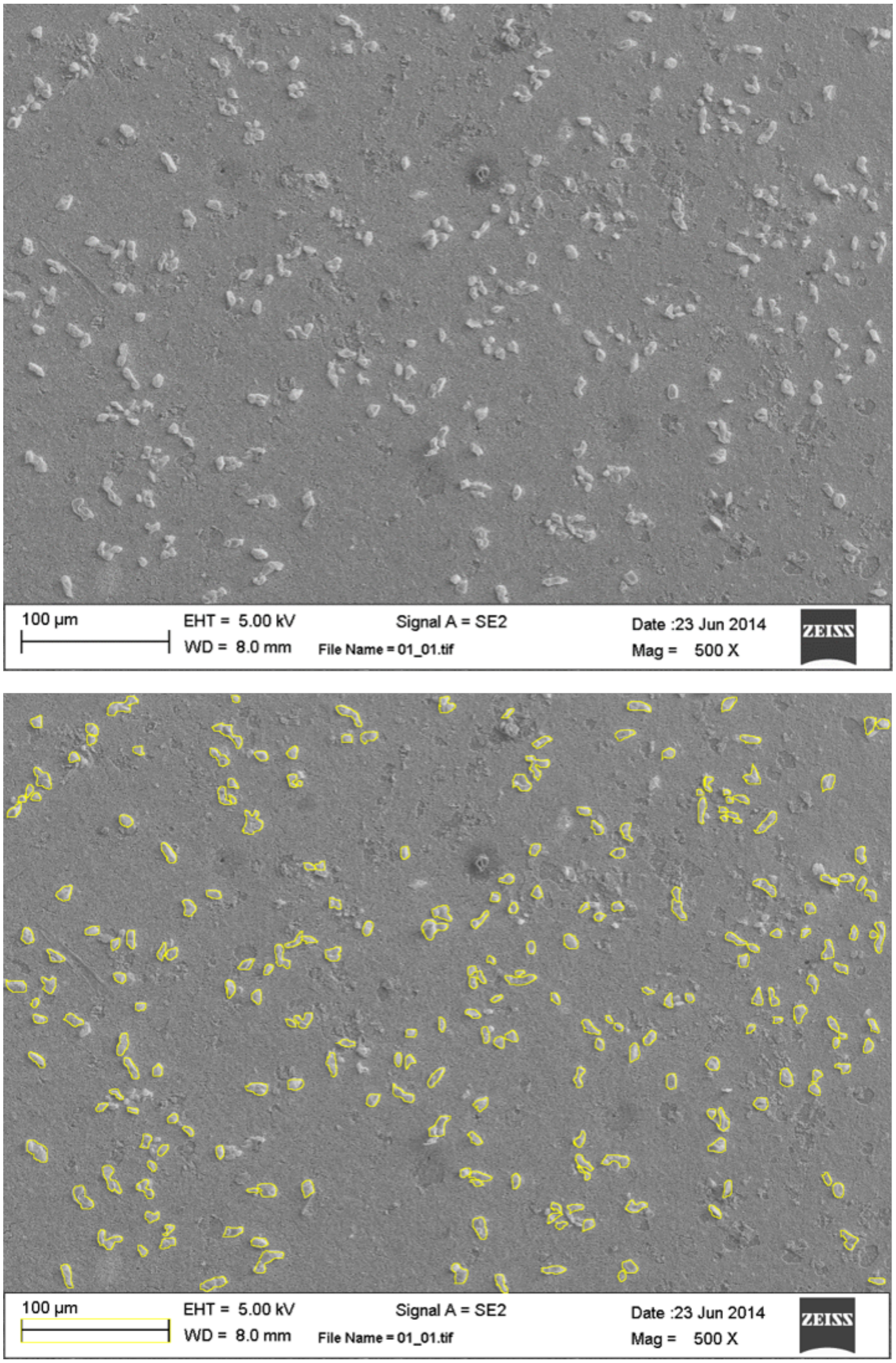

Figure 1: (a) A 500X FESEM microscopic photo of the particle sample taken from the North Tower plume. (b) Same as (a) with the outlines of the distinguishable particles drawn using the microscopic image processing software ImageJ.

In order to better resolve the sizes of small particles, for the sample from the 1st Niskin bottle, we apply the method above to a total of 6 photos of a given view (i.e. the visible area in a 500X photo) taken with 500X, 1000X, 2000X, 4000X, 8000X, and 16000X magnification respectively. The minimal particle radius that can be reliably measured under each magnification is: 3 μm under 500X, 1.5 μm under 1000X, 1 μm under 2000X, 0.5 μm under 4000X, 0.24 μm under 8000X, and 0.12 μm under 16000X. For the sample from the second Niskin bottle, we apply the same method to only three photos of a given view taken with 500X, 1000X, 2000X magnification respectively. The choice of fewer photos per view for the sample from the 2nd bottle is because the lack of distinguishable particles within the 0.12 to 0.5 μm range on the filter.

We then calculate the histogram of particle radius between 0.2 and 18 μm (the largest observed particle radius) in 0.1 μm intervals. Subsequently, we calculate the weighted average of the histograms obtained from the 6 photos of the same view. The weight applied to each photo is the ratio of the visible area in that photo to the visible area in a 500X photo (e.g., the weight applied to a 500X photo is 1 and the weight applied to a 1000X photo is 4). The result is a discrete size distribution showing the number of particles per view within each 0.1 μm radius bin between 0.2 and 18 μm. Lastly, we repeat the aforementioned procedures on 25 views of the particle sample from the first Niskin bottle and 30 views of the sample from the second bottle and average the results to obtain the final discrete particle size distribution shown in Figure 2.

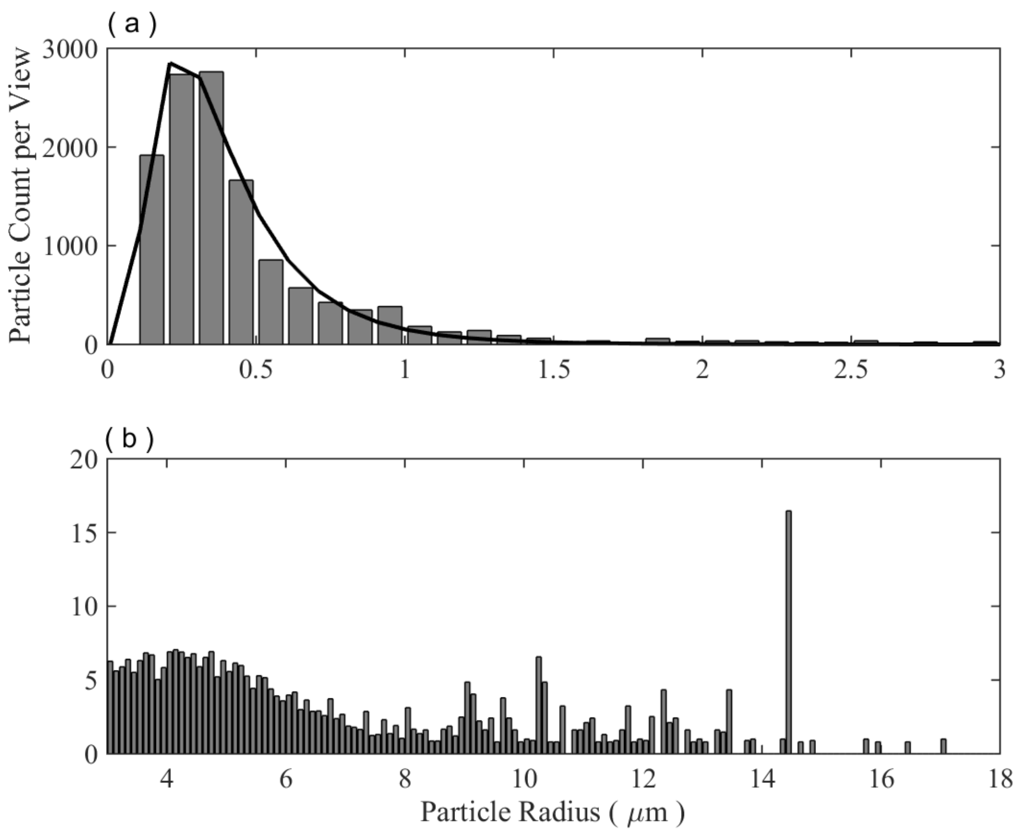

Figure 2: Discrete size distribution of particles having radii (a) from 0.01 to 3 μm and (b) from 3 to 18 μm. The solid curve in (a) is the least-squares fit of the log-normal curve having the form: $n(a) = \frac{\Lambda}{a} \exp\left(-\frac{[\log(a)-\Upsilon]^2}{\Sigma^2}\right)$, where n is particle count, a is particle radius, $\Lambda = 849.94$, $\Upsilon = -1.08$, and $\Sigma = 0.82$.

We then extend the particle size distribution obtained above to account for small particles (<0.2 μm) that cannot be resolved and large particles (>18 μm) that could have been missed in our analysis. For particles having radii smaller than 0.2 and larger than 0.01 μm (the hypothesized smallest particle radius), we fit a log-normal distribution curve having the following mathematical expression to the discrete size distribution obtained above

$$n(a) = \frac{\Lambda}{a} \exp\left(-\frac{[\log(a) - \Upsilon]^2}{\Sigma^2}\right) \tag{1}$$

where n is particle count, a is particle radius, and Λ, Υ, Σ are adjustable constants. The choice of the log-normal distribution is based on its similarity to the overall shape of the discrete particle size distribution shown in Figure 2a.

For particles having radii greater than 18 μm, we assume the particle count decreases with the radius exponentially as

$$n(a) = n_{18} \exp\left(-[a - 18\,\mu\mathrm{m}]^2/b^2\right) \tag{2}$$

where n_{18} is the number of particles having radii of 18 μm, and b is the e-folding radius of the exponential decrease which is arbitrarily set to 2 μm. In this way, we obtain a discrete particle size distribution over the size range $0.01 \leq a \leq 500\,\mu$m (the maximum particle size reported in Feely *et al.* [1987] based on the samples taken from several vents on the Endeavour Segment) in 0.1 μm intervals. We then calculate the probability density function (PDF) of particle size distribution as

$$P(a) = \frac{n(a)}{N\Delta a} \tag{3}$$

where $P(a)$ is the PDF so that $P(a)\Delta a$ is the probability of a plume particle having a radius between a and $a + \Delta a$, N is the total number of particles over the whole size range, and $\Delta a = 0.1\,\mu$m. The effective mean grain size corresponding to the PDF is $a_0 = \sum_{0.01}^{500} aP(a)\Delta a = 0.75\,\mu$m.

Lastly, we conduct Monte-Carlo simulations to quantify the uncertainty in the PDF obtained above. We first randomly pick 30 out of the total of 55 views to calculate a new PDF following the same procedures mentioned above. We then repeat the preceding step 5000 times and calculate the 95% confidence interval of the resulting PDFs.

REFERENCES

Feely, R. A., Lewison, M., Massoth, G. J., Robert-Baldo, G., Lavelle, J. W., Byrne, R. H., Damm, K. L. V., and Herbert C.Curl, J. (**1987**). “Composition and Dissolution of Black Smoker Particulates from Active Vents on the Juan de Fuca Ridge”, Journal of Geophysical Research **92**, 11347–11363.