

Response to Anonymous Referee #2

We thank the reviewer for the constructive suggestions and comments concerning our manuscript entitled “Characterization of tandem aerosol classifiers for selecting particles: implication for eliminating multiple charging effect” (ID: amt-2021-436). Those comments are valuable and very helpful for improving our paper, as well as the important guiding significance to our studies. Below, we provide a point-by-point response to individual comment (Reviewer comments in italics, responses in plain font; page numbers refer to the AMTD version)

1. Comments and suggestions:

Line 88, Eq. (3): The equation for the transfer function of the DMA in non-diffusing case is not given in the general form. Eq. (3) is only correct when aerosol inlet and aerosol sampling flow rates are equal (i.e. the DMA is operated in balanced flow mode), leading to $\delta = 0$ in the general form of DMA transfer function [Eq. (7) in Stolzenburg and McMurry (2008)]. This information should be noted in the paper.

Responses and Revisions:

Thank you for your comment. We have changed it in the revised manuscript:

“Assuming that aerosol inlet and aerosol sampling flow rates are equal, the transfer function of DMA can be expressed as follows when particle diffusion is negligible”.

2. Comments and suggestions:

Line 139: The primary reference for the miniature inverted soot generator is as follows, which should be cited in the text: Kazemimanesh, M., Moallemi, A., Thomson, K., Smallwood, G., Lobo, P. and Olfert, J.S., 2019. A novel miniature inverted-flame burner for the generation of soot nanoparticles. Aerosol Science and Technology, 53(2), pp.184-195.

Responses and Revisions:

Thank you for the reminder. We have added it in the text.

“Soot particles were generated by a miniature inverted soot generator (Argonaut Scientific Ltd., Canada) with a propane flow of 74.8 SCPM and an air flow rate of 12 SLPM. Although this operation setting is not in the open-tip flame regime, the flame is open-tip consistent with Fig. 2d in Moallemi et al. (2019). Detailed aerosol generation methods can be found in Kazemimanesh et al. (2019b) and Moallemi et al. (2019)”

3. Comments and suggestions:

Line 138-140: To reproduce the experiments in this study, it is necessary to give details of propane and air flow rates used in the soot generator.

Responses and Revisions:

Thank you for your reminder. We have changed it in the revised manuscript:

“Soot particles were generated by a miniature inverted soot generator (Argonaut Scientific Ltd., Canada) with the propane flow of 74.8 mL min⁻¹ and the air flow rate of 12 L min⁻¹. Although this operation setting is not in the open-tip flame regime, the flame is open-tip consistent with the Fig. 2d in Moallemi et al. (2019)”.

4. Comments and suggestions:

Lines 145-155: For one or two mobility-selected particles, please add representative plots for the measured spectral density of mass ($dN/d\log m_p$) and aerodynamic diameter ($dN/d\log d_{ae}$), either in the paper or in the supplementary material.

Responses and Revisions:

The measured spectral density of mass ($dN/d\log m_p$) and aerodynamic diameter ($dN/d\log d_{ae}$) for particles with d_m of 150 nm and 250 nm have been added in the supplementary material.

5. Comments and suggestions:

Lines 198-199: It states that the ability of DMA-CPMA to eliminate multiply charged particles depends on the resolutions of both DMA and CPMA; however, dependence on the resolution of CPMA is not obvious from Eq. (25). Can the authors clarify this?

Responses and Revisions:

Thank you for your comment. The resolution can be calculated by

$$R_m = \frac{m_1}{m_{1,max} - m_1} = \frac{2\pi B_{1,max} L_{CPMA} \tau_c^2 \omega^2 m_1}{Q_{CPMA}},$$

We have changed the Eq. (25) to

$$D_{fm} > PP_0 = \frac{\log(m_{2,max}/m_1)}{\log(d_{m2,min}/d_{m1})} = \frac{\log(2 + \frac{1}{R_m(1+\beta_{DMA})})}{\log\left(\frac{2}{(1+\beta_{DMA})} \frac{Cc(d_{m2,min})}{Cc(d_{m1})}\right)}$$

in the text and the relationship between the slope of PP_0 and the resolution is more obvious.

6. Comments and suggestions:

Line 201: The sentence states that a larger β is necessary to reduce the potential of multiply-charged particles, but it seems that Eq. (25) shows the opposite. Can the authors double-check this?

Responses and Revisions:

Thank you for your comment. $(1 - \beta_{DMA})$ has been replaced by $(1 + \beta_{DMA})$. We have revised the Eq. (25) to

$$D_{fm} > PP_0 = \frac{\log(m_{2,max}/m_1)}{\log(d_{m2,min}/d_{m1})} = \frac{\log(2 + \frac{1}{R_m(1+\beta_{DMA})})}{\log\left(\frac{2}{(1+\beta_{DMA})} \frac{Cc(d_{m2,min})}{Cc(d_{m1})}\right)}$$

7. Comments and suggestions:

Line 303: In Fig. 5c and all related figures, the unit used incorrectly for absorption cross section is Mm^{-1} , which is the unit for absorption coefficient ($length^{-1}$). The correct unit for absorption cross section should be $m^2 particle^{-1}$ (derived from absorption coefficient/particle number concentration).

Responses and Revisions:

Thank you for the comment. The Y axis is the absorption coefficient α_{abs} (Mm^{-1}). We have revised it in the Fig. 5c and 6c. The text has also been changed accordingly.

8. Comments and suggestions:

Lines 306-310: It is not very clear to me why multiple charging effects due to the use of DMA-CPMA would affect the MAC or DRF of soot particles. As far as I understand, global climate models consider a specific (mostly constant) MAC value for black carbon particles to estimate their DRF, without regard for multiple charging. Unless the authors are claiming that the MAC values used in current climate models

are grossly incorrect.

Responses and Revisions:

Sorry we didn't make it clear. Previous studies used DMA-APM or DMA-CPMA to investigate the mass-specific MAC (Radney et al., 2013; Zangmeister et al., 2018). Our study tried to illustrate that multiple charging effect can affect classification of DMA-CPMA and the measured value of MAC. If using the measured MAC to evaluate the DRF of fresh soot particles, it can cause uncertainties.

9. Comments and suggestions:

Introduction and discussion section: There are a few recent studies that have looked at tandem measurements of mobility diameter, mass, and aerodynamic diameter to study the effective density and shape factor of spherical and non-spherical particles. These studies have used a combination of DMA, AAC, and APM or CPMA and, in my view, are relevant to this paper and should be mentioned in the introduction and their results discussed where necessary:

Yao, Q., Asa-Awuku, A., Zangmeister, C.D. and Radney, J.G., 2020. Comparison of three essential sub-micrometer aerosol measurements: Mass, size and shape. Aerosol Science and Technology, 54(10), pp.1197-1209.

Kazemimanesh, M., Rahman, M.M., Duca, D., Johnson, T.J., Addad, A., Giannopoulos, G., Focsa, C. and Boies, A.M., 2022. A comparative study on effective density, shape factor, and volatile mixing of non-spherical particles using tandem aerodynamic diameter, mobility diameter, and mass measurements. Journal of Aerosol Science, 161, p.105930.

Responses and Revisions:

Thank you for your suggestion. We have added these literatures in the introduction (Line 72 and Line 73) and discussion (Line 265)

Editorial and technical corrections

10. Comments and suggestions:

Line 272: I cannot find Eq. (30) in the paper.

Responses and Revisions:

Sorry for our mistake. We have revised it to Eq. (16).

11. Comments and suggestions:

Line 301: Change the sentence to "Subsequently, absorption cross section, σ_{abs} , was derived using the absorption coefficient and total number concentration of particles with different charging states."

Responses and Revisions:

We have changed it in the revised manuscript:

"Subsequently, absorption coefficient, α_{abs} , was derived using the Mie theory and $PNSD_{ve}$ of particles with different charging states."

12. Comments and suggestions:

Lines 308-310: This sentence is written very poorly (huge amount? huge error?). Please rephrase this sentence and avoid ambiguous adjectives.

Responses and Revisions:

We have changed it in the revised manuscript:

“A large amount of 70 nm -90 nm soot particles was emitted from a diesel engine (Wierzbicka et al., 2014), and neglecting the multiple charging effect in the measurement of mass-specific MAC on this size range will result in significant bias in the estimation of radiative forcing of automobile-emitted soot particles”.

13. Comments and suggestions:

Line 312: Reference to Table 3 should be given in the earlier paragraph (perhaps in line 302).

Responses and Revisions:

Thank you for the comment. Table 3 now is given in line 374.

14. Comments and suggestions:

Grammar mistakes.

Responses and Revisions:

Grammar mistakes have been corrected.