

A point-by-point response

Dear Editor,

We are very pleased to submit a revised manuscript entitled with “The density of ambient black carbon retrieved by a new method: implications to CCN prediction” for possible publication in journal of ACP.

We'd like to thank you for your efforts and time on handling the paper. We also would like to thank the reviewers for their valuable comments and suggestions, all of which have been considered carefully during the revision (a point-by-point response to reviewers as follows). We believe all the comments from the reviewers have been addressed, and the paper have been greatly improved after the revision.

Yours sincerely,

Fang Zhang

On behalf of all authors

Comments from the editor:

The revised manuscript has been improved a lot. However, I would suggest that the authors address the following issues raised by the reviewer: 1. please clearly state the reason and uncertainty of the assumptions in the deriving of the effective density; 2. the chemical components of the non-BC component and BC coating should be different, the uncertainties should be discussed; 3. the number fraction of the BC-containing particles in the ambient particle is (less than 25%) low, so is it reasonable to attribute the uncertainties of the CCN number concentration to the BC density;

Re: Thanks for the editor's constructive comments and suggestions.

Regarding to the first concern about the reason and uncertainty of the assumptions in the deriving of the effective density, we have addressed this issue carefully in section 2.3 of the revised paper, or as follows:

2.3 Uncertainties and limitations

For the retrieval, the assumptions on the values of κ_{SOA} , ρ_{POA} , ρ_{SOA} and $\rho_{\text{Ex-BC}}$ as well as the fraction of primary organic aerosols in non-hygroscopic mode would add uncertainty in the inferred values of ambient internally mixed BC density. For example, the freshly emitted POA particles might consistently be coated with secondary particles during the aging process, resulting in changes of the $NF_{\text{NH-POA}}$. However, a real-time variation of the $NF_{\text{NH-POA}}$ is not yet available due to the lack of such measurement data.

Applying only rough fractions of hydrophobic POA for three different atmospheric conditions could still cause uncertainties. Also, the densities of POA and SOA may differ due to their precursors, emission sources and the formation mechanisms in ambient atmosphere (Alfarra et al., 2006; Reyes-Villegas et al., 2018). The density of Ex-BC is generally characterized by the morphology and size (Wu et al., 2019). In addition, the value of κ_{SOA} spans largely due to the variations in the emissions of gas precursors and formation processes under different atmospheric conditions (Zhang et al., 2015; Liu et al., 2021b). Therefore, we examined the sensitivities of In-BC density to the variations of these factors, as exhibited in Fig. 1 and Fig. 2.

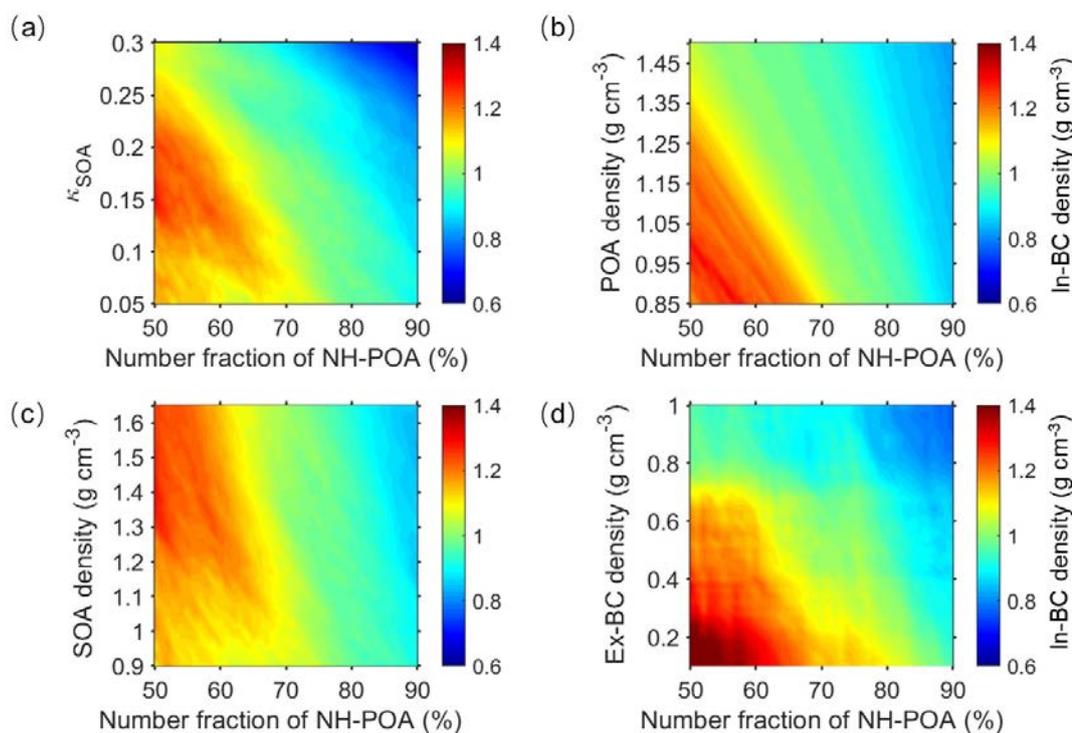


Figure 1. Sensitivities of In-BC density to the variations in the number fraction of nearly hydrophobic (NH) POA and hygroscopic parameter of OA (κ_{SOA}) (a), POA

density (b), SOA density (c) and the externally mixed BC density (d).

The figures show that the In-BC density gradually decreased with the increment of the NF_{NH-POA} , implying the high fraction of bare POA particles corresponded to the early aging stage of aerosol particles. With the increase of κ_{SOA} , the In-BC density was generally reduced, but with small fluctuations (Fig. 1a, Fig. 2b). This suggests a complex impact of assumptions of κ_{SOA} on the retrieved BC density. In addition, the In-BC density decreased slightly as ρ_{Ex-BC} increased (Fig. 2e), suggesting applying a larger ρ_{Ex-BC} would derive smaller values for In-BC density. The In-BC density was insensitive to the changes of the density of POA and SOA, showing an almost negligible effect on the retrieved results (Fig. 2c and d).

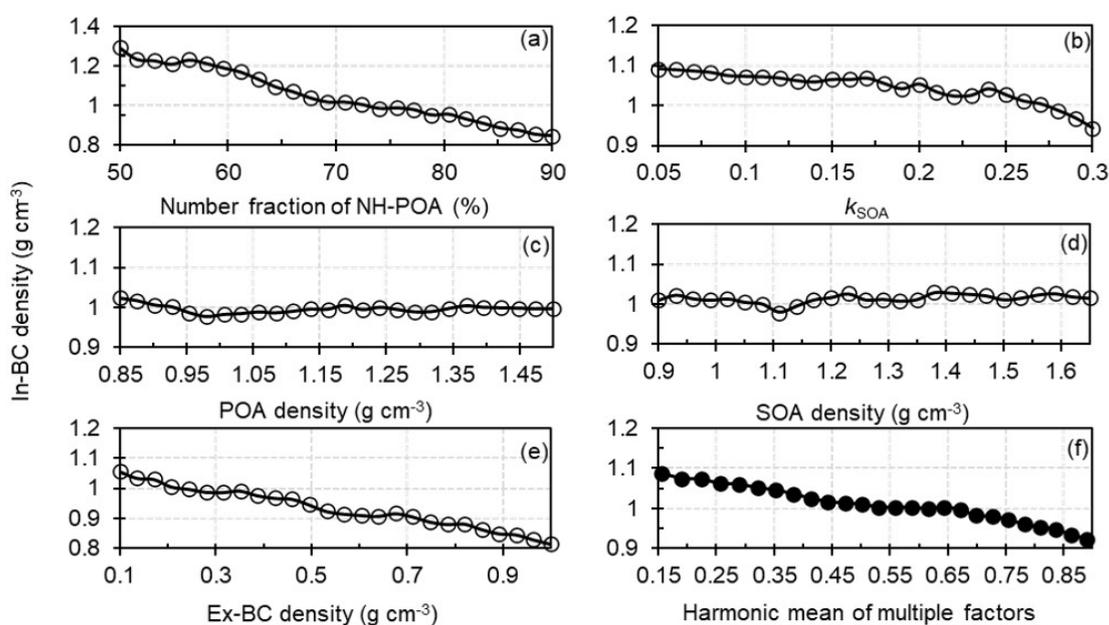


Figure 2. Sensitivity of the In-BC density to variations in the number fraction of nearly hydrophobic (NH) POA (a), the hygroscopic parameter of SOA (b), the POA density (c), the SOA density (d), the externally mixed BC density (e) and the harmonic mean of multiple factors (f).

The uncertainty analysis shows that, by comparing the results based on the mean fraction of the NF_{NH-POA} with a typical atmospheric observed range of 50-90 % for the NF_{NH-POA} (Liu et al., 2021a), the assumption on NF_{NH-POA} can lead to relative deviations (uncertainty) of -17 % ~ +27 % for the retrieved BC density (Fig. 3a).

In addition, unlike inorganics (eg., NH_4HSO_4 , $(NH_4)_2SO_4$ and NH_4NO_3), for

which the hygroscopicity has been already well-understood (Petters and Kreidenweis, 2007), the hygroscopicity of organic species varies largely due to the complexity in organic aerosol constituents. Therefore, the assumption of the values of κ_{SOA} will add the uncertainty in the calculation of BC density. Previous studies have suggested that the organics have a wide range of κ values ranging from 0.05 to 0.3 (Jimenez et al., 2009; Mei et al., 2013). Thus, the sensitivity test has also been done to examine the effect due to changes in κ_{SOA} on calculating the density of BC (Fig. 1a). The result shows that the assumption of κ_{SOA} values can cause an average relative deviation of -10% ~ +3% in calculating the density of In-BC (Fig. 3b).

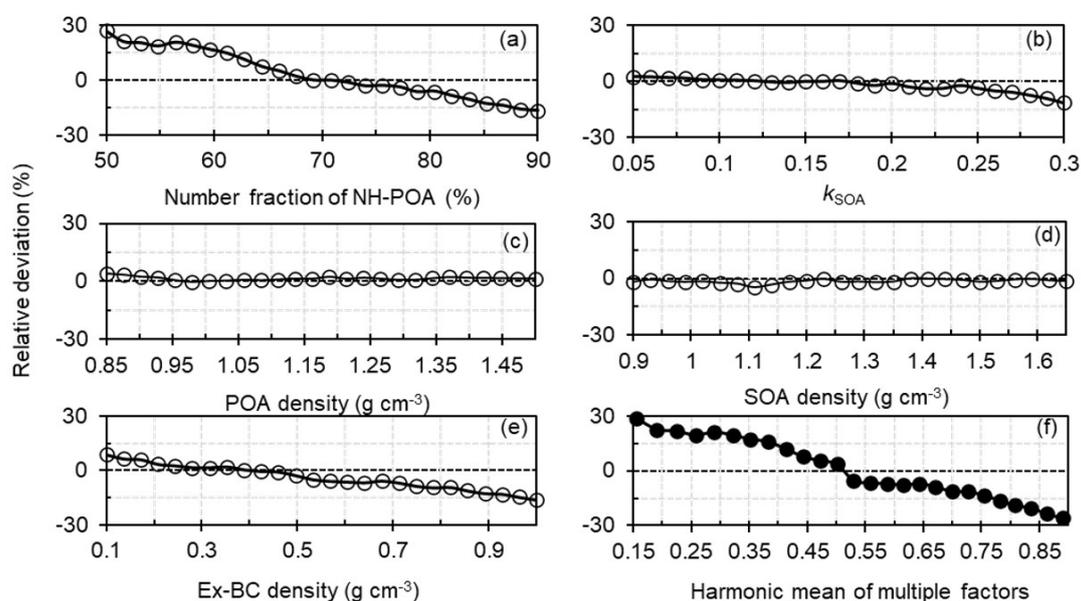


Figure 3. Relative deviations of the number fraction of nearly hydrophobic (NH) POA to the In-BC density (a), the hygroscopic parameter of OA to the In-BC density (b), the POA density to the In-BC density (c), the SOA density to the In-BC density (d), the externally mixed BC density to In-BC density (e) and the combined deviations based on multiple factors mentioned above (f).

However, the sensitivity test shows that the impact of both the ρ_{POA} and ρ_{SOA} variations on the BC density estimation was very small or even negligible (Fig. 1b, c). By varying the ρ_{POA} from 0.85 to 1.5 g cm^{-3} and the ρ_{SOA} from 0.9 to 1.65 g cm^{-3} according to the literature (Noureddini et al., 1992; Alfarra et al., 2006; Reyes-Villegas et al., 2018; Kostenidou et al., 2007), the retrieval uncertainties in the BC density were

both within $\pm 5\%$ (Fig. 3c, d). For $\rho_{\text{Ex-BC}}$, it exhibited that the evolution of the $\rho_{\text{Ex-BC}}$ could lead to an average deviation of $-16\% \sim +9\%$ in calculating In-BC density (Fig. 3e) when increasing the values of $\rho_{\text{Ex-BC}}$ from 0.1 to 1.0 g cm^{-3} , which represents a typical range in ambient atmosphere (Wu et al., 2019; Liu et al., 2020). A combined uncertainty (δ) caused by the multiple factors (δ_i), which was calculated by equation (12), was $-26\% \sim +29\%$ as shown in Fig. 3f.

$$\delta = \sqrt{\sum_{i=1}^n \delta_i^2} \quad (12)$$

In addition, it should be noted that the mass concentration of BC obtained from AE33 based on aerosol light absorption may lead some uncertainties. However, the comparison of the simultaneously measured data by SP2 with those by AE33 during the campaign shows that the temporal variations of BC mass concentrations measured by the two techniques were well consistent (Fig. S8). Note that the BC mass measured by SP2 is occasionally low probably because of the low detection efficiency in small size (McMeeking et al., 2010; Schwarz et al., 2006). In addition, the SP2 is unable to quantify the BC mass beyond a certain limit because of the saturation of electronic devices recording signals (Pileci et al., 2021). We show that, compared the results that were retrieved if applying the BC mass measured by SP2, the BC density retrieved based on AE33 can be 18% higher. Given the measurement bias from SP2, this overestimation indicates an upper limit of the uncertainty.

For the second comment about the uncertainties from differences in composition of the non-BC and BC-containing particles, we also have included statements and more discussions (see lines 228-236) or as follows,

“...Third, since only one hydrophobic and/or one hygroscopic mode was observed by the HTDMA in most cases during the campaign (Fig.S1, S5), the chemical components of the more hygroscopic (MH) mode at a given diameter should contain both these hygroscopic non-BC and the coatings on BC-containing particles, which would be measured by the HR-AMS instrument together. Therefore, by subtracting the externally mixed POA in non-hygroscopic mode (see section 2.3), the concentration and mass fraction of each component measured by HR-AMS can represent the overall

chemical composition of MH modes, and thus was applied in the ZSR mixing rule for the retrieval of the density of internally mixed BC in this study ...”

For the third comment, some statements have been added in the revised version or as follows,

Lines 95-102 “...Moreover, although the BC accounts for very small mass fractions (5~10 %) in total fine aerosols, according to our previous field observed results, the BC-containing particles could contribute 60 %-78 % toward the total number concentration in urban Beijing (Chen et al., 2020). This is comparable to the other results using SP2 instrument, which measured that the number fractions of the coated BC-containing aerosols could be as high as about 50-80% at the field sites in north China (Liu et al., 2019b; Zhao et al., 2022). Therefore, the effect of BC density on the uncertainty of CCN prediction should be concerned carefully...”

Lines 528-532 “... In addition, although the BC accounts for small mass fractions in ambient fine aerosols, according to the measurements simultaneously conducted at the site, the BC-containing particles could contribute 60 %-78 % toward the total number concentration in urban Beijing (Chen et al., 2020). Our results further highlight the effect of BC density on the uncertainty of CCN prediction should be concerned carefully ...”

Comments from the reviewer 1:

There are some minor issues needed to be resolved before accepting for the final publication. A list is given below:

1. It is strongly suggested that the authors carefully doublecheck the manuscript before submitting the next revision.

Re: Thanks a lot, the manuscript has been carefully double checked.

2. Abstract: L29, November 14; L32-33, it is not necessary to say “in non-hygroscopic or hygroscopic mode”, I suggest to remove this expression; L36, literature (lower case); L38, concentration (singular).

Re: revised. L29, from 15 November to 14 December 2016. L36, and L38, revised.

L32-33, the sentence has been revised as “The uncertainty of the retrieval method was

evaluated within $\pm 30\%$, which was primarily caused by assumptions on both the hygroscopic parameter of organics and the proportional distribution of primary organic aerosols in different hygroscopic modes”

3. L167, referring to.

Re: revised.

4. L190, a mean of.

Re: revised.

5. L274 and throughout the whole manuscript, the numbers after “equation” are suggested to be consistent, either in the parentheses or just the numbers.

Re: Thanks for the comments, revised.

6. L284, with secondary particles; L286, measurement data; L294, “And” should be deleted.

Re: revised.

7. L299, Fig. 2., check throughout the paper. There should be a space between the Fig. and the figure number.

Re: revised.

8. L306, corresponds to.

Re: revised.

9. L324, for which, otherwise the conjunction which does not have a role in the clause.

Re: revised.

10. L332, kSOA values.

Re: revised.

11. L337 and throughout the manuscript, literature. Use singular instead of plural form.

Re: revised.

12. L348, with those by AE33, data is in plural form.

Re: revised.

13. L353-354, what is “electronic devices recording signals”?

Re: As detailed description by Pileci et al. (2021), the SP2 is unable to quantify rBC mass above a certain limit due to the saturation of the electronics that record the signals. This saturation limit can be varied via detector gains, with typical settings resulting in upper limits of quantification ranging from $DrBC \approx 500$ nm to around $DrBC \approx 1$ μ m. Consequently, the total BC mass may be underestimated if BC cores greater than the upper limit of quantification contributes substantially to total BC mass.

Pileci, R. E., Modini, R. L., Bertò, M., Yuan, J., Corbin, J. C., Marinoni, A., Henzing, B., Moerman,

M. M., Putaud, J. P., Spindler, G., Wehner, B., Müller, T., Tuch, T., Trentini, A., Zanatta, M., Baltensperger, U., and Gysel-Beer, M.: Comparison of collocated refractory black carbon (rBC) and elemental carbon (EC) mass concentration measurements during field campaigns at several European sites, *Atmos. Meas. Tech.*, 14, 1379–1403, <https://doi.org/10.5194/amt-14-1379-2021>, 2021.

14. L355, were retrieved if applying...

Re: revised.

15. L356, given the measurement bias, no “that” in between.

Re: revised.

16. L359, 3.1 subsection title is suggested to change to “A comparison and validation of retrieved mixing state and density of BC”

Re: Thanks for the comments, revised.

17. L362, present not presented.

Re: revised.

18. L367, that should be deleted, and this is not a clause.

Re: revised.

19. L378, Chen et al. (2020).

Re: revised.

20. L380, the “,” between “using a tandem... SP2” should be deleted.

Re: revised.

21. L383, what is “those” being represented? it seems to referring to the mass fraction.

Re: The sentence has been revised as “Overall, the mass fraction obtained in our study was comparable with that reported in urban Beijing”.

22. L385, coating processes.

Re: revised.

23. L388, what is its referring to? BC density or BC, from the sentence, it seems BC density. Suggested to be modified.

Re: revised. The sentence has been revised as “Accordingly, the densities of the bulk and internally mixed BC present apparent fluctuations as shown in Fig. 4b, which is significantly affected by the variations of BC emission sources and BC aging processes”.

24. L390, due to that? What is “that”for? Please delete.

Re: revised.

25. L393, lift? rising seems better.

Re: revised.

26. L394, with secondary inorganic aerosol.

Re: revised.

27. L400, the continuous or the continuing.

Re: revised.

28. L404-405, The campaign average values of the bulk and internally mixed BC densities are

Re: revised.

29. L407, are not void-free spheres.

Re: revised.

30. L408, with those observed at other sites....

Re: revised.

31. L410, within the range of density from field measurements.

Re: revised.

32. L415-416, there is a “,” after e.g.

Re: revised.

33. L419, as was summarized and ...

Re: revised.

34. L422, a peak value at ..., because it is not the peak value.

Re: revised.

35. L433, in large biases.

Re: revised.

36. L436, same issue as #23.

Re: The sentence has been revised as “Considering the large variation range of BC density during the campaign, which is closely associated with BC morphology or degree of BC aging ...”

37. L448, represents the range of BC density in the atmosphere. The lower and upper limits are just two values.

Re: revised. The sentence has been corrected as “The results show that, by varying the value of density from 0.14 to 2.1 g cm⁻³ that represents the range of BC density in the atmosphere...”

38. L451, delete “And”.

Re: revised.

39. L461, much worse than the use of...

Re: revised. “much worse compared to the use of ...” has been corrected to “much worse than the use of ...”

40. L462, at night or during nighttime.

Re: revised. “at nighttime” has been corrected to “at night”

41. L465, particles are; delete “And now”.

Re: revised.

42. L466, delete “;”.

Re: revised.

43. L468, delete “those”.

Re: revised.

44. L473, as the fraction of organics is.

Re: revised.

45. L480, the reference from Ren et al. (2018).

Re: revised.

46. Subsection title is suggested to change to “NCCN prediction based on the real-time variations of BC density and mixing state.

Re: Thanks for the suggestion, revised.

47. Figure 7. Predicted CCN number...

Re: revised.

48. L494, what are those periods?

Re: The sentence has been revised as “The diurnal variations in the ratio of predicted-to-measured N_{CCN} shows the N_{CCN} can be underestimated by up to 15 % at $S=0.40$ % during noontime.”

49. L498-499, A better closure at $S=0.23\%$ is because the bulk k of particles is closer to that at the critical diameter of 100-150 nm.

Re: revised.

50. L504, what is CCN closure? Do you mean CCN number concentrations? Same in L531.

Re: Closure study for CCN provides an indirect way to evaluate the effects of aerosol microphysical properties (eg., particle size distribution, chemical composition, mixing state and etc.) in calculating CCN number concentrations. When the bias between the predicted and measured CCN concentrations is small or even negligible, it means the closure achieves. Some descriptions about this issue have been included in the text, see **lines 457-468** or as follows:

“...Closure studies provide a useful way to investigate the importance of aerosol properties to CCN concentration prediction. If the closure study is achieved, it means the bias between the predicted and measured CCN concentrations is within $\pm 15\%$ (Chang et al., 2007). The detailed calculation methods are presented in the supporting information (SI: Methods) or the reference in Ren et al. (2018) ...”

51. L521, range or ranges.

Re: revised.

52. L526, The uncertainties of the NCCN prediction are -28 % ~11 %...

Re: revised. The sentence has been revised as “Further examination shows the uncertainties of the N_{CCN} prediction were -28 % ~11 % at the typical S of 0.23 % and 0.40 % by varying the BC density from 0.14 to 2.1 g cm⁻³ that represented the range of ambient BC particles.”

53. L529, the variations of BC density.

Re: revised. The sentence has been corrected as “Moreover, the prediction was found more sensitive to the variations of BC density when it was <1.0 g cm⁻³, suggesting a great significance of accounting for the effect of BC density for the aerosol particles with low aging degree when evaluating the climate effect.”

54. L530, a great significance of accounting for...

Re: revised. Same as reply 53.

55. L541, in future studies.

Re: revised. “in further studies” has been corrected to “in future studies”.

56. L542, provide more comprehensive understanding of the variations of BC density.

Why do you use variability?

Re: Thanks for the comments, “variability” has been corrected to “variations”.

57. Assessing not accessing.

Re: revised.

Comments from the reviewer 2:

This manuscript is much better than before.

My major concern is (1) that there are too many assumptions in the deriving of the effective density.

Re: Thanks for the comments. We are well aware of the uncertainty of such a new

method due to the assumptions. Therefore, as we addressed in section 2.3, a comprehensive analysis of the reason and uncertainty of this method has been presented in the revised paper. See section 2.3 or as follows:

2.3 Uncertainties and limitations

For the retrieval, the assumptions on the values of κ_{SOA} , ρ_{POA} , ρ_{SOA} and $\rho_{\text{EX-BC}}$ as well as the fraction of primary organic aerosols in non-hygroscopic mode would add uncertainty in the inferred values of ambient internally mixed BC density. For example, the freshly emitted POA particles might consistently be coated with secondary particles during the aging process, resulting in changes of the $NF_{\text{NH-POA}}$. However, a real-time variation of the $NF_{\text{NH-POA}}$ is not yet available due to the lack of such measurement data.

Applying only rough fractions of hydrophobic POA for three different atmospheric conditions could still cause uncertainties. Also, the densities of POA and SOA may differ due to their precursors, emission sources and the formation mechanisms in ambient atmosphere (Alfarra et al., 2006; Reyes-Villegas et al., 2018). The density of EX-BC is generally characterized by the morphology and size (Wu et al., 2019). In addition, the value of κ_{SOA} spans largely due to the variations in the emissions of gas precursors and formation processes under different atmospheric conditions (Zhang et al., 2015; Liu et al., 2021b). Therefore, we examined the sensitivities of In-BC density to the variations of these factors, as exhibited in Fig. 1 and Fig. 2.

The figures show that the In-BC density gradually decreased with the increment of the $NF_{\text{NH-POA}}$, implying the high fraction of bare POA particles corresponded to the early aging stage of aerosol particles. With the increase of κ_{SOA} , the In-BC density was generally reduced, but with small fluctuations (Fig. 1a, Fig. 2b). This suggests a complex impact of assumptions of κ_{SOA} on the retrieved BC density. In addition, the In-BC density decreased slightly as $\rho_{\text{EX-BC}}$ increased (Fig. 2e), suggesting applying a larger $\rho_{\text{EX-BC}}$ would derive smaller values for In-BC density. The In-BC density was insensitive to the changes of the density of POA and SOA, showing an almost negligible effect on the retrieved results (Fig. 2c and d).

The uncertainty analysis shows that, by comparing the results based on the mean fraction of the $NF_{\text{NH-POA}}$ with a typical atmospheric observed range of 50-90 % for the

$NF_{\text{NH-POA}}$ (Liu et al., 2021a), the assumption on $NF_{\text{NH-POA}}$ can lead to relative deviations (uncertainty) of -17 % ~ +27 % for the retrieved BC density (Fig. 3a).

In addition, unlike inorganics (eg., NH_4HSO_4 , $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3), for which the hygroscopicity has been already well-understood (Petters and Kreidenweis, 2007), the hygroscopicity of organic species varies largely due to the complexity in organic aerosol constituents. Therefore, the assumption of the values of κ_{SOA} will add the uncertainty in the calculation of BC density. Previous studies have suggested that the organics have a wide range of κ values ranging from 0.05 to 0.3 (Jimenez et al., 2009; Mei et al., 2013). Thus, the sensitivity test has also been done to examine the effect due to changes in κ_{SOA} on calculating the density of BC (Fig. 1a). The result shows that the assumption of κ_{SOA} values can cause an average relative deviation of -10 % ~ +3 % in calculating the density of In-BC (Fig. 3b).

However, the sensitivity test shows that the impact of both the ρ_{POA} and ρ_{SOA} variations on the BC density estimation was very small or even negligible (Fig. 1b, c). By varying the ρ_{POA} from 0.85 to 1.5 g cm^{-3} and the ρ_{SOA} from 0.9 to 1.65 g cm^{-3} according to the literature (Noureddini et al., 1992; Alfarra et al., 2006; Reyes-Villegas et al., 2018; Kostenidou et al., 2007), the retrieval uncertainties in the BC density were both within ± 5 % (Fig. 3c, d). For $\rho_{\text{Ex-BC}}$, it exhibited that the evolution of the $\rho_{\text{Ex-BC}}$ could lead to an average deviation of -16 % ~ +9 % in calculating In-BC density (Fig. 3e) when increasing the values of $\rho_{\text{Ex-BC}}$ from 0.1 to 1.0 g cm^{-3} , which represents a typical range in ambient atmosphere (Wu et al., 2019; Liu et al., 2020). A combined uncertainty (δ) caused by the multiple factors (δ_i), which was calculated by equation (12), was -26 % ~ +29 % as shown in Fig. 3f.

$$\delta = \sqrt{\sum_{i=1}^n \delta_i^2} \quad (12)$$

In addition, it should be noted that the mass concentration of BC obtained from AE33 based on aerosol light absorption may lead some uncertainties. However, the comparison of the simultaneously measured data by SP2 with those by AE33 during the campaign shows that the temporal variations of BC mass concentrations measured by the two techniques were well consistent (Fig. S8). Note that the BC mass measured

by SP2 is occasionally low probably because of the low detection efficiency in small size (McMeeking et al., 2010; Schwarz et al., 2006). In addition, the SP2 is unable to quantify the BC mass beyond a certain limit because of the saturation of electronic devices recording signals (Pileci et al., 2021). We show that, compared the results that were retrieved if applying the BC mass measured by SP2, the BC density retrieved based on AE33 can be 18% higher. Given the measurement bias from SP2, this overestimation indicates an upper limit of the uncertainty.

(2) the chemical components of the non-BC component and BC coating should be different; the uncertainties should be discussed.

Re: Some statements about this have included in the revised text (see **lines 228-236**) or as follows,

“...Third, since only one hydrophobic and/or one hygroscopic mode was observed by the HTDMA in most cases during the campaign (Fig.S1, S5), the chemical components of the more hygroscopic (MH) mode at a given diameter should contain both these hygroscopic non-BC and the coatings on BC-containing particles, which would be measured by the HR-AMS instrument together. Therefore, by subtracting the externally mixed POA in non-hygroscopic mode (see section 2.3), the concentration and mass fraction of each component measured by HR-AMS can represent the overall chemical composition of MH modes, and thus was applied in the ZSR mixing rule for the retrieval of the density of internally mixed BC in this study ...”

(3) the author contributes the uncertainties of the CCN number concentration to the BC density. To my best knowledge, the number fraction of the BC-containing particles in the ambient particle is (less than 25%) low based on the SP2 measurement.

Re: According to our previous field observed results, the BC-containing particles could contribute 60 %-78 % toward the total number concentration in urban Beijing (Chen et al., 2020), which is comparable to the results using SP2 instrument at the field sites in north China (Liu et al., 2019b; Zhao et al., 2022). Therefore, the effect of BC density on the uncertainty of CCN prediction should also be concerned. We have included some statements in the revised paper or as follows:

Lines 95-102 “...Moreover, although the BC accounts for very small mass

fractions (5~10 %) in total fine aerosols, according to our previous field observed results, the BC-containing particles could contribute 60 %-78 % toward the total number concentration in urban Beijing (Chen et al., 2020). This is comparable to the other results using SP2 instrument, which measured that the number fractions of the coated BC-containing aerosols could be as high as about 50-80% at the field sites in north China (Liu et al., 2019b; Zhao et al., 2022). Therefore, the effect of BC density on the uncertainty of CCN prediction should be concerned carefully...”

Lines 528-532 “... In addition, although the BC accounts for small mass fractions in ambient fine aerosols, according to the measurements simultaneously conducted at the site, the BC-containing particles could contribute 60 %-78 % toward the total number concentration in urban Beijing (Chen et al., 2020). Our results further highlight the effect of BC density on the uncertainty of CCN prediction should be concerned carefully ...”

(4) I also doubt the meaning of the retrieved BC density in this work.

Re: The effective density of BC is a crucial factor relevant to its aging degree that would add uncertainty in evaluating its climate effect. However, as we stated in the introduction section of manuscript, the mixing state and the density of BC particles are usually directly measured by several techniques, such as VTDMA-SP2 (Zhang et al., 2016b), or DMA-SP2 (Olfert et al., 2007; Rissler et al., 2014; Wu et al., 2019), and DMA-CPMA-SP2 system (Liu et al., 2019b; Yu et al., 2020), etc. But, such techniques or measurements are not available in many previously conducted field campaigns. In this study, we develop a new method for retrieving the mixing state and effective density of ambient BC particles by combining field measured hygroscopic growth factor and aerosol chemical composition and Köhler theory. This study provides a unique way of utilizing field measurements to infer ambient BC density and highlights the importance of applying variable BC density values in models when predicting CCN and assessing its relevant climate effect. The method is with some limitations. However, a comprehensive uncertainty analysis has been made in the paper so that the scientists can use it more scientifically.

References:

- Alfarra, M. R., Paulsen, D., Gysel, M., Garforth, A. A., Dommen, J., Prévôt, A. S. H., Worsnop, D. R., Baltensperger, U., and Coe, H.: A mass spectrometric study of secondary organic aerosols formed from the photooxidation of anthropogenic and biogenic precursors in a reaction chamber, *Atmos. Chem. Phys.*, 6, 5279–5293, <https://doi.org/10.5194/acp-6-5279-2006>, 2006.
- Reyes-Villegas, E., Bannan, T., Le Breton, M., Mehra, A., Priestley, M., Percival, C., Coe, H., and Allan, J. D.: Online Chemical Characterization of Food-Cooking Organic Aerosols: Implications for Source Apportionment, *Environ. Sci. Technol.*, 52, 5308–5318, <https://doi.org/10.1021/acs.est.7b06278>, 2018.
- Wu, Y. F., Xia, Y. J., Huang, R. J., Deng, Z. Z., Tian, P., Xia, X. G., et al.: A study of the morphology and effective density of externally mixed black carbon aerosols in ambient air using a size-resolved single-particle soot photometer (SP2), *Atmos. Meas. Tech.*, 12, 4347–4359, <https://doi.org/10.5194/amt-12-4347-2019>, 2019.
- Zhang, R., Wang, G., Guo, S., Zamora, M. L., Ying, Q., Lin, Y.: Formation of urban fine particulate matter, *Chemical Reviews*, 115(10), 3803–3855, <https://doi.org/10.1021/acs.chemrev.5b00067>, 2015.
- Liu, J., Zhang, F., Xu, W., Sun, Y., Chen, L., Li, S.: Hygroscopicity of organic aerosols linked to formation mechanisms, *Geophysical Research Letters*, 48, e2020GL091683, <https://doi.org/10.1029/2020gl091683>, 2021b.
- Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., et al.: Evolution of Organic Aerosols in the Atmosphere, *Science*, 326, 1525–1529, <https://doi.org/10.1126/science.1180353>, 2009.
- Noureddini, H., Teoh, B. C., Davis Clements, L.: Densities of vegetable oils and fatty acids, *J. Am. Oil Chem. Soc.*, 69 (12), 1184–1188, 1992.
- McMeeking, G.R., Hamburger, T., Liu, D., Flynn, M., Morgan, W.T., Northway, M., Highwood, E.J., Krejci, R., Allan, J.D., Minikin, A., Coe, H.: Black carbon measurements in the boundary layer over western and northern Europe. *Atmos. Chem. Phys.* 10, 9393-9414, <https://doi.org/10.5194/acp-10-9393-2010>, 2010.
- Kostenidou, E., Pathak, R. K., & Pandis, S. N.: An Algorithm for the Calculation of Secondary Organic Aerosol Density Combining AMS and SMPS Data, *Aerosol Science and Technology*, 41:11, 1002-1010, <https://doi.org/10.1080/02786820701666270>, 2007.
- Liu, H., Pan, X., Liu, D., Liu, X., Chen, X., Tian, Y., Sun, Y., Fu, P., and Wang, Z.: Mixing characteristics of refractory black carbon aerosols at an urban site in Beijing, *Atmos. Chem. Phys.*, 20, 5771–5785, <https://doi.org/10.5194/acp-20-5771-2020>, 2020.
- Mei, F., Setyan, A., Zhang, Q., and Wang, J.: CCN activity of organic aerosols observed downwind of urban emissions during CARES, *Atmos. Chem. Phys.*, 13, 12155–12169, <https://doi.org/10.5194/acp-13-12155-2013>, 2013.
- Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, 7, 1961–1971, <https://doi.org/10.5194/acp-7-1961-2007>, 2007.
- Pileci, R. E., Modini, R. L., Bertò, M., Yuan, J., Corbin, J. C., Marinoni, A., Henzing, B., Moerman, M. M., Putaud, J. P., Spindler, G., Wehner, B., Müller, T., Tuch, T., Trentini, A., Zanatta, M., Baltensperger, U., and Gysel-Beer, M.: Comparison of co-located refractory black carbon (rBC)

- and elemental carbon (EC) mass concentration measurements during field campaigns at several European sites, *Atmos. Meas. Tech.*, 14, 1379–1403, <https://doi.org/10.5194/amt-14-1379-2021>, 2021.
- Chang, R. Y. W., Liu, P. S. K., Leaitch, W. R., and Abbatt, J. P. D.: Comparison between measured and predicted CCN concentrations at Egbert, Ontario: Focus on the organic aerosol fraction at a semirural site, *Atmos. Environ.*, 41, 8172–8182, <https://doi.org/10.1016/j.atmosenv.2007.06.039>, 2007.
- Ren, J., Zhang, F., Wang, Y., Collins, D., Fan, X., Jin, X., et al.: Using different assumptions of aerosol mixing state and chemical composition to predict CCN concentrations based on field measurements in urban Beijing, *Atmos. Chem. Phys.*, 18, 6907–6921, <https://doi.org/10.5194/acp-18-6907-2018>, 2018.
- Liu, L., Zhang, J., Zhang, Y., Wang, Y., Xu, L., Yuan, Q., et al.: Persistent residential burning-related primary organic particles during wintertime hazes in North China: insights into their aging and optical changes, *Atmos. Chem. Phys.* 21, 2251–2265, <https://doi.org/10.5194/acp-21-2251-2021>, 2021a.
- Tan, H., Yin, Y., Gu, X., Li, F., Chan, P. W., Xu, H., Deng, X., and Wan, Q.: An observational study of the hygroscopic properties of aerosols over the Pearl River Delta region, *Atmos. Environ.*, 77, 817–826, <https://doi.org/10.1016/j.atmosenv.2013.05.049>, 2013.
- Chen, L., Zhang, F., Zhang, D., Wang, X., Song, W., Liu, J., Ren, J., Jiang, S., Li, X., and Li, Z.: Measurement report: Hygroscopic growth of ambient fine particles measured at five sites in China, *Atmos. Chem. Phys.*, 22, 6773–6786, <https://doi.org/10.5194/acp-22-6773-2022>, 2022.
- Chen, L., Zhang, F., Yan, P., Wang, X., Sun, L., Li, Y., Zhang, X., Sun, Y., and Li, Z.: The large proportion of black carbon (BC)-containing aerosols in the urban atmosphere, *Environ. Pollut.*, 263, 114507, <https://doi.org/10.1016/j.envpol.2020.114507>, 2020.
- Liu, H., Pan, X.L., Wu, Y., Wang, D.W., Tian, Y., Liu, X.Y., et al.: Effective densities of soot particles and their relationships with the mixing state at an urban site in the Beijing megacity in the winter of 2018, *Atmos. Chem. Phys.* 19, 14791–14804, <https://doi.org/10.5194/acp-19-14791-2019>, 2019b.
- Zhao, G., Tan, T., Hu, S., Du, Z., Shang, D., Wu, Z., Guo, S., Zheng, J., Zhu, W., Li, M., Zeng, L., and Hu, M.: Mixing state of black carbon at different atmospheres in north and southwest China, *Atmos. Chem. Phys.*, 22, 10861–10873, <https://doi.org/10.5194/acp-22-10861-2022>, 2022.
- Zhang, Y., Zhang, Q., Cheng, Y., Su, H., Kecorius, S., Wang, Z., Wu, Z., Hu, M., Zhu, T., Wiedensohler, A., and He, K.: Measuring the morphology and density of internally mixed black carbon with SP2 and VTDMA: new insight into the absorption enhancement of black carbon in the atmosphere, *Atmos. Meas. Tech.*, 9, 1833–1843, <https://doi.org/10.5194/amt-9-1833-2016>, 2016b.
- Olfert, J. S., Symonds, J. P. R., and Collings, N.: The effective density and fractal dimension of particles emitted from a light-duty diesel vehicle with a diesel oxidation catalyst, *J. Aerosol Sci.*, 38, 69–82, <https://doi.org/10.1016/j.jaerosci.2006.10.002>, 2007.
- Rissler, J., Nordin, E. Z., Eriksson, A. C., Nilsson, P. T., Frosch, M., Sporre, M. K., Wierzbicka, A., Svenningsson, B., Londahl, J., Messing, M. E., Sjogren, S., Hemmingsen, J. G., Loft, S., Pagels, J. H., and Swietlicki, E.: Effective Density and Mixing State of Aerosol Particles in a Near-Traffic Urban Environment, *Environ. Sci. Technol.*, 48, 6300–6308, <https://doi.org/10.1021/es5000353>, 2014.

- Wu, Y. F., Xia, Y. J., Huang, R. J., Deng, Z. Z., Tian, P., Xia, X. G., et al.: A study of the morphology and effective density of externally mixed black carbon aerosols in ambient air using a size-resolved single-particle soot photometer (SP2), *Atmos. Meas. Tech.*, 12, 4347–4359, <https://doi.org/10.5194/amt-12-4347-2019>, 2019.
- Yu, C., Liu, D., Broda, K., Joshi, R., Olfert, J., Sun, Y., Fu, P., Coe, H., Allan, J.D.: Characterising mass-resolved mixing state of black carbon in Beijing using a morphology-independent measurement method, *Atmos. Chem. Phys.*, 20, 3645–3661. <https://doi.org/10.5194/acp-20-3645-2020>, 2020.