

## ***Interactive comment on “Dynamic shape factor and mixing state of refractory black carbon particles in winter in Beijing using an AAC-DMA-SP2 tandem system” by Xiaole Pan et al.***

**Anonymous Referee #2**

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In this paper, the authors investigate the dynamic shape factor and the mixing state of rBC-containing particles in Beijing using a system including an aerodynamic aerosol classifier, a differential mobility analyzer, and a single particle soot photometer. They conclude their study with a series of T-matrix optical simulations. The topic is certainly of interest and worth publishing; however, the paper is not always clear, and some aspects require some more explanation and detail. I think the paper could maybe be published but only after significant improvements/clarifications are made to address the comments from all the reviewers.

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GENERAL COMMENTS 1. More details need to be provided on the optical model, at a minimum the exact number of monomers, how their diameters were chosen, the index of refraction values used, etc.

2. More discussion should be provided on the accuracy and precision of the chi parameter, how these accuracies and precisions are calculated/estimated, and how significant (in a statistical sense) are the differences discussed in the paper.

3. Tense consistency and grammar should be checked in the entire paper.

4. The bibliography is OK, but maybe a bit biased and limited. Some additional references that might be useful (I am not suggesting the authors should cite all of them, I let them judge the relevance) to the paper discussion are added at the end of this document.

SPECIFIC COMMENTS Line 22: “aerosol particles” include all particles or only rBC-containing particles?

Lines 27-28: Maybe I am misreading this sentence, but why would condensation result in larger rBC cores? And why clean conditions would result in more coagulation processes and therefore larger particles. I would expect the opposite (more pollution should result in a higher likelihood of coagulation and therefore larger sizes). Also - a clarification - the assertion “smaller rBC core” refers to D<sub>mev</sub> from the SP2, correct?

Line 28: Does the chi value refer to all particle or only the rBC-containing particles?

Line 43: In general BC particles can also be mixed through (or be injected into) the upper troposphere above the PBL (for example by large fires).

Line 67: Also photoacoustic and extinction-minus-scattering techniques.

Lines 68-69: The term soot is also often used in microscopy.

Line 82: To enhance clarity for a broader readership, it might be good to briefly define the “lensing effect”.

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Line 103: Electron or x-ray microscopy in general, not just transmission.

Line 108: By definition, one would think that for a spherical particle, chi should be exactly one.

Line 197: Please define the setpoint Rs.

Line 199: Consider revising “at a scanning mode”

Line 219: “. . .the same as that of a spherical particle” of what size? Equation (9) line 239: The subscript BC here indicates BC or r-BC? Also, as written the equation seems to imply that  $D_{mob}=D_{ve}$ . . . is a chi missing?

Line 271: Maybe consider revising as “the tandem DMA-AAC system was experimentally tested”

Line 296: Is this the Dmev of rBC particles or of any particle? This is confusing to me especially in view of the comment in lines 298 and 300. (See also one of the previous comment).

Lines 300 – 303: As mentioned above, I do not clearly understand this argument. rBC indicates the refractory part of the particle which should identify with the core of the particle, so how would the core grow due to condensation? Even more confusing to me is the sentence in lines 307 - 308: how would water-soluble inorganic matter change the rBC core mass? I can understand that could change the rBC core shape to more compact, but the mass equivalent diameter should not change in the compaction process (it is just a reorganization of the monomer location).

Line 310: How large are the uncertainties on chi? Are these variations significant (in a statistical sense)? An uncertainty is mentioned in line 322 for that value of chi, but I do not recall any discussion on what that uncertainty represents and how was it estimated.

Line 312: What is an “In-BC” core?

Lines 327-329: I am not clear why coagulation would result in higher chi. To me, it

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makes more sense what suggested in the following sentences on diesel heavy-duty vehicle contributions (at night) and the effect of photochemical processing (during the day).

Lines 349 – 353: I don't follow the argument here. It might help some further discussion or clarification.

Line 390: MR indicates what? The mass ratio of core to shell?

Line 396: Is “exponentially” meant to actually represent an analytical exponential increase, or just to mean qualitatively a large increase?

Lines 401 – 403: Those two numbers (1.71 +/- 0.05 and 1.56 +/- 0.15) appear not to be significantly different within the uncertainties provided.

Line 414: The authors probably meant to write “fractal” not “fractional” (?)

Line 415: “were reported” by who?

Lines 417-418: I am not clear what the authors mean by “a faster decreasing tendency”, decreasing with what?

Line 420: Just a clarification, is the effective density here referring to the entire rBC-containing particle or only to the rBC (core)?

Line 421-424: Does this refer to a result from the authors or from the literature? It is not clear to me what the authors refer to.

Line 442: “to present as aggregation. . .” should maybe be “to be present as aggregated. . .”, or “to be present as aggregation of. . .”? Or something similar.

Line 436: Missing “of” in front of “climate effect”? Or alternatively, “Quantification” should maybe be “Quantifying”

Line 449: Is this “simultaneity” an assumption, or an actual observation? If an observation, based on what?

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Line 459-460: Could the authors please elaborate a bit more on the “novel aggregate model” and provide some detail? What’s novel about? There has been a lot of published work on this topic (see some limited example in the reference below)

Line 462: Maybe “sensitive” should be “sensitivity”?

Line 470: What’s the reason for this diameter choice, from the literature? If so, please cite some published work. If not, please justify.

Line 473: I thought (I) indicates fresh particles and (II) indicates thinly coated, as from lines 462 – 463? Please clarify. Also, how much coating is on the thinly coated particles, and how much on the thickly coated particles in your model?

Line 502: “reconstruction” or “restructure”?

Line 513: “was” should be “were”

Line 533: “stale” should be “stable”?

Line 535: “novel” in what sense? There have been several studies (experimental as well as numerical) using similar models and mixing configurations.

Lines 520-522: I am still confused by this assertion (see previous comments on this topic)

Figure 2, 4, 5 and 7: Consider using the symbol for chi as in the text instead of DSF for the plot axis title to avoid confusions.

Figure 4: What is the top plot, a residual plot? Please provide an axis label and explain in the caption.

Figure 5: In the caption, explain the meaning of the colored bands.

Some relevant publications: 1. Cross, E.S., T.B. Onasch, A. Ahern, W. Wrobel, J.G. Slowik, J. Olfert, D.A. Lack, P. Massoli, C.D. Cappa, J.P. Schwarz, J.R. Spackman, D.W. Fahey, A. Sedlacek, A. Trimborn, J.T. Jayne, A. Freedman, L.R. Williams, N.L.

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Ng, C. Mazzoleni, M. Dubey, B. Brem, G. Kok, R. Subramanian, S. Freitag, A. Clarke, D. Thornhill, L.C. Marr, C.E. Kolb, D.R. Worsnop, and P. Davidovits, Soot Particle StudiesInstrument Inter-ComparisonProject Overview. *Aerosol Science and Technology*, 2010. 44(8): p. 592-611.

2. Moffet, R.C., R.E. O’Brien, P.A. Alpert, S.T. Kelly, D.Q. Pham, M.K. Gilles, D.A. Knopf, and A. Laskin, Morphology and mixing of black carbon particles collected in central California during the CARES field study. *Atmos. Chem. Phys.*, 2016. 16(22): p. 14515-14525.

3. China, S., B. Scarnato, R.C. Owen, B. Zhang, M.T. Ampadu, S. Kumar, K. Dzepina, M.P. Dziobak, P. Fialho, J.A. Perlinger, J. Hueber, D. Helmig, L.R. Mazzoleni, and C. Mazzoleni, Morphology and mixing state of aged soot particles at a remote marine free troposphere site: Implications for optical properties. *Geophysical Research Letters*, 2015. 42(4): p. 1243-1250.

4. Jacobson, M.Z., Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. *Nature*, 2001. 409(6821): p. 695-697.

5. Zangmeister, C.D., J.G. Radney, L.T. Dockery, J.T. Young, X. Ma, R. You, and M.R. Zachariah, Packing density of rigid aggregates is independent of scale. *Proceedings of the National Academy of Sciences*, 2014.

6. Buseck, P.R., K. Adachi, A. Gelencsér, É. Tompa, and M. Pósfai, ns-Soot: A Material-Based Term for Strongly Light-Absorbing Carbonaceous Particles. *Aerosol Science and Technology*, 2014. 48(7): p. 777-788.

7. Petzold, A., J.A. Ogren, M. Fiebig, P. Laj, S.M. Li, U. Baltensperger, T. Holzer-Popp, S. Kinne, G. Pappalardo, N. Sugimoto, C. Wehrl, A. Wiedensohler, and X.Y. Zhang, Recommendations for reporting “black carbon” measurements. *Atmos. Chem. Phys.*, 2013. 13(16): p. 8365-8379.

8. China, S., N. Salvadori, and C. Mazzoleni, Effect of Traffic and Driving Character-

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istics on Morphology of Atmospheric Soot Particles at Freeway On-Ramps. *Environmental Science & Technology*, 2014. 48(6): p. 3128-3135.

9. Saliba, G., R. Subramanian, R. Saleh, A.T. Ahern, E.M. Lipsky, A. Tasoglou, R.C. Sullivan, J. Bhandari, C. Mazzoleni, and A.L. Robinson, Optical properties of black carbon in cookstove emissions coated with secondary organic aerosols: Measurements and modeling. *Aerosol Science and Technology*, 2016. 50(11): p. 1264-1276.

10. Zanatta, M., P. Laj, M. Gysel, U. Baltensperger, S. Vratolis, K. Eleftheriadis, Y. Kondo, P. Dubuisson, V. Winiarek, S. Kazadzis, P. Tunved, and H.W. Jacobi, Effects of mixing state on optical and radiative properties of black carbon in the European Arctic. *Atmos. Chem. Phys.*, 2018. 18(19): p. 14037-14057.

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16. Chakrabarty, R.K., H. Moosmüller, W.P. Arnott, M.A. Garro, J.G. Slowik, E.S. Cross, J.-H. Han, P. Davidovits, T.B. Onasch, and D.R. Worsnop, Light scattering and absorp-

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