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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Technical note: Measurement of chemically-resolved volume equivalent diameter and effective density of particles by AAC-SPAMS" *by* Long Peng et al.

Anonymous Referee #2

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Peng et al present results from coupling an aerodynamic aerosol classifier (AAC) with a single particle aerosol mass spectrometer (SPAMS) to determine individual particle effective density and shape. While this is a worthwhile endeavor, the manuscript misrepresents the prior work on the topic (deriving effective density using a size measurement prior to a single-particle mass spectrometer, which was shown several times in the 2000s). I agree with Johannes Schneider's review and provide additional comments here. A main additional point is that the results need to include propagated measurement error reporting, in the figures and text, for accurate comparisons to theoretical/manufacturer values and to support size-dependent trends, or the lack thereof.

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As the prior review stated, deriving effective density using a size measurement prior to a single-particle mass spectrometer is not new, and this prior work needs to be discussed in a dedicated added section in the introduction to properly place the current work in context. There are statements, such as those on lines 90, 150-151, and 298-299, that give the impression that this is the first work to measure chemically-resolved particle effective density, and therefore, this phrasing needs to be revised. The lack of discussion of this prior work is surprisingly given the author's prior paper, which is cited (Zhang et al. 2016, Sci. China Earth Sci., Measurement of aerosol effective density by single particle mass spectrometry). The introduction of the current manuscript only mentions one prior paper (Moffet and Prather 2005) that derived effective density using an aerosol mass spectrometer (and that work was using scattering signals) and does not discuss prior derivations of shape factors. In addition to Zelenyuk et al. (2006) and Schneider et al. (2006) referenced in the previous review, additional work, not cited in the current paper, includes: - Murphy et al. (2004), J. Aerosol Sci., Particle density inferred from simultaneous optical and aerodynamic diameters sorted by composition - Spencer et al. (2007), Environ. Sci. Technol., Simultaneous Measurement of the Effective Density and Chemical Composition of Ambient Aerosol Particles - Slowik et al. (2007), Aerosol Sci. Technol., Measurements of morphology changes in fractal soot particles using coating and denuding experiments: Implications for optical absorption and atmospheric lifetime - Zelenyuk et al. (2008), Analytical Chem., Simultaneous measurements of individual ambient particle size, composition, effective density, and hygroscopicity - Zelenyuk et al. (2008), Environ. Sci. Technol., A new real-time method for determining particles' sphericity and density: Application to secondary organic aerosol formed by ozonolysis of a-pinene - Zelenyuk et al. (2008), J. Phys. Chem. A, "Depth-profiling" and quantitative characterization of the size, composition, shape, density, and morphology of fine particles with SPLAT, a single-particle mass spectrometer - Alexander et al. (2016), Aerosol Sci. Technol., Measurement of size-dependent dynamic shape factors of quartz particles in two flow regimes This may not be a complete list, and therefore, the authors need to do a thorough litera-

ture search. In addition to addition to the Introduction, this prior work should also be discussed/compared to in the Results & Discussion. For example, paragraph 2 on page 11 does a good job of comparing to and discussing previous literature, and the manuscript would benefit from this type of comparison and discussion at other points in the manuscript.

The manufacturer reported uncertainties on the PSL sizes need to be reported in the Methods, given the goal of measurement comparison to these values, and then they need to be included when calculating "discrepancies" with the measurements. It is quite possible that the measurement comparison is well within the expected uncertainties. Currently the sizes are stated at 0.X nm accuracy, but my understanding is that the manufactured PSLs are not this monodisperse. For example, a quick look at the manufacturer website suggests that the 203 nm PSLs are provided at +/- 5 nm. Given the direct comparison in the Results to these sizes, the reported accuracy by the manufacturer is key and needs to be reported. In addition, for the ammonium sulfate, ammonium nitrate, and ambient particles, what is the error in the size selection by the AAC? This is also critical to the method uncertainty. The AAC brochure says that size resolution depends on the sheath to sample flow ratio, so this should also be reported in the methods. Throughout the Results and Discussion text and associated figures, the full measurement uncertainty (that takes into account the width of the size distribution and that it is not monodisperse to the tenth of a nanometer, as implied by reporting values to 0.X nm) needs to be calculated and included in the difference ("discrepancy") calculations. These error bars are particularly needed in Figures 2 and 3 (similar to the inclusion in Figure 4) and in the assessment of any size dependence of effective density. The Figure 4 caption should also state the origin of the error bars shown. Further, on lines 202-203, it is stated that "...in the SPAMS [the] size calibration curve possesses the systematic error." However, this systematic error is not stated or shown (nor is it's origin explained). Further, on Line 206, it is stated that the size-dependent pattern observed is "divergent with the previous studies", but without inclusion of measurement uncertainty, any "pattern" or trend cannot be assessed. Further, often too

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many decimal places are reported in the manuscript, beyond the appropriate number of significant figures, and this should be evaluated once error is calculated.

In the methods section, Section 2.2 should be separated into laboratory experiments and ambient sampling, for improved clarity. Information should be provided about the diffusion drying tube shown in Figure 1, especially since the water content of the particles is key to the results. The description of the clustering methodology should be moved from the Results to the Methods and expanded. The location and dates of ambient sampling also need to be provided, as well as the actual number of ambient particles measured at each size selected.

Additional comments:

Lines 209-211: This sentence is not clear and makes mention of a separate paper being written on the topic of effective density and size-dependent evaluation, but it is not clear why that isn't included here or how it is different.

Line 239: By "different", do you mean "wider"?

Lines 255-256: Add a reference to this methods sentence and move to the Methods section.

Line 281 and Table 1: Note that error should only be reported with 1 significant figure.

Table 1 caption: State what the error corresponds to here.

Figure 1 caption: Add flow rates to the figure to make it more informative.

Figure S2b is missing.

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