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Comment on acp-2022-105

Anonymous Referee #1

Referee comment on "Mixing state of black carbon at different atmospheres in north and southwest China" by Gang Zhao et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-105-RC1, 2022

This study presents the measurements of BC microphysical properties at three different sites spanning urban, semi-urban and rural environments over the North China Plain region. These are potentially valuable datasets in order to understand the evolution of BC over this anthropogenically influenced region. However, a few key issues regarding the atmospheric processes of BC have not been analysed and demonstrated sufficiently. Discussions in many places are loose and not referenced. The technical part, data analysis and discussion need expansion. I therefore suggest major revision, and list a few suggestions.

Major points:

1.The time-lag method of SP2 is associated with large uncertainties, which can only roughly separate the thinly and thickly coated BC, based on the time for the coating to be sufficiently evaporated and display a deceased scattering signal before incandescence. Given you have used the DMA-SP2 set up, I would suggest comparing this metric with the DMA-SP2 derived coating status at different core size range.

In addition, the more advanced technique using the coated over core size diameter derived from the SP2 should be acknowledged, particularly over this region on the ground (10.5194/acp-19-6749-2019) and vertical profiles (10.1021/acs.est.9b03722). These coating status of BC, rather than only separating the thinly and thickly coated BC, should be compared with your DMA-SP2 results.

At the moment, the DMA-SP2 method and time-lag has been mixed and it is not clear how the coatings have been calculated, and what metric have you actually used. Please check through the texts and make the statement clearer.

2. The definition of "fresh and aged" BC using time-lag method may be sometimes questionable, because not like traffic source, some other sources such as biomass burning (10.1029/2008GL033968) or solid fuel burning (10.5194/acp-14-10061-2014; 10.5194/acp-19-6749-2019) initially have a higher coating but may not be that aged. These should be referenced and discussed. I think the thinly and thickly coated BC is a fair classification for this purpose. Because you have three sites, which are three levels of ageing, then you can discuss the fraction of thickly-coated BC at different ageing scale at the three sites.

3. The method using the magnitude of scattering signal remaining after the particle experiencing evaporation in the SP2 laser beam, to indicate the attached or coated type of BC should be detailed. What is the threshold used, and what is the uncertainty. It may be useful to show readers how this has been happened, especially for the audience without previous knowledge of the SP2 instrument.

How this is related to the thickly-coated BC determined by time-lag method. Are we supposed to see a less fraction of attached BC when more thickly-coated?

4. Regarding the BC size, firstly, the all-BC core size distribution should be given, and the geometric mean and deviation should be compared with such measurements in this region (10.5194/acp-19-6749-2019). This may be source-related, or related with ageing time.

I would suggest performing the same lognormal fitting on Fig. 6 like in Fig. 5 to obtain the distribution of both core and coated sizes. The core size and coated size should be discussed together and possibly derive a general coating thickness for each site. The BC core distribution associated with each coated size will worth showing, in conjunction with and compared with other studies in this region, such as using uncoated and coated sizing to derive optical properties (10.1016/j.chemosphere.2020) and CCN properties (10.1021/acs.est.9b03722).

The possible issue is the defined "aged" BC had a smaller core than "fresh" BC is because the time-lag method is quite sensitive to the BC core size, as a smaller BC core will take a longer time to evaporate the coatings on it. This does not mean the BC is definitely "aged" (as aforementioned), but the higher time-lag one just has a smaller core and larger coating/BC core ratio, i.e. a larger relative coating thickness. Indeed, the aged BC should have a larger core if no precipitation experienced, because of coagulation among BC cores. This seemingly contradiction needs explanation.

About the coated BC size distribution, the rural site even had the smallest size for both modes of BC, compared to urban site. Shouldn't BC have larger size after ageing from urban to rural site? This needs explanation.

It is not careful that sometimes you used Dm or Dp for the coated BC diameter, which should be consistent throughout the texts.

Section 3.4 definitely needs expansion and referenced.

There have been wide observations that heavy pollution has caused substantial increase of BC coatings and sizes (10.5194/acp-18-9879-2018; 10.5194/acp-19-6749-2019), which should be compared with your study.

5. The DMA-SP2 has been used to derive the particle shape of BC-containing particles (10.1029/2021GL094522), which should be referenced to aid your results. The interesting part is you found the attached faction increased with larger Dp, which is essentially consistent with the results in the above reference that larger Dm will contain more fractal BC hard to be enveloped by coatings.

6. The coagulation between non-BC and BC-containing particles is an interesting one. It shows a higher concentration or more polluted condition caused a higher fraction of attached type. That means heavier pollution will lead to a higher fraction of attached type (not core-shell) BC, this is somewhat contradictory to the normal knowledge that for a more polluted condition, where more secondary aerosol forms, more condensation process could lead to a higher fraction of more coated and spheric BC, so less attached. This needs explanation. How are these related to the aged/fresh BC.

7. The mereological analysis is largely missing, at least like air mass backtrajectories, RH etc. should be analysed to show how these are related to your results.

Others:

Fig. 3. a study at remote site also using time-lag method could be also compared (10.5194/acp-10-7389-2010).

The conclusion really needs to be polished to show the key messages.

There are many places where techniques and discussions are mixed, I only list a few places, such as line 308; line 370-376, please also check through the texts to write as methods and discussions separated.

The title should use black carbon, not refectory BC, as the mixing state is associated with refractory BC and coatings. "in" different atmosphere. China is too broad, you may need to specify the region.

Line 23-28, there are too many numbers here, which really needs tidy up.

The conclusion about mixing state of BC is not clear. How to know the mixing is necessarily by coagulation. I probably understand you mean here, but the attached one only takes a small proportion but most is still driven by condensation.

Line 53-54, it should point out the mixing state of BC importantly determines its absorbing properties but Cappa et al. did not find that, and study found it displayed as two regimes (10.1038/ngeo2901). The statements of the references need correction.

Line 159, this sentence can't be one single paragraph.

The unit of Fig. 9 is %?

Can we make similar plots as in Fig. 9 for the other two sites.