

We would like to thank the reviewer for the valuable and constructive comments, which help us to improve the manuscript. Listed below are our point-by-point responses to the comments, including the corresponding changes made to the revised manuscript. The reviewers' comments are marked in black and our answers are marked in blue, and the revision in the manuscript is further formatted as *'Italics'*.

Referee #2:

The work developed a new approach to simulate the BC mixing state based on an emissions inventory and back-trajectory analysis. They quantified the mass-averaged aging degree of total BC particles and found the aging process of BC during atmospheric transport showed that it strongly dependent on emission levels. In general, from the modelling development to the modelling implication, I think that the result is worth to publish in the ACP after one minor revision.

1. Introduction P2L16-17 need references such as Li et al., JGR, 121(22), 13,784-713,798.

Response: Thanks. We have added some related references (Li et al., 2016; Jacobson, 2001; Moffet et al., 2009; Liu et al., 2017).

2. P2L39 The author review several method to determine the BC particles. Also, Wang et al., ESTL, 2017 measure the Df in polluted air following soot particle aging. Does the model possibly consider the dimension fraction of BC? That would be more interesting.

Response: Thanks for the comment. We have added some statement on change in BC morphology (quantified by dimension fraction) associated with BC mixing state under polluted conditions, as *“Moreover, Li et al., (2016) and Wang et al., (2017) revealed that the significant change in BC morphology (e.g. increase of fraction dimension) associated with their mixing state (i.e., from bare-like or partly coated to embedded BC) in polluted air, which could enhance BC light absorption (Liu et al., 2015; Cappa et al., 2012).”*

3. P7L24 deleted were P7L25 identified to indicated P7L27 import?

Response: Thanks. We have revised.

4. Change the word P9L2-L3 Here it is possible to consider how RH influence BC coating? If the authors want to get the conclusion, firstly you need to exclude other possible factors. Therefore, as you try to do it.

Response: Thanks. In our model calculation, we just considered the effect of emissions on BC mixing state, and excluded the influences of other possible factors. A rather simplified scheme was adopted where the aging rate is assumed proportional to the emissions. Actually, the aging rate coefficient (k) in our models was related to meteorological factors (e.g., temperature, relative humidity (RH)), chemistry (e.g., oxidation), aerosol phase state (e.g., heterogeneous reaction) as well as other parameter (e.g., particle size). The aging rate coefficient was presumed as a constant in the model calculation, and therefore the change in BC aging degree obtained from our simulations is only caused by distinct emissions.

To make this point clear, the related statement in the revised manuscript was “*In this study, a rather simplified scheme was adopted where the aging rate is assumed proportional to the emissions without detailed consideration of the effects of temperature, particle sizes, phase state, hygroscopicity and chemistry (Riemer et al. 2009; Cheng et al. 2008, 2012, 2015; Mu et al. 2018). Actually, these factors can influence the model parameter of aging rate coefficient. Our simulations strongly depends on the emission, initial value of D_p/D_c ratio and aging rate coefficient.*”

5. P9L6-13 Here some logical problem. if the aerosol in clean regions pass through the polluted area, how you know these BC don't deposit. How do you know BC at the sampling site were from clean region instead of the close polluted areas. Seemly, these are difficult questions about the mixture of air masses during the transport. How the authors separate them here?

Response: We thank the reviewer for raising these questions. To the first question, our model calculations consider the dry and wet deposit. In Eq. (7) in the manuscript, the TE represents the transport efficiency, taking into account transformation, transport, and removal processes of BC in the atmosphere. The TE is be treated as a weighting factor to quantify effective amount of BC from the emission origin to receptor site.

To the second question, we can separate BC from various emission origins (i.e., $0.25^\circ \times 0.25^\circ$ grid). Here, we discussed the gridded D_p/D_c ratio, representing the aging degree of BC particles that are transported to the receptor site from a certain origin (i.e., a grid h) rather than all origins.

Our model calculation can track the evolution of BC aging degree from a certain origin (i.e., a grid h) to the receptor site throughout different regions.

6. P10-P11, the implications and discussion should be combined? The implications should not be in Result section. The authors need to consider the structure in the two section.

Response: Thanks. We have changed “3.3 Implication for BC light absorption” into “BC light absorption in the atmosphere”

7. P12L10-11, capability should be individual BC or total here? My question is that the statement could be one problem. If you think total capability of BC in upper layer is higher than that of BC in the lower. You need to know the BC distribution in the column within PBL. Normally the BC concentration is much higher than them in upper layer. Even mixing state is higher in upper layer but their concentration is lower. Therefore, the total capacity need be questioned.

Response: Thanks. Sorry for the misleading and here the capability of BC light absorption represents the light absorption per unit BC mass, namely mass cross section (MAC) of BC. The MAC of BC strongly depends on their mixing state due to lens effect of coating materials on BC surface. The MAC and BC mass concentration (C_{BC}) are two factors to determine BC light absorption (σ_{ab}), namely $\sigma_{ab} = \text{MAC} \times C_{BC}$.

To make it clear, we have changed the “*capability of BC light absorption*” into “*capability of BC light absorption (i.e., mass cross section of BC-containing particles including coating materials on BC surface)*”.

8. P12 section 5 concluding remarks. Most of them repeat the results or others. Is that possible to shorten it? The details should not be concluded here such as P13L2-5 and so on. The reference should be removed in the section.

Response: Thanks. Following the reviewer’s suggestion, we have sharpened the Concluding Remarks section, as “*The effect of BC-containing particles on air quality and climate is not only dominated by BC mass concentration but also controlled by their mixing state. To better understand the mixing state of atmospheric BC in China, we developed a new approach to simulate the BC aging process during atmospheric transport based on the BC emission*”

inventory and back-trajectory analysis. Our models track the BC mixing state (i.e., D_p/D_c ratio) from an emitted source origin (e.g., a $0.25^\circ \times 0.25^\circ$ grid) to a receptor (i.e., Tsinghua site). The model calculation can quantify the mass-averaged D_p/D_c ratio of overall BC particles over the receptor site from various origins, which agreed well with observed ones. The simulations can provide information on the BC mixing state with fine temporal and spatial resolutions.

Based on the simulations of BC mixing state during atmospheric transport, we find a strong dependence of BC mixing state on emissions during atmospheric transport. BC particles with higher aging degrees at our site were mainly from more intensive emission origins (e.g., southern Hebei) due to higher aging rates. On the other hand, when BC particles emitted from clean origins passed through polluted regions, they were also characterized by a greater D_p/D_c ratio by speeding up the aging process in polluted air. Our simulations demonstrated the importance of regional transport in BC light absorption in Beijing under polluted conditions. This provides a new perspective on the phenomenon of pollution building up in Beijing, further demonstrating that this is driven by regional transport and transformation rather than local sources and processes.”

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