

Response to comments

Response: We thank the reviewers for thoughtful suggestions and constructive criticism that have helped us improve our 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. Reference to line numbers are added to the revised manuscript.

Referee#1: Johannes Schneider

General Comments: *This technical note is appropriate for ACP. The combination of single particle results (via the clustering algorithms), effective density and shape factors is interesting and is worth to be exploited further. However, some important comments have to be addressed before this paper can be published:*

Specific comments:

1) *Both ZEL2006 and SCH2006 used the mobility diameter measured by a differential mobility analyzer (DMA) and not the AAC that measures the aerodynamic diameter, but the basic idea is the same. In SCH2006, we already presented the equation for ρ_{eff_II} (your Equ. 8). Thus, please give proper credit to our work.*

Response: Thanks for your suggestion. We have included the citation (Schneider et al., 2006) as suggested. Please refer to Lines 82-83: "The detailed derivation of Eq. (8) was presented in Schneider et al. (2006)."

2) *Lines 68-76: You rely here on DeCarlo et al., 2004, but the numbering of the effective density is different. ρ_{e_I} is the same, but you changed ρ_{e_II} and ρ_{e_III} compared to DeCarlo et al. Please use the same numbering to avoid confusion. Please also refer to Hand et al., 2002 who introduced your ρ_{e_III} (which in de Carlo et al. is termed ρ_{eff_II}).*

Response: Thanks for your suggestion. To avoid confusion, we change the numbering of the definitions of the effective density based on DeCarlo et al., 2004. Besides, we refer Hand and Kreidenweis (2002) to the definition of ρ_e^{II} . Please refer to Lines 68-76: “At present, three definitions of ρ_e are introduced in atmospheric science (DeCarlo et al., 2004): the first definition (ρ_e^I) is the ratio of the measured particle mass (m_p) to the particle volume (V) calculated assuming a spherical particle with a diameter equal to the measured D_m ; the second definition (ρ_e^{II}) is the ratio of ρ to χ (Hand and Kreidenweis, 2002); and the third definition (ρ_e^{III}) is the ratio of D_m and D_{va} , all of which are expressed in Eqs. (4)-(6), respectively. ”

$$\rho_e^I = \frac{6m_p}{\pi D_m^3} \quad (4)$$

$$\rho_e^{II} = \frac{\rho_p}{\chi} \quad (5)$$

$$\rho_e^{III} = \frac{D_{va}}{D_m} \rho_0 \quad (6)$$

3) *Line 80, equation 8: rho_0 is wrong here, needs to be deleted to get the units right.*
 Response: Thanks for your comment. It has been corrected accordingly.

4) *Line 81: "The detailed derivation will be presented in a separate paper". The derivation of Equ. 8 was given in deCarlo et al., 2004, and also in SCH2006, so please give proper reference here, and for completeness, give the derivation of Equ. 7 here as well.*

Response: Thanks for your comment. The citation and derivation of Eq. 7 have been added accordingly. Please refer to Lines 81-83: “The Eq. (7) is derived from combining the Eq. (1) with Eq. (4) in which m_p is equal to $1/6 \rho \cdot D_{ve}^3$. The detailed derivation of Eq. (8) was presented in Schneider et al. (2006).”

5) *Line 157, Eq. 11: To calculate D_ve, you need the Cunningham slip correction values here. How are they obtained? The differences between D_ve and D_a (e.g. Fig. S2) are rather large, so the Cunningham correction can not be neglected.*

Response: We agree with the comment. Actually, the Cunningham Slip Correction

Factor was not neglected in this study. The calculating process of the D_{ve} is presented in the Section 2.3, please refer to Lines 169-182:

“Combining Eqs. (2) and (3), we obtain the following Eq. (10):

$$C_c(D_a) \frac{D_a^2}{D_{va}} = D_{ve} C_c(D_{ve}) \frac{\chi_v}{\chi_t} \quad (10)$$

Based on the approximation between χ_v and χ_t ($\chi_v \approx \chi_t = \chi_a$) (DeCarlo et al., 2004), Eq. (10) becomes Eq. (11):

$$C_c(D_a) \frac{D_a^2}{D_{va}} = D_{ve} C_c(D_{ve}) \quad (11)$$

The Cunningham Slip Correction Factor is calculated by Eq. (12):

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

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. Substituting Eq. (12) into Eq. (11) obtains the Eq. (13).

$$\frac{D_a^2}{D_{va}} + \frac{D_a \cdot \lambda}{D_{va}} \left(A + B \cdot \exp\left(\frac{C \cdot D_a}{\lambda}\right) \right) = D_{ve} + \lambda \left(A + B \cdot \exp\left(\frac{C \cdot D_{ve}}{\lambda}\right) \right) \quad (13)$$

Thus if the D_a and D_{va} of an unknown particle are measured, its D_{ve} could be calculated according to Eq. (13).”

6) Line 161: I think that Equ. 12 results from combining Equ. 3 and Equ. 6. Correct?

Response: Correct, and it has been clarified based on the above comments. Please refer to Lines 182-184: “Finally, the ρ_e value of the particles is calculated by the D_{va} and D_{ve} values according to Eq. (14), which is obtained by combining Eq.(3) and Eq.(5): ”

$$\rho_e = \frac{\rho_p}{\chi_a} = \frac{D_{va}}{\rho_0 \cdot D_{ve}} \quad (14)$$

7) Line 180 – 182: Please clarify that for spherical particles like PSL, $\rho_e = \rho$ (see deCarlo et al., Equ [43] and thereafter).

Response: It has been clarified accordingly. Please refer to Lines 204-206: “The deviations of $\rho_{e,me}$ are determined to be 4.3%, -5.2%, -5.2%, and 4.3%, respectively, by comparing to the theoretical ρ_e ($\rho_{e,th}$) that is equals to the ρ_p for the spherical particles (i.e. 1.055 g/cm³ of PSL particles).”

8) Line 206-209: *If different definitions of the effective densities are used, the statement "This pattern is divergent with the previous studies, which showed that effective density decreased as the size increasing" has to be removed or at least reworded.*

Response: Thanks for your comment. The statement has been reworded, please refer to Lines 235-237: "It is determined by the definition of effective density used in this study, which keeps constant as long as the χ_a does not change with particle size for pure compounds."

9) Line 259-260: *Please refer to the respective Figures in the Supporting Information.*

Response: It has been clarified accordingly. Please refer to Lines 282-284: "Details of the chemical composition and number fraction of the eight types of particles are presented in the Figures S3 and S4, respectively, which are discussed in the Supporting Information."

10) Line 275-280: *$\rho_e = \rho_p / \text{shape factor}$ (Equ. 6). Thus, either the density of the particle material is a function of size, or the particle shape factor (or both). I think you can not rule out that the material density changes with size. SPMS is not quantitative, so particles of the same cluster type may have different quantitative composition (e.g. the ratio OC/EC or organic/inorganic). Thus, you can't tell whether the changing ρ_e with size is an effect of shape or composition*

Response: We agree with the comment. They have been revised to Lines 302-304: "Specifically, the ρ_e of K-Na increases with D_{ve} , while the ρ_e of OC-N-S and OC-EC-N-S decreases with D_{ve} , which may be influenced by the particle shape and/or the material density."

11) *Supplement: Fig.S2b) is missing.*

Response: Thanks for your comment. Sorry for this mistake. Fig. S2 do not include the Fig. S2b, so we delete the description of Fig.S2b in the caption. Please refer to Lines 27-28 in Supporting Information.