

Interactive comment on “Effective density of Aquadag and fullerene soot black carbon reference materials used for SP2 calibration” by M. Gysel et al.

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As black carbon (BC) particles are typically not spherical but rather composed of aggregates of smaller spherules, determination of the mass m of such a particle from its mobility diameter D_{mob} is not straightforward. The authors parameterize the rela-

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tion between m and D_{mob} in terms of the effective density ρ_{eff} , which is the density calculated under the assumption that the particle is spherical with diameter equal to the mobility diameter: $m = (\frac{\pi}{6})\rho_{\text{eff}}D_{\text{mob}}^3$. Further, they present relations between this effective density and mobility diameter for Aquadag particles with D_{mob} between 60 and 710 nm and for Fullerene particles with D_{mob} between 65 and 900 nm, which they display (along with their measurements) on a log-log plot. The relation for Aquadag is an 8th order polynomial and that for Fullerene a 9th order polynomial, with coefficients presented in Table 1, together with a warning not to round to coefficients to fewer significant digits (most are given to six significant digits). However, despite the appearance of high accuracy, the estimated accuracy of the measurements is only $\pm 10\%$. In light of such a large uncertainty it would seem that such a function form for the fits is a bit excessive, and that the data could be represented nearly as well with a simpler fit.

Indeed, if the effective density for Aquadag and Fullerene is graphed against the mobility diameter on a linear-linear plot, as shown in Figure 1, an underlying linear relationship is seen to represent the data exceedingly well over the vast majority of the range of mobility diameters. The relations $\rho_{\text{eff}}/(\text{kg m}^{-3}) = 820 - 0.64(D_{\text{mob}}/\text{nm})$ for Aquadag and $\rho_{\text{eff}}/(\text{kg m}^{-3}) = 770 - 0.53(D_{\text{mob}}/\text{nm})$ for Fullerene can be clearly seen to be quite accurate over most of the range of accumulation mode mobility diameters studied, agreeing to within 5% for mobility diameters greater than 100 nm for Aquadag and 150 nm for Fullerene, and to within approximately 1% for mobility diameters greater than 115 nm for Aquadag and 185 nm for Fullerene. For the sake of completeness, and to facilitate calculation of the effective density for lower sizes, the fits $\rho_{\text{eff}}/(\text{kg m}^{-3}) = 1490 - 10.9(D_{\text{mob}}/\text{nm}) + 0.039(D_{\text{mob}}/\text{nm})^2$ for Aquadag and $\rho_{\text{eff}}/(\text{kg m}^{-3}) = 1640 - 10.8(D_{\text{mob}}/\text{nm}) + 0.031(D_{\text{mob}}/\text{nm})^2$ for Fullerene (shown in Figure 1 by dashed lines) match those presented by the authors to within 1% for D_{mob} in the ranges 60–140 nm for Aquadag and 65–165 nm for Fullerene.

The departure of ρ_{eff} from the linear trend at small mobility diameters reflects the fact that as the size of the aggregate decreases the effective density must approach that of

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the primary particles (i.e., individual spherules) that make up the aggregate (in the limit of the 'aggregate' consisting of a single primary particle the effective density would equal that of the primary particle itself). To illustrate this point, consider a Fullerene particle comprised of primary particles of diameter 50 nm and density 1200 kg m^{-3} (the values stated in the paper). A particle with $D_{\text{mob}} = 100 \text{ nm}$ would consist of approximately 5 primary particles, and one with $D_{\text{mob}} = 70 \text{ nm}$ (the leftmost data point on Figure 4 of the paper) of only 2.4 primary particles. If it were assumed instead that the individual spherules had diameter 60 nm, the number of primary particles would be 3 and 1.4, respectively. An aggregate consisting of so few primary particles cannot attain the "wispieness" or fractal nature that larger aggregates can, and thus it is not surprising that the density deviates from the linear relationship exhibited by these larger aggregates. Additionally, it should be noted that discrete values of the mobility diameters should occur at small sizes, reflecting the small number of configurations possible for few primary particles.

Finally, we would also like to point out that the Gysel dataset offers the opportunity to examine the fractal dimension, defined as $D_f = \partial \ln m / \partial \ln D_{\text{mob}}$, for these two substances. Using the expression for mass in terms of effective density given above, this expression is equivalent to $D_f = 3 + (D_{\text{mob}}/\rho_{\text{eff}})(\partial \rho_{\text{eff}}/\partial D_{\text{mob}})$. The values of D_f calculated using the linear relations presented above are shown in Figure 2 as a function of mobility diameter. As per expectations, the fractal dimension approaches 3 at the smaller aggregate sizes, reflecting the limited clustering that can occur with few spherules, and decreases as the number of primary particles that make up the aggregate increases, reflecting the increased wispieness of these larger aggregates. (There is some anomalous behavior of the fractal dimension at mobility diameters below that at which the data deviate from the linear fit, possibly due to the few number of primary particles resulting in D_{mob} not being a continuous function, as required for the derivative in the definition of fractal dimension, but rather a discrete quantity.)

It is hoped that this brief "back-of-the-envelope" analysis will inspire the authors to

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examine additional lines of inquiry. For example, why is the effective density approximately linear in mobility diameter, and what is the criterion, in terms of mobility diameter or number of primary particles, above which this linear relation holds?

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