A point-by-point response to reviewers

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.

I'd like to thank you for your efforts and time on handling the paper. I 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). Particularly, regarding to the major concern of Reviewer #3 who questioned the uncertainty of the method, a further comprehensive uncertainty analysis has been done. We show that the overall retrieval uncertainties in the BC density are ranged from -26 % to +29 % (within ±30%), which we think is reasonable and more scientific rather than divorced from reality. We don’t think it is scientific to evaluate the uncertainty by using some extreme values, ranges or values that cannot represent the typical conditions of the actual atmosphere. For example, the range of number fraction of NH-POA (50-90%) was chosen in this study because which is within a typical range of the NH-POA representing the actual atmosphere according to the literatures. When considering the extreme conditions (eg., the number fraction of NH-POA is 0 or 100%), the deviations caused by the assumption on NFNH-POA can increase to the range of -68%~+44%. However, such evaluation may not be representative of the actual atmospheric conditions. Actually, we compared the results with the laboratory and field measurements reported in the literature. It shows generally good agreement with the previously reported values, indicating that the method is feasible. Therefore, 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

A point-by-point response to reviewer

Reviewer #1

The authors basically resolved all the issues. However, by carefully reading through the manuscript, numerous typos/ill sentences still exist in the revision. A careful proofread is strongly suggested before resubmitting the next revision. Below are some of the examples (the line numbers are based on the ATC2):

1. L32: a period of time, use “during” instead of “on”;

Re: revised.

2. L37 and throughout the manuscript: internally before a past tense, usually don’t need a hyphen to connect;

Re: revised.

3. L38: a campaign mean density;

Re: revised.

4. L39: Literature not literature;

Re: revised.

5. L42: “would lead to” is better than “led to” since subjunctive mood is used here;

Re: revised.

6. L241: nearly hydrophobic group;

Re: revised.

7. L252-253: By reviewing and summarizing the existing results about, what is “about” used here?

Re: revised. “about” is referred the typical values of density for the freshly emitted or externally mixed BC reported in the Literature.

8. L255: span, not spans; with a mean of …;

Re: revised.

9. L257: a large proportion of what?

Re: The sentence has been revised as “By reviewing and summarizing the existing results, we show that typical values of density for the freshly emitted or externally mixed BC observed in the winter of urban Beijing or North China Plain span over 0.14-0.50 g cm^{-3}, with mean of ~0.40±0.10 g cm^{-3} (Fig. S3), in the size range of 100 to 300 nm, where the mass concentration of externally mixed BC mostly concentrated".

10. L274 and throughout the manuscript: the density signs, please use subscript for the types of BC;

Re: All the density signs have been subscripted for the types of BC.

11. L274: in the calculated In-BC density, not calculating…

Re: revised.

12. L293: since the ZSR rule assumes…

Re: revised.

13. L344: by subtracting or through subtracting, need a prep;

Re: revised.

14. L354: awkward "then", "and" might work better;

Re: revised.

15. L355-356, what do you mean? "Because the maximum value of the ….in this campaign";

Re: According to the simplified ion pairing scheme (Gysel et al., 2007), $n_{H_2SO_4}$was zero. The sentence has been revised as "Because the value of the $n_{H_2SO_4}$was zero in this campaign."

16. L374: that of the lubricating oil…

Re: revised.

17. L375-378: an awkward sentence, please revise;

Re: The sentence has been revised as "Since the cooking organic aerosols represent a high contribution to POA in urban environments, we also choose the mean density of the rapeseed oil and oleic acid (~0.85 g cm-3) (Reyes-Villegas et al., 2018) to evaluated the result as shown in section 2.3."

18. L383, Note that usually follows no comma; organics is singular and accounts for not account for;

Re: revised.

19. L385: lower total volume, here compare to what?

Re: Thanks for the comments. We have noted that, this method fails to retrieve the BC density when organics account for a large fraction (>60 %). This is because that a high fraction of OA usually corresponds to low total volume of all the other species like BC, yielding negative values for $v_{\mathrm{In\text{-}BC}}$ introduced in equation 9. As a result, 39 % of the data observed during the campaign were excluded when calculating the BC density.

20. L386: how vIN-BC was introduced in an equation? Please revise;

Re: According to the review's comments, we have added some introductions about the in the revised text. See **Lines 224-239** or as follows:

"The density of internally mixed BC (In-BC), $\rho_{\mathrm{In\text{-}BC}}$ is then derived from the following equations:

$$\kappa_{gf-MH} = \kappa_{chem} = \sum_i \varepsilon_i \kappa_i = \frac{v_{inorg}}{v_{total}} \kappa_{inorg} + \frac{v_{SOA}}{v_{total}} \kappa_{SOA} + \frac{v_{In-POA}}{v_{total}} \kappa_{POA} + \frac{v_{In-BC}}{v_{total}} \kappa_{BC} \tag{8}$$

where $\kappa_{\mathrm{gf\text{-}MH}}$ is the hygroscopic parameter of the more hygroscopic (MH) mode, κ_{chem} is the hygroscopic parameter of aerosol particles in the mixed composition and can be calculated based on chemical volume fractions using a simple rule (Stokes and Robinson, 1966; Petters & Kreidenweis, 2007), κ_{i} is the hygroscopic parameter of each pure composition and ε_{i} is the volume faction of the individual components in the internally mixed particle. v_{inorg}, v_{SOA} and $v_{\mathrm{In\text{-}POA}}$ are the volume of the inorganic, SOA and internally mixed POA species, and can be calculated as follows: $v_{inorg} = \frac{m_{inorg}}{\rho_{inorg}}$, $v_{SOA} = \frac{m_{SOA}}{\rho_{SOA}}$, and $v_{In-POA} = \frac{m_{In-POA}}{\rho_{POA}}$. v_{total} is the total volume of all the species and can be written as $v_{total} = \frac{m_{inorg}}{\rho_{inorg}} + \frac{m_{SOA}}{\rho_{SOA}} + \frac{m_{In-POA}}{\rho_{POA}} + \frac{m_{In-BC}}{\rho_{In-BC}}$. In equation (8), κ_{BC} and κ_{POA} are assumed to be 0. So, the total volume, v_{total} can be further written as $v_{total} = \frac{v_{inorg}\kappa_{inorg} + v_{SOA}\kappa_{SOA}}{\kappa_{gf-MH}}$. The volume of internally mixed $v_{\mathrm{In\text{-}BC}}$ can be calculated as follows,

$$v_{In-BC} = \frac{v_{inorg}\kappa_{inorg} + v_{SOA}\kappa_{SOA}}{\kappa_{gf-MH}} - v_{inorg} - v_{SOA} - v_{In-POA}$$

$$= \frac{\frac{m_{inorg}}{\rho_{inorg}}\kappa_{inorg} + \frac{m_{SOA}}{\rho_{SOA}}\kappa_{SOA}}{\kappa_{gf-MH}} - \frac{m_{inorg}}{\rho_{inorg}} - \frac{m_{SOA}}{\rho_{SOA}} - \frac{m_{In-POA}}{\rho_{POA}} \tag{9}$$

"

21. L391: … is applied assuming that all the aerosol…., “with the assumption of” can not be used as a conjunction;

Re: revised.

22. L408-409: this is also an awkward sentence;

Re: The sentence has been revised as “Applying only the rough fraction of hydrophobic POA for three different atmospheric conditions could still causes uncertainty.”

23. L419: the higher fraction? Here again, compare to what?

Re: revised. “The figures show that the In-BC density gradually decreases with the increment of the $NF_{\mathrm{NH\text{-}POA}}$, implying the high fraction of bare POA particles correspond to the early aging stage of aerosol particles.”

24. L444 and throughout the rest: -+ sign? This is a formal publication!

Re: revised.

25. L624: this is really hard to understand what the authors want to express, “directly measured or indirectly retrieved previously reported results”

Re: The sentence has been revised as “Our results are basically comparable with those previously reported results, which are directly measured or indirectly retrieved.”

26. L625-626: to be is redundant;

Re: revised.

27. L791: is higher at a smaller particle size??

Re: Here we want to illustrate that the organics are more concentrated on small particles, and the surface tension depression may be more pronounced at small particle sizes.

28. L798: Ren et al. (2018).

Re: revised.

29. L853-854: …caused by assuming…; however, the sentence is still ambiguous;

Re: The sentence has been revised as “The uncertainty of the new retrieval method was evaluated within ±30 %, which is primarily caused by assuming the value of κ_{SOA} and the fraction of primary organic aerosols in non-hygroscopic mode.”

30. L857: internally mixed;

Re: revised.

31. I believe there are still lot of those typos/ill sentences.

Re: Thank you very much for the comments, we have carefully revised the wording and writing in the revised version.

A point-by-point response to reviewer

Reviewer #3

The authors tried to develop a new method to retrieve the effective density of internally mixed BC (In-BC) based on Köhler theory combining with field observations. However, when they retrieve the effective density of BC using the new method, they assumed too much. This would lead great uncertainties in their results, particularly the mass loading of internally mixed BC. Therefore, the retrieved effective density of internally mixed BC is unbelievable. Although the authors evaluated the uncertainties in their results, the uncertainties are largely underestimated due to that some important effect factors are not considered. More details are listed as follows.

(1) In this work, the authors assumed the same effective density of externally mixed BC (Ex-BC) with different sizes. However, the effective density of externally mixed BC decreases with increasing BC mobility diameters. For Ex-BC with mobility diameter smaller than 80 nm, the effective density can be more than 1 g cm-3, but it would be lower than 0.3 g cm-3 for Ex-BC with mobility diameter larger than 300 nm. Because the authors did not consider the substantial difference in effective densities among Ex-BC with different mobility diameters, the mass size distribution of Ex-BC shown in Fig. S4 would have a large uncertainty. This would affect quantification of Ex-BC mass loading, and further lead to a large uncertainty in In-BC mass loading.

Re: Thanks for the comments. It's true that there is some bias in deriving the mass concentration by assuming a fixed value for $\rho_{\text{Ex-BC}}$. Generally, the $\rho_{\text{Ex-BC}}$ would change as a function of the mobility diameter and its morphology (Park et al., 2003; Rissler et al., 2014; Wu et al., 2019). By reviewing and summarizing the existing results, we found that typical values of density for the freshly emitted or externally mixed BC observed in the winter of urban Beijing or North China Plain spans over 0.14-0.50 g cm^{-3} with mean of ~0.40±0.10 g cm^{-3} in the size range of 100 to 300 nm (Fig. S3), where the mass concentration of externally mixed BC accounted for a large proportion in urban Beijing ((Peng et al., 2016, 2017; Wu et al., 2019; Liu et al., 2020; Zhao et al., 2022). Therefore, an average $\rho_{\text{Ex-BC}}$ of 0.4 g cm^{-3} was used for calculating the mass concentration of externally mixed BC in our study. According to the review's comments, we have updated the uncertainty analysis of $\rho_{\text{Ex-BC}}$ in the range of 0.1-1.0 g cm^{-3} as shown in Fig. R1 or Fig. S4. By increasing the $\rho_{\text{Ex-BC}}$ from 0.1 to 1.0 g cm^{-3}, it exhibits that the variations of the $\rho_{\text{Ex-BC}}$ could lead to an uncertainty of -29% ~18% in the calculating the mass fraction of In-BC.

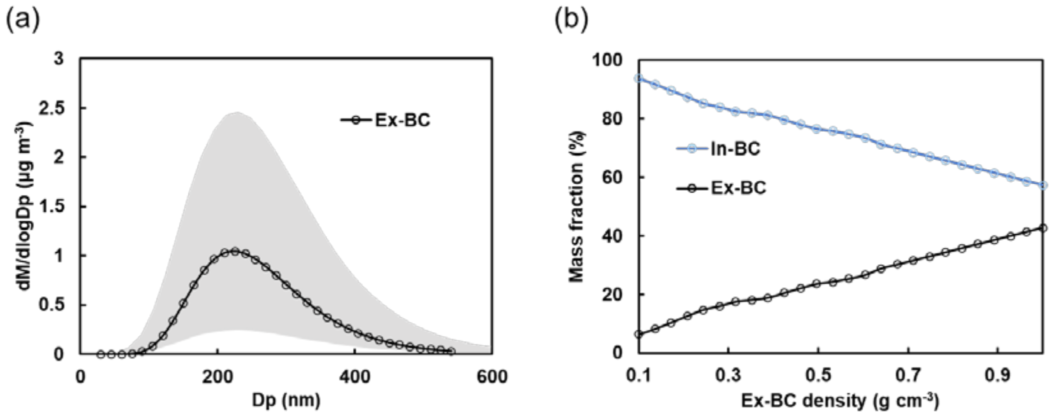

Fig. R1 (a) Average mass size distribution of Ex-BC by modeling as a single log-normal distribution. The shaded part represents the boundary by setting the Ex-BC density as 0.1 g cm^{-3} (lower limit) and 1.0 g cm^{-3} (upper limit). (b) Mean mass fraction of In-BC and Ex-BC by increasing the $\rho_{\text{Ex-BC}}$ from 0.1 to 1.0 g cm^{-3}.

(2) The authors used the AE33 measurement to quantify the total refractory BC (Ex-BC+In-BC), which would largely overestimate the mass loading of total refractory BC. The BC concentrations measured by AE33 based on aerosol light absorption reflect an equivalent BC mass loading, because the aerosol light absorption measured by AE33 not only include the absorption of refractory BC itself, but also the light absorption enhancement caused by lensing effect of BC mixing state. When the light absorption of BC is significantly enhanced by other species coated on BC surface during atmospheric aging, the mass concentrations of atmospheric BC measured by AE33 can be more than twice the refractory BC. The overestimate of the mass loading of total refractory BC would lead to an overestimate of In-BC mass loading. This would strongly affect the

retrieved effective density of In-BC.

Re: Yes, the BC concentrations measured by AE33 based on aerosol light absorption may lead to some overestimations of BC mass loading. Thanks for the reviewer's comments which remind us to make a comparison of the simultaneously measured data by SP2 with that by AE33 during the campaign as shown in Fig R2. It shows that the temporal variations of BC mass concentrations measured by the two techniques are very consistent. 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 above a certain limit due to the saturation of the electronics that record the signals (Pileci et al., 2021). Furthermore, we show that, compared the results that retrieved if applying the BC mass measured by SP2, the BC density retrieved based on AE33 can be 18% higher. Given that the measurement bias from SP2, this overestimation indicates an upper limit of the uncertainty. Some discussions about this issue have been included in the revised paper, see **Lines 346-357** or as follows:

"… In addition, it should be noted that the mass concentration of BC obtained from AE33 based on aerosol light absorption may lead some uncertainty. However, the comparison of the simultaneously measured data by SP2 with that by AE33 during the campaign shows that the temporal variations of BC mass concentrations measured by the two techniques are well consistent (Fig S5). 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 retrieved if applying the BC mass measured by SP2, the BC density retrieved based on AE33 can be 18% higher. Given that the measurement bias from SP2, this overestimation indicates an upper limit of the uncertainty ..."

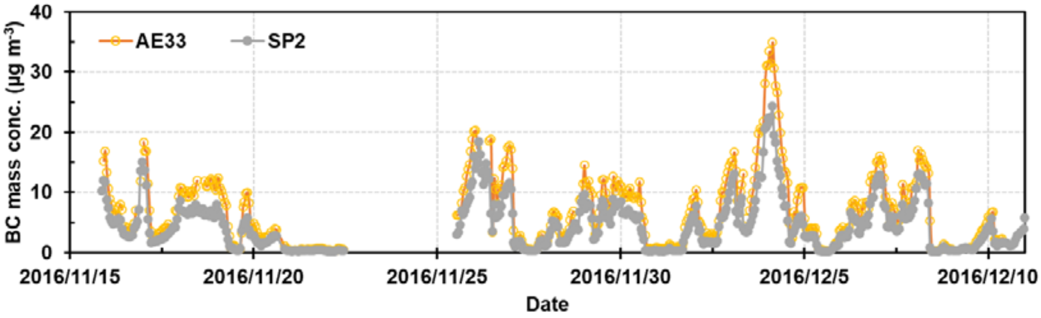

Fig. R2 Time series of mass concentration of BC measured by AE33 and SP2.

(3) Because the authors used a narrow range of the variabilities in the effect factors to make sensitive analysis, the uncertainty (±30%) in In-BC density is underestimated. The authors highlighted that the In-BC density was sensitive to the number fraction of nearly hydrophobic POA (NH-POA), but they selected a narrow range of the number fraction of NH-POA (50-90%) to estimate the uncertainty in In-BC density (Figure 1). For source emission, the number fraction of NH-POA can lower than 50%. To take an extreme example, when all NH-POA is attached to BC particles by coagulation process during source emission, the number fraction of NH-POA can be nearly 0. When the sensitive analysis is made with number fraction of NH-POA in the range of 0-100%, the uncertainty in In-BC density will be much higher than ±30%, which is determined by a narrower range of the number fraction of NH-POA (50-90%). Similarly, they also use a narrow range of POA (0.85-1 g cm-3) and SOA (1.2 -1.4 g cm-3) densities to make sensitive analysis.

Re: The range of number fraction of NH-POA (50-90%) was chosen in this study because that, it showed from a simultaneous measurement at the site that the fractions of hydrophobic POA in the range of 50-90% for nearly 93% for a two-week period of sampled aerosols by referring the literature (Liu et al., 2021a). So, the number fraction of NH-POA (50-90%) is within a typical range of the NH-POA representing the actual atmosphere.

In addition, according to the reviewer's comments, in the revise version, we have applied a wider range of ρ_{POA} (0.85-1.5 g cm-3) and ρ_{SOA} (0.9-1.65 g cm-3) to make uncertainty analysis, which showed the uncertainties in the retrieval BC density are both within ±5 %, as shown in Fig. R3 and R4. See **Lines 334-339** or as follows:

"…However, the sensitivity test shows that the impact of both the ρ_{POA} and ρ_{SOA} variations on the BC density estimation is 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 literatures (Noureddini et al., 1992; Alfarra et al., 2006; Reyes-Villegas et al., 2018; Cai et al., 2020; Kostenidou et al., 2007), the retrieval uncertainties in the BC density are both within ±5 % (Fig. 3c, d) …"

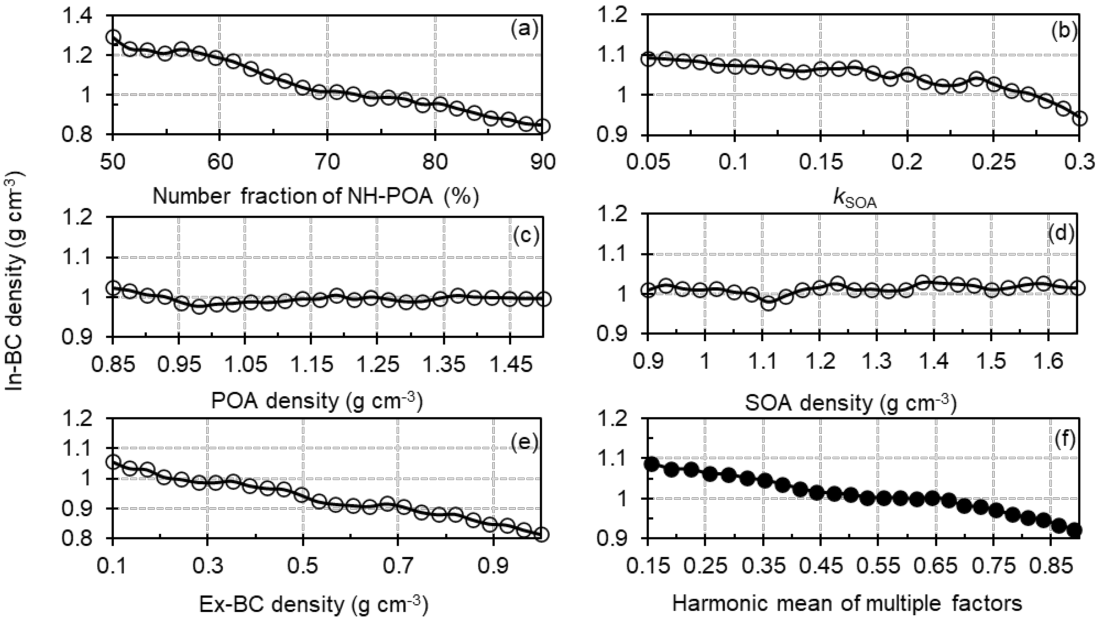

Fig. R3 Sensitivity of the In-BC density to variations in the number fraction of nearly hydrophobic (NH) POA (a), the hygroscopic parameter of SOA (b), the POA density (c), the SOA density (d), the externally mixed BC density (e) and the harmonic mean of multiple factors (f).

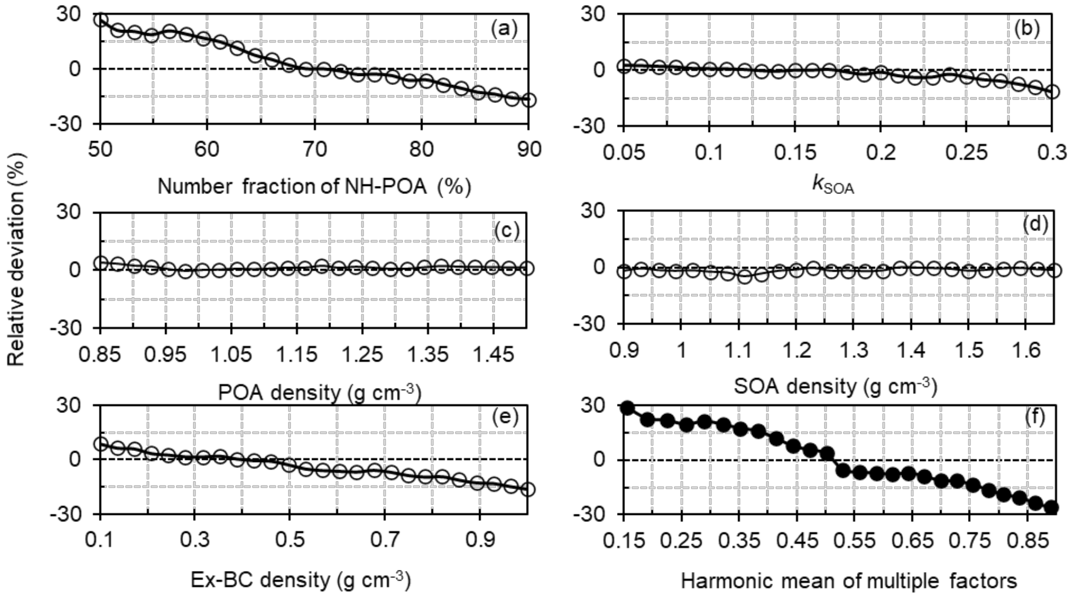

Fig. R4 Relative deviations of the number fraction of nearly hydrophobic (NH) POA to the In-BC density (a), the hygroscopic parameter of OA to the In-BC density (b), the POA density to the In-BC density (c), the SOA density to the In-BC density (d), the externally mixed BC density to In-BC density (e) and the combined deviations based on multiple factors mentioned above (f).

(4) The large variability in the retrieved BC density (0.14-2.1 g cm-3) based the new method developed in this work can not exclude the effect of the great uncertainties in their results. Therefore, the range of the retrieved BC density in this work is meaningless.

Re: Yes, the assumptions on the values of κ_{SOA}, ρ_{POA}, ρ_{SOA} and $\rho_{\mathrm{Ex\text{-}BC}}$ as well as the fraction of primary organic aerosols in non-hygroscopic or hygroscopic mode would add uncertainty in the inferred values of ambient internally mixed BC density, as we addressed in the section 2.3 uncertainties and limitations. The overall retrieval uncertainties in the BC density are ranged from -26 % to +29 %. When considering the extreme conditions (eg., the number fraction of NH-POA is 0 or 100%), the deviations caused by the assumption on NFNH-POA can increase to the range of -68%~+44%. However, the simultaneous measurement at the site that the fractions of hydrophobic POA in the range of 50-90% for nearly 93% for a two-week period of sampled aerosols by referring the literature (Liu et al., 2021a). So, the number fraction of NH-POA (50-

90%) may be more representative of the actual atmospheric NH-POA within the typical range.

In addition, as mentioned in comment 2, the uncertainty in mass loading of BC obtained from AE33 may lead to a maximum bias of 18% when compared with SP2 measurements. Although the retrieved method may have some uncertainties and limitations, it provided a new sight to investigate real-time variations of BC density in the polluted urban atmosphere, and to further study the effects of BC density variations on CCN activity and prediction.

Since the BC density is difficult to measure, it is worth trying to develop new methods for retrieving accurate BC density from available measured data. We think that the current work is with scientific significance for that the effective density of BC is a crucial factor relevant to its aging degree that would add uncertainty in evaluating its climate effect. It is thus necessary to conduct field measurements and obtain more observational data to verify this methodology in further studies.

(5) The CCN closure study can not demonstrate that the prediction CCN number concentrations are improved by using the retrieved BC density. A good agreement between the predicted and observed CCN number concentrations (Fig.7) is due to prediction CCN number using the size-resolved κ based on hygroscopic growth factor measurement (κgf). In this work, the BC density is retrieved by assuming that κgf is equal to κ derived from aerosol chemical composition (κchem), shown in Equation (8). Compared with κchem, the measured κgf can improve prediction CCN number concentrations. When the authors made the sensitivity analysis of predicted CCN number concentrations to change of BC density with prescribed valued of 0.14 and 2.1 g cm-3 (Fig. 6), the κ was calculated based on aerosol chemical composition (i.e., κchem). For these cases, the uncertainties in the predicted CCN number concentrations are dominated by κchem. Therefore, the differences in the predicted CCN number concentrations shown in Figure 6 and Figure 7 is mainly due to using κchem and κgf, respectively.

Re: In this study, it should be noted that here we use the κ_{chem}, not κ_{gf} to calculate the critical diameter both in Figure 6 and Figure 7. The measured N_{CCN} is based on the measurement of CCN counter. The precited N_{CCN} is calculated by using the critical diameter and the particle number size distribution. The critical diameter is calculated based on Köhler theory and ZSR rule. For a multicomponent particle, the Zdanovskii–Stokes– Robinson (ZSR) mixing rule (Stokes and Robinson, 1966) can also estimate κ_{chem} using chemical composition data:

$$\kappa_{chem} = \sum_i \varepsilon_i \kappa_i \tag{1}$$

where ε_{i} and κ_{i} are the volume fraction and κ for the ith chemical component, respectively.

So, the three cases (CCN closures) are just showed with a constant lower limit of BC density of 0.14, a constant upper limit of BC density of 2.1, and a variable retrieved BC densities when calculating the κ_{chem}. Therefore, the CCN closure study could indicate that the sensitivity of the prediction CCN number concentrations to variations in BC density.

Considering a large uncertainty in the retrieved BC density, I do not think the new method developed in this work will be used in the future studies.

Re: Thanks for the comments. Although various techniques have been developed to quantify the mixing state and density of BC in recent years, as we addressed in the Introduction section. However, such techniques or measurements are always not available in many previously conducted filed campaigns. Since the Gf-PDF measured by the HTDMA can reflect the mixing state of aerosol particles (Tan et al., 2013; Hong et al., 2018; Chen et al., 2022), and the hydrophobic mode generally refers to components like freshly emitted BC or POA, which are externally mixed with the aerosol particles populations; while the hydrophilic mode is those secondary inorganic or organic salts and aged BC. Based on this knowledge gained, we proposed a new method and try to retrieve BC density to solve the unavailability of such dataset. We are well aware of the uncertainty of such a new method. Therefore, as we addressed in above comments, a comprehensive analysis of the uncertainty of this method has been presented in the paper. We show that the overall retrieval uncertainties in the BC density are ranged from -26 % to +29 % (within ±30%), which we think is reasonable and more scientific rather than divorced from reality. We don’t think it is scientific to evaluate the uncertainty by using some extreme values, ranges or values that cannot represent the typical conditions of the actual atmosphere. In addition, we compared our results with the laboratory and field measurements reported in the literature. It shows generally good agreement with the previously reported values, indicating that the method is feasible. In future, however, it is necessary to conduct field measurements and obtain more observational data to verify this methodology.

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