

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2021-904

Anonymous Referee #1

Referee comment on "The chemical composition and mixing state of BC-containing particles and the implications on light absorption enhancement" by Jiaying Sun et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-904-RC1>, 2021

The manuscript investigated the mixing state and light absorption enhancement of BC-containing particles at Beijing and Gucheng during winter. They found that coating of second inorganics has a larger enhancement than organic coatings. Overall, this study can improve the knowledge of BC light absorption enhancement and evaluation of BC light absorption during the atmospheric aging process. This can help reduce the uncertainties in BC climate effects. However, this manuscript is not well written. Many cases require additional information to clarify motivation, methodology, results, and interpretation. My impression of this paper is that it could be improved by considering the following suggestions. The revised paper should discuss these points, not just answer in the authors' response. I am willing to review the revised manuscript. Therefore, my recommendation for the editor is that this manuscript needs major revision.

General comments:

- Captions of figures do not include all necessary information. Also, some figures are very confusing. Please see my specific comments below.
- In the Methods section, the authors could provide more information about sampling, analysis methods, instrumentation, etc. I understand some methods have already been published and are widely used in the literature (e.g., L125-127, "The detailed ... in factor *i*."). However, it is better to provide short summaries of these methods in the main manuscript or SI. It is not clear how these methods work for me, and I have to go back to the original references.
- Many discussions are not clear to me. Please see my specific comments.
- I did not see any discussion about uncertainties. Please add these like uncertainties in instruments and data.

Specific comments:

- In this manuscript, you used terms such as 'coating' and 'internally mixed'. In this case, I assume you mean particles are internally mixed and core-shell morphology. However, it could also be partially coated or aggregated together, which you cannot tell from AMS or SP-AMS. Do you have any TEM images, EDX mapping, or shape factor measurement? If you do not have evidence to support core-shell morphology, I would like to see some discussions about the effects of different morphologies.
- I understand that you used 870 nm PAX to measure light absorption properties since many studies believe only BC can absorb at 870 nm. However, many studies have pointed out that brown carbon (BrC) can also absorb at 870 nm, leading to overestimating your eBC. Moreover, BrC can also scatter light at 870 nm, leading to overestimating your BC scattering properties. These two can result in different results of ΔF_R . I did not see any discussion about these. Please add discussions.
- In this study, the max temperature of the thermodenuder (TD) is 250 °C. However, this temperature might not remove all BrC and inorganics (see *"Two-stage aerosol formation in low-temperature combustion"* and *"The Brown-Black Continuum of Light-Absorbing Combustion Aerosols"*). How do you account for that? Also, did you count particle loss in the TD?
- In your calculation of absorption enhancement ($E_{\text{abs}} = b_{\text{abs, total}} / b_{\text{abs, BC}}$), b_{abs} can be different for the same component if their concentration is different before and after TD. It is better to use MAC or imaginary part of the refractive index.
- L114-118, "Seven particles ... are shown in Table S1." How many signals of each fragment do you need to classify a particle to a type? How did you decide the thresholds?
- L130-131, "BC-containing ... (Xie et al., 2020)." I am curious why there is a discrepancy between your study and Xie et al.?
- L135-137, "Comparatively, ... at the urban site." This is true for large particles that SO₄ is generated from fog or cloud processing. SO₄ could be generated from anthropogenic sources such as coal combustion for small particles. Do you have any measurements to show that these SO₄ are coming from the aging processes?
- Section 3.2 is not clear to me. I might misunderstand some concepts. Please clarify my following questions:
 - L163-165, "Moreover, the number ... in urban region." Why do you say this? The correlation between increased BC_N concentration and their role in pollution formation is not clear to me. First, do you have any particle concentration and air quality measurements to show these days are highly polluted? Second, what important roles are you mentioned here? I can see the potential correlation between BC_N formation and RH, but you need to explain a little more between BC_N and urban region pollution. Also, I suggest you do some statistic analysis to show correlations. Similar to any comparison you did in the manuscript.
 - L166-167, "This result ... in rural area." See comment 8(a).
 - L166-171, "In addition, we ... zhang et al./ 2021)." I do not understand this. Higher RH has lower BCOC_N. Does that mean more BCOC_N formation depends on photochemical? Moreover, how could sulfate formation not affect BC mixing state and light-absorption? This does not make sense to me.
 - In figure 3, why did you name the earlier case as case 2? How do you define P0-P5?
 - L175, "As a consequence, ... in half-day." You showed that E_{abs} increased by decreasing BCOC_S and increasing BOCO_N. Could that be because BCOC_S is less volatile than BOCO_N, so that after TD, more BOCO_N (coating) can be removed?
 - L180-187, "As shown in Fig. 3d, ... at high PM level." First, how do you conclude the

mountain valley winds influenced the concentration of the fine particles? Do you have any evidence to show the wind direction, or have any references explained that before? Moreover, since you mentioned the fine particles were dominated by fossil fuel OA, which indicates local traffic emission based on my knowledge. Then, should the diurnal variations cause by the changes in traffic conditions? Second, You did not show the diurnal cycle of E_{abs} . Moreover, I also do not understand the effect of mountain valley winds since you did not clearly explain that. Furthermore, could the variation in E_{abs} be due to changes in chemical composition? Third, You mentioned that after P5, BBOA was stable, and FFOA increased. However, I found BBOA decreased, and FFOA increased until midnight of 11/10, then decreased. I do not see you providing BBOA and FFOA concentration at other times. It will be interesting to see that. Last, you said the BCOC_N and BC were high in the last sentence. I am not sure what the period is. Could you clarify that? Moreover, I also do not understand how you conclude that fresh fossil fuel BC might mix with OC and nitrate at a high PM level.

- L190-191, "Such differences were ... during daytime." Have you considered the heterogenous nitro reactions during nighttime?
- L198, should it be Fig. S1 instead of Fig. S2? Also, Fig S2 is very confusing. What are the solid line and dashed line?
- Figure 4: what are these triangles in the figures? For figures 4 a and b, I suggest switching the x and y axis since the x axis depends on the E_{abs} , which depends on the $(NO_3+SO_4)/C_n$ and $(OC+Metal)/C_n$.
- L208-209, "These results indicated ... exceeded ~ 6 ." This is not clear to me. Can you explain a little bit more? You are not showing a time series of these two ratios. Does eBC increase with aging time? Also, I do not know how you calculate $(NO_3+SO_4)_{AMS}/eBC$. Based on your context, it seems like this ratio will keep increasing due to aging (although you did not provide the time evolution of that ratio). Then if that keeps increasing, why will the $(NO_3+SO_4)/C_n$ ratio stop increasing? I guess that eBC is initially increased due to increasing NO_3 and SO_4 , which have higher E_{abs} ($eBC = b_{abs}/MAC = E_{abs} * b_{abs_BC}/MAC$). NO_3+SO_4 increased at the beginning and then reached a steady state. Then something happened, which decreased the overall E_{abs} (maybe coating of organics?). Anyway, I might be wrong. However, please show me more evidence.
- L209-211, "Different from ... at both sites." What does this tell you? What is your purpose in comparing these two variables, and why do you not use the same format as Fig. 4a?
- L211-212, "Moreover, ... ($R^2=0.95$)". Where are these results presented?
- L223-225, "These results ... at the urban site." Did you compare mass spectra of TD and bypass? Could that be because OC and metals are less volatile, they were not removed by TD efficiently?
- L227-229, "Combined with ... at the rural site." How do you find that? Do you have any TEM images or EDX mapping? Mass spectra only can tell you these species exist in the same particle. Based on mass spectra, you cannot get morphology (partially coating or aggregate or core-shell).
- Equation 1-6: How did you get these equations? Why are there so many variables? What is the difference between 1 and 2, 3 and 4, 5 and 6? You did not provide any explanations for these equations.
- L232-233, "The predicted ... (Fig. S4)." Have you done any statistic analysis to show the correlation between predicted and measured E_{abs} ?
- L239-240, "Note that ... Factor5 in BJ." It is not clear which factor is which. Please clarify that either here or in the Methods section.