Reply to the comments of anonymous reviewer #3 on manuscript Entitled " Vertical distribution of black carbon and its mixing state in urban boundary layer in summer"

We sincerely appreciate the patient and insight comments and recommendations of the reviewer in improving this paper. Here, we will response to all the comments one by one as follows:

General Comments:

The authors can give more discussion on the significance of the work. In earlier studies, what had been left open, and what the present paper now to address? How their finding are relevant to atmospheric chemistry and radiative transfer? What the novel insights of their work are?

Reply: Thanks for the advice from the reviewer, we have rewritten the conclusion part to make the significance of the work clearer. (Line 405-411)

Specific comments:

(1) Abstract. Please indicate where the measurements are taken in the abstract and the date range of the measurements.

Reply: We have added the detailed information about the measurement location and time in the abstract. (Line 17)

In this study, vertical measurements were conducted through a moveable container based on a meteorology tower in Beijing urban area during June and July.

(2) SP2 data analysis: The minimum core size (from LII) and minimum total particle size (from scattering) differ, with the latter greater than the former. Have the authors filtered to account for any mismatch in the SP2 size for incandescence vs scattering, which can bias results? What are the limits on the Dp? When calculating BC concentration calculation, have the authors considered the SP2 limitations?

Reply: It is worth noting that the size range of pure scattering particles (~170-500 nm) differs from that of BC particles (~70-600 nm) due to the distinct responses of scattering and incandescence (LII) signals to particle size. This detection size range has been reported in the instructions of the SP2 instrument and in previous studies (Schwarz et al., 2008).

Regarding the BC core size (Dc), only the LII signal was used, so there is no mismatch between the LII and scattering signals.

The scattering signal was used to derive the optical diameter of BC-containing particles (Dp) through the LEO-fitting method. However, due to uncertainties in refractive indices, BC morphology, and the fitting area in the LEO method, Dp can sometimes be lower than Dc. We believe this is the mismatch referred to by the reviewer. In such cases, we retained the Dp derived from the LEO-fitting method. Although the Dp/Dc ratio may be lower than 1 for some individual particles, which is not physically correct, it did not affect the bulk characteristic of Dp/Dc. This mismatch has also been reported in previous studies(Laborde et al., 2013;Liu et al., 2019).

The calibration material and limited detection size range can affect the determination of BC concentration. To address this issue, we used the log-normal function to fit the BC size distribution and determine the missing fraction of BC mass concentration. This missing fraction was then used to calibrate the results. In the Method section (lines 110-117), we have described the major parameters such as refractive indices, calibration material, calibration compensation factor, and so on. Introducing additional parameters related to Dp and BC concentration determination would make the section too long, so we refer readers to our previous study (Liu et al., 2020) for more detailed information.

(3) Page 5/Line 149-151: The authors are trying to add some discussions on the causes of BC coating growth during ozone pollution day. However, there is no sufficient evidence to support. The authors can support their explanation by previous studies, e.g.https://doi.org/10.1021/acs.est.2c00090.

Reply: The reference provided by the reviewer mainly reported the influence of O_3 and atmospheric oxidation ability on BC's aging, which is well relevant to the discussion in this part. Thanks for the reviewer, we have added this reference to support our argument and extended our discussion. (Line 169-173)

Previous studies have suggested that BC aging could occur more rapidly in the presence of increased O3 concentrations (Zhang et al., 2022). Furthermore, Liu et al. (2020b) found the Dp/Dc could increase at a faster rate during the daytime under high Ox condition. These findings suggest that severe photochemical processes and high atmospheric oxidation rates could contribute to the aging of BC and the subsequent increase in its coating thickness.

(4) Page 6/Line 165-166: The author can give NO data rather than NOx to demonstrate titration reaction.

Reply: Thanks for the reviewer, we have now changed the expression NO_x to NO. (Line 185-187)

The presence of substantial NO emissions from traffic sectors would decrease the concentration of ozone through the titration reaction between ozone and NO, leading to the decreasing trend of ozone concentration with height.

(5) Page 6/Line 173-174: The author can discuss the emission sources of the measured BC based on MMD.

Reply: We have added more discussion about BC's core size to make the discussion clearer. (Line 194-197)

Studies have shown that the size of the BC core varies depending on the emission source. For instance, BC emitted from traffic sources typically has a lower MMD of about 150-180 nm, while that from solid fuel burning sources has a higher MMD of about 170-230 nm (Schwarz et al., 2008;Pan et al., 2017;Holder et al., 2014). In Beijing and London, (Liu et al., 2014;Liu et al., 2019a) found that the MMD of BC was much larger in winter than in summer, likely due to increased solid fuel burning for heating purposes.

(6) Page 6/Line 175-176: O3 decreased with increase height? I think that should be "O3 increased with ……".

Reply: Thanks for the reviewer, it's a typo. It should be "O_3 increased with …", we have modified the mistake. (Line 200)

The concentration of O3 increased with increasing height at a sharper slope than that in the uniform case.

(7) Page 7/Line 296-308: The measurement found higher O3 and Dp/Dc in the residual layer. The authors can give more discussion on BC aging in the residual layer. How the finding is relevant to atmospheric chemistry and radiative transfer?

Reply: We have added more discussion in this part. (Line 341-344 and Line 349-350)

The existence of aerosol, especially the absorbing aerosols, would depress the development of mixing layer (Ding et al., 2016;Wang et al., 2018b). The higher Dp/Dc of BC in the residual layer may amplify the absorbing of BC in the upper height in the early mornings, resulting in a more stable structure and deteriorating the air pollution.

Besides the high actinic flux, the higher O3 concentration may also contribute to the Dp/Dc increase in the upper height.

(8) Conclusions: The authors need to clear the novel insights of their work and the significance of their findings.

Reply: The conclusion part has been rewritten. (Line 379-412)

Reference

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Liu, D. T., Joshi, R., Wang, J. F., Yu, C. J., Allan, J. D., Coe, H., Flynn, M. J., Xie, C. H., Lee, J., Squires, F., Kotthaus, S., Grimmond, S., Ge, X. L., Sun, Y. L., and Fu, P. Q.: Contrasting physical properties of black carbon in urban Beijing between winter and summer, Atmospheric Chemistry and Physics, 19, 6749-6769, 10.5194/acp-19-6749-2019, 2019.

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