Response to comments

Response: We thank the reviewer for his comments and recommendations to improve the manuscript. Below we have detailed the responses and resulting edits to all of the reviews' comments. The review comments are listed in italics and black, followed by our responses in normal font and blue. To make it clear, the contents in revised manuscript are presented in quotes and normal font.

Referee#1：Johannes Schneider

Specific comments：*lines 169 - 185, the discussion of the Cunningham slip correction:- please give a reference for equation (12). "A, B and C are empirically determined constants specific to the analysis system." Did you determine these constants for your system, and if so, how? What are the numbers for A, B, C that you have used?*

Response: We have added the references and the values of A, B, C. Please refer to Lines 190-194:

Lines 190-194: “The Cunningham Slip Correction Factor is calculated by Eq. (12) (DeCarlo et al., 2004):

$$C_c(D) = 1 + \frac{\lambda}{D}\left(A + B \cdot \exp\left(\frac{C \cdot D}{\lambda}\right)\right), \qquad (12)$$

where λ is the mean free path of the gas molecules, and A, B and C are empirically determined constants specific to the analysis system. The values of A, B and C are 2.33, 0.966, and -0.498, respectively, which are provided by the manual of the AAC.”

Reference

DeCarlo, P. F., Slowik, J. G., Worsnop, D. R., Davidovits, P., and Jimenez, J. L.: Particle morphology and density characterization by combined mobility and aerodynamic diameter measurements. Part 1: Theory, Aerosol Sci. and Technol., 38, 1185-1205, https://doi.org/10.1080/027868290903907, 2004.

Response to comments

Response: We thank the reviewer for his/her thoughtful suggestions and constructive criticism that have helped us further improve our manuscript. Below we have detailed the responses and resulting edits to the review's comments. The comments are listed in italics and black, followed by our responses in normal font and blue. To make it clear, the contents in revised manuscript are presented in quotes and normal font.

Referee#2: Anonymity

General Comments: Peng et al present a revision of their manuscript combining an AAC and single-particle mass spectrometry to determine chemically-resolved single-particle effective densities. As noted previously by both reviewers, this work is interesting and useful. However, both reviewers pointed to a major manuscript weakness being limited citations to previous, highly relevant work and some explanations needing clarifications. In addition, several previous comments focused on stated uncertainties and errors in the results. Several of these issues have been corrected, but some still remain, as noted below.

1) *The authors added sentences in the introduction discussing the use of particle light scattering to obtain effective density, and these added sentences on Lines 87-94 are useful. However, most previous and similar work has used a DMA with single-particle mass spectrometry, and both reviewers pointed to the need to discuss this previous work in the introduction to place the current similar work (that combines AAC with single-particle mass spectrometry) in context and make it clearer how this current work builds upon this previous work. However, the authors chose not to do this, with the exception of adding a statement that the derivation of Eq 8 was previously presented in Schneider et al (2006) (lines 82-83). I still believe it is a major weakness of the paper to not discuss previous work combining a DMA with single-particle mass spectrometry (see previous reviews) in this manuscript's introduction.*

Response: Thanks for the comments. After careful consideration, we also think the inclusion of previous work combining a DMA with single-particle mass spectrometry would be better for the intact, although it is different to our method. We have added the discussion of such work accordingly, please refer to Lines 83-94:

"A variety of methods are developed to characterize $\rho_e{}^{I}$ and $\rho_e{}^{III}$, among which the more advanced methods are to achieve the measurement of the chemically-resolved effective density. Combining a single particle soot photometer (SP2) with a (volatility) tandem differential mobility analyser ((VT)DMA) can measure the $\rho_e{}^{I}$ of particles mixed with soot (Zhang et al., 2016b; Wu et al., 2019; Han et al., 2019). The measurement of chemically-resolved $\rho_e{}^{III}$ can be achieved by coupling a DMA with an on-line aerosol mass spectrometer including the single particle laser ablation time-of-flight mass spectrometer (SPLAT I/II) (Zelenyuk et al., 2005; Zelenyuk et al., 2006; Alexander et al., 2016), aerosol mass spectrometer (AMS) (Dinar et al., 2006; Schneider et al., 2006; Kiselev et al., 2010), aerosol time-of-flight mass spectrometer (ATOFMS) (Spencer and Prather, 2006; Spencer et al., 2007), and single-particle aerosol mass spectrometry (SPAMS) (Zhang et al., 2016a; Zhai et al., 2017)."

2) *Following on the previous comment, the authors did not rephrase the problematic statements that give the impression that this is the first work to measure chemically-resolved particle effective density (previous lines 90, 150-151, 298-299, now lines 98,). These sentences include: "The aim of the present work is to develop a method to obtain Dve and pe." (line 98 in current manuscript) "These two properties cannot yet be measured for unknown particles by current techniques." (when referring to Dve and pe on lines 167-168 in the current manuscript) "…to first characterize the Dve, pe, and chemical compositions of atmospheric particles…" (lines 321-322 of the current manuscript) As previously requested, these misleading statements should be rephrased.*

Response: Sorry for the misunderstanding. The problematic statements have been rephrased. Please refer to:

a) Line 109: "The aim of the present work is to develop a method to simultaneously obtain D_{ve} and ρ_e."

b) Lines 183-184: “These two properties cannot yet be simultaneously measured for unknown particles by current techniques.”

c) Lines 336-339: “Coupled with the ability of SPAMS to characterize the chemical composition of individual particles, we conducted a sample proof of the AAC-SPAMS equipment to first simultaneously characterize the D_{ve}, ρ_e and chemical compositions of atmospheric particles, showing the potential application of this system in field observations.”

3) *Throughout the manuscript, error should be reported with one significant figure. In addition, while error was added to the PSL sizes on Lines 139-142, it is overstated. For example, 203.0 +/- 5.0 nm shows greater certainty than 203. +/- 5. nm, which would be in line with the manufacturer’s stated uncertainty. Also, in the response, the authors state that they fixed error to be reported with one significant figure in Table 1, but all of the errors shown in this table still are 2-3 significant figures.*

Response: Thanks for pointing out this. We have revised the description to “Dried spherical PSL (Nanosphere Size Standards, Duke Scientific Corp., Palo Alto) (ρ_p = 1.055 g/cm^3 and χ = 1.0) with D_{ve} values of 203 ± 5 nm, 310 ± 6 nm, 510 ± 5 nm, and 740 ± 6 nm were used in the AAC-SPAMS system, and the D_{ve} was verified by Scanning Mobility Particles Sizer (Model 3938, TSI Inc., USA).”, which is in line with the manufacturer’s stated uncertainty.

For the data shown in Table 1, as shown in the previous response, we indicated that the resolution for the measured D_{ve} values can be as small as < 1 nm, and thus we have revised the mean values and standard deviation calculated from several measurements to 1 decimal place in the revised manuscript. Note that the decimal place for the mean D_{ve} is consistent with the measurement error in the AAC-SPAMS system.

4) *The authors state in their response that “Considering the precision of the PSL size is less than that of the instrument of AAC, the discrepancy between the measured value (from AAC-SPAMS) and the true value (the size and density of PSL) is used to represent*

the measurement uncertainty, which is presented in Figure 2." However, the error in the AAC size needs to be stated in (added to) to the manuscript for the reader to evaluate this statement. It appears that these measurement uncertainties were not propagated through to the reported deviations in size and density in Figure 2, so the reader cannot evaluate where the major source of error is originating, which is important for the results. The authors at least need to state the errors in the manuscript to justify their method. It is also critical that these uncertainties be clearly explained so that the number of decimal places used in presenting the data in the results can be properly evaluated.

Response: We agree with the comment that it is critical to evaluate the measurement error accurately. Firstly, we have added the measurement uncertainty of the AAC. Please refer to Lines 157-161:

"Besides, to present the measurement uncertainty of the AAC, the D_a values of these PSL particles were measured to be 212.8 ± 0.2, 324.7 ± 0.4, 529.9 ± 0.4, and 767.5 ± 0.4, respectively, by the system of AAC- Condensation Particle Counter (CPC), which shows that the AAC has the deviations of 1.1%, 1.3%, 0.8%, and 0.7% for determining the D_a values of the particles.".

Then, the uncertainties for the Gaussian fitting to obtain the peak D_{va} for PSL was estimated to be of 6.6%, 4.4%, 2.3% and 2.2%.

Finally, we calculated the deviations between the theoretical D_{ve} ($D_{ve,th}$) and $D_{ve,me}$, which is < 1% and <6%, respectively. And thus we may conclude that the errors from AAC (~1%) and fitting of D_a (~2-7%) should explain the errors of $D_{ve,me}$ (< 1%) and $\rho_{e,me}$ (<6%) measured by the AAC-SPAMS system, as discussed in section 3.1.

5) *The ambient sampling section of the Methods (Sec 2.3) needs to state the dates of sampling, as previously requested, since the seasonality will impact particle composition, for example.*

Response: Thanks for the comment. We have added the dates of sampling. Please refer to Lines 172-173:

"The sampling time for the particles of each D_a was approximately 10 minutes. From July 6th to 8th, 2019, approximately 129,869 ionized particles were obtained from nine rounds of measurement."

6) *Lines 304-306: Errors were reported for all particle types in the previous version of the manuscript, but now these are missing for the Amine, EC-N-S, and EC-S particle types for an unknown reason.*

Response: Thanks for pointing out this mistake. They have been added. Please refer to Lines 320-325:

"Additionally, the average ρ_e of each type of particle is in the order from small to large: 1.2 ± 0.2 g/cm^3 for OC-EC-N-S, 1.3 ± 0.2 g/cm^3 for OC-N-S, 1.4 ± 0.1 g/cm^3 for K-rich, 1.4 ± 0.1 g/cm^3 for Amine, 1.5 ± 0.1 g/cm^3 for EC-N-S, 1.5 ± 0.1 g/cm^3 for EC-S, 1.6 ± 0.1 g/cm^3 for K-Na and 1.6 ± 0.1 g/cm^3 for Metal-rich. It is reasonable to find that the average ρ_e of internally mixed particles distributes in the range of their material densities (ρ_m)."

7) *Consistency between Methods and Figure 1 caption: It is now stated that the system flow rate is 0.3 lpm, but line 153 states an "overall sampling flow of 3 lpm". Line 124 states "The exhaust flow from the AAC was about 0.3 lpm." However, the sampling flow rates of the AAC and SPAMS are not stated in Section 2.1. Please add these to Section 2.1, clarify the flow rates at each part of the system in Figure 1, and clarify the discrepancy between the figure caption and line 153.*

Response: Thanks for pointing out this, which is due to our incorrect description in the *Methods*. As stated in the figure caption, the sampling flow in the system of AAC-SPAMS is 0.3 lpm, which mean that the sampling flow from AAC to SPAMS was 0.3 lpm. The overall sampling flow of 3 lpm refers to the sampling flow for the $PM_{2.5}$ cyclone inlet. To reduce the residence time of particles in the conductive silicone, a diaphragm pump was connected in parallel with the diffusion drying tube and ran at a flow rate of 2.7 lpm, which was not present in Figure 1. We have revised the description to “The sampling flow from the $PM_{2.5}$ cyclone inlet was 3 lpm, and the residence time in the conductive silicone tube was approximately 5 seconds.”, please refer to Lines 168-170.