

Response to comments

Thanks to the editor for his unremitting efforts in processing and reviewing our manuscript, which helps to improve the manuscript. We have made appropriate revisions accordingly in the revised manuscript to address the editor's concerns, and our explanations of such revisions could be found in the uploaded files.

Specific Comments

1) The authors have addressed some of reviewer 2's comments, but some of the key problems remain. Specifically, the authors still make statements that could be interpreted as saying that this is the first technique to determine D_{ve} and P_{eff} , which is simply not correct because people have been determining these parameters using different combinations of sizing metrics (e.g. D_m , D_a , D_{ve} , M) for decades. As requested, the authors have cited the previous works based on DMA-SPMS measurements, but they still need to more explicitly compare what their combination of measurements offers over previously published techniques. This is a key requirement when demonstrating the novelty of the work.

Response: Thanks for the comments. We agree that D_{ve} and effective density could be determined by various methods. However, there are three definitions of ρ_e used in atmospheric science. The methods of DMA-SP2 and DMA-SPMS are developed to measure the ρ_e^I and ρ_e^{III} , respectively, while the AAC-SPAMS in this study is developed to measure the ρ_e^{II} . The ρ_e^I and ρ_e^{III} are demonstrated to have the inherent characteristics of decreasing with increasing particle size, while ρ_e^{II} is independent of particle size. To our best knowledge, there is no appropriate technique to achieve the measurement of ρ_e^{II} for aspherical particles, so the novelty of this present work is that we developed the AAC-SPAMS system to first achieve the measurement of ρ_e^{II} for aspherical particles. To make it clear, we have revised the sentence to be specific to ρ_e^{II} clarified this in the revised manuscript. Please refer to Lines 97-118 in the revised manuscript.

Lines 97-118:

“However, the ρ_e^I and ρ_e^{III} are demonstrated to have the inherent characteristics of decreasing with increasing particle size, which will be presented in a separate publication. Therefore, it will introduce systemic error when assessing the particle impacts on visibility, human health and climate change from the physical quantities in ρ_e^I and ρ_e^{III} . In contrast, ρ_e^{II} is independent of particle size. For example, for soot particles with χ of 2.5 and ρ_p of 1.80 g/cm³, the calculated ρ_e^I , ρ_e^{II} , and ρ_e^{III} are 0.43, 0.72, and 0.45 g/cm³ at D_m of 40 nm, and 0.22, 0.72, and 0.36 g/cm³ at D_m of 550 nm, respectively. The big gap between the three definitions of effective density suggests that they should be carefully treated when characterizing the particles. However, the ρ_e^{II} has not been widely applied in atmospheric sciences because of the lack of measurement techniques. Previous literatures tried to retrieve the ρ_e^{II} and the real part in the refractive index (n) through a fitting procedure that compares the measured light-scattering intensity of particles (R_{meas}) to the theoretical values ($R_{theory,test}$) calculated by a series of n and ρ_e^{II} values (Moffet and Prather, 2005; Moffet et al., 2008; Zhang et al., 2016a). Moffet and Prather (2005) successfully obtained ρ_e^{II} for spherical particles by single particle mass spectrometry. However, subject to the accuracy of Mie theory for the aspherical particles, dry NaCl and calcium-rich dust particles were failed to fit the $R_{theory,test}$ well to R_{meas} (Moffet et al., 2008). Similarly, Zhang et al. (2016a) failed to simultaneously retrieve ρ_e^{II} and n for (NH₄)₂SO₄ and NaNO₃ particles. To our best knowledge, there is no appropriate technique to achieve the measurement of ρ_e^{II} for aspherical particles.”

In this study, we only focus on ρ_e^{II} . For simplicity, the symbol ρ_e in “Experimental section” and “Results and discussion” refers to the definition of ρ_e^{II} . We have clarified in Lines 120-121. In order to avoid ambiguity, we emphasized the definition of the ρ_e^{II} in the Abstract and Conclusion. Please refer to Lines 21-24 and 329-331.

Lines 21-24: “Three definitions of ρ_e are generally used to characterize the physical property of a particle as an alternative to particle density, in which only the ρ_e^{II} , defined

as the ratio of particle density (ρ_p) to a dynamic shape factor (χ), has the characteristic of being independent of particle size.”

Lines 329-331: “We develop an AAC-SPAMS system to first achieve the measurement of the D_{ve} and ρ_e (defined as the ratio of ρ_p to χ) of the aspherical particles through characterizing their D_a and D_{va} .”

2) In addition, the use of the term "proof" is inappropriate, because in the context of instrumentation, this implies a real-world application has been independently verified. "Demonstrated" would be more appropriate.

Response: We agree with the comment. We have rephased the statement as suggested. Please refer to Lines 334-338: “Coupled with the ability of SPAMS to characterize the chemical composition of individual particles, the AAC-SPAMS system was demonstrated to be capable of characterizing the D_{ve} , ρ_e (ρ_p/χ) and chemical compositions of atmospheric particles simultaneously, showing the potential application of this system in field observations.”

Besides, we further polished the language of the manuscript.