Reply to the comments of anonymous reviewer #2 on manuscript Entitled " Vertical distribution of black carbon and its mixing state in urban boundary layer in summer"

We sincerely appreciated the comments from the reviewer. Many advices from the reviewer are really helpful in the improving of this paper. We have carefully considered the comments and revised the paper. Here, we will response to all the comments one by one as follows:

Major comments:

(1) English expression needs further improvement, which is not bad but needs to be re-checked carefully. Some sentences are not clear enough.

Reply: We have followed the specific comments (see below) from the reviewer to make the expression clearer. Besides, we have also checked the whole manuscript several times to improve the words and sentences.

(2) A detail introduction of the vertical measurements of BC and its mixing state is missing. although the authors note that there are limited measurements, their results should be included for intact.

Reply: The vertical measurement about BC and its mixing state is limited and mainly conducted by aircraft. We have added more descriptions about BC's mixing state in the introduction and a brief summary about the vertical measurement about BC's mixing state in Table S2. (Line 51-53)

Limited vertical measurements of BC and its mixing state using the aircraft approach have been conducted (Katich et al., 2018;Schulz et al., 2019;Schwarz et al., 2013;Schwarz et al., 2017). Generally, BC concentration decreases with altitude, but the vertical distribution of size distribution and coating thickness could vary a lot due to the regional transportation (Hu et al., 2020), pollution levels (Ding et al., 2019;Zhao et al., 2020), biomass burning plumes (Ditas et al., 2018) and other factors. However, as summarized in Table S2, aircraft measurement mainly focuses on the vertical difference between boundary layer, upper troposphere and lower stratosphere. The data points in the boundary layer were limited due to flight height restrictions.

Table S2 Detail timetable of the vertical profile measurement

Measurement area	Measurement description	BC conc.	BC core Size distribution	Coating thickness	Reference
Europe	Aircraft measurement (0-1000m), EUCAARI campaign.	~6-200 ng/m3, decreasing with altitude.	With MMD of 150-210 nm, decreasing with altitude.	-	Ding et al., 2019)
Rural Beijing	Aircraft measurement (0-3000m).	~50-3000 ng/m3, decreasing with altitude.	With MMD of 160-230 nm, the vertical profile of MMD varied from cases.	Higher coating thickness in the boundary layer under pollution conditions.	(Ding et al., 2019;Zhao et al., 2020)
Arctic	Aircraft	1.4-50 ng/m3, generally	With MMD of 130-200	-	(Schulz et al.,

	measurement (0.1-5500m), NETCARE campaign	decreasing with altitude, but existing concentration peaks at certain height	nm, decreasing with altitude in Spring but uniform in summer.		2019)
Europe to North America	Aircraft measurement (2-20km)	1-40 ng/m3, decreasing with altitude	-	Significant coating thickness increase during plume affected period	(Ditas et al., 2018)
Global	Aircraft measurement, HIPPO and Atom campaign	1-10 ng rBC/kg air in the upper troposphere, 0.5-2 ng rBC/kg in the lower stratosphere	-	-	(Katich et al., 2018;Schwarz et al., 2013;Schwarz et al., 2017)

(3) Some conclusions are not supportive enough. for instance, the authors mentioned that the boundary layer in summer is relatively high. Then, how much does 0-240 m take up in the boundary layer? In addition, the variation in the boundary layer leads to such vertical profile type changes in Fig.3, but without showing the variations of boundary layer data. It is necessary to include boundary layer height throughout the day to support the discussion.

(4) "In fact, the several gradual decrease cases observed during the daytime generally occurred on cloudy days when the radiation was relatively weak." I do not find such data in Fig.3. Supporting data should be provided.

Reply: We have added data of mixing layer height and cloud amount in the discussion to support our discussion.

1) In the method section, we have added the sources of new data. (Line 100-104)

The height of boundary layer height (BLH) was determined (Tang et al., 2015;Tang et al., 2016) using the vertical profile of the attenuated backscatter coefficient measured by a single-lens ceilometer (CL51, Vaisala, Finland) at the same observation site. The cloud amount data was extracted from the reanalysis data product ECMWF Reanalysis v5 (ERA5, https://cds.climate.copernicus.eu/cdsapp#!/home) with spatial resolution of 0.25°*0.25°and time resolution of 1 hour.

2) In Fig. 1, we have added the time series of BLH and cloud amount.

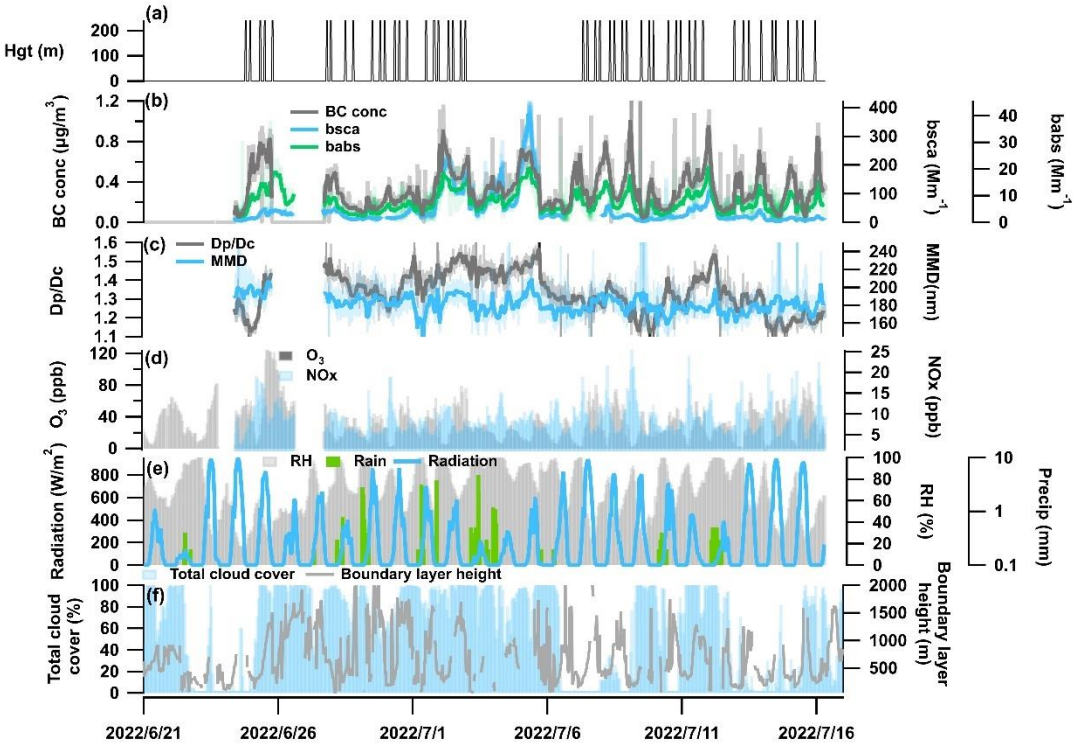

Figure. 1 Time series of the major observed parameters during the observation; the data were processed to a 1-hour resolution. (a) The height of the moveable container. (b) Mass concentration of black carbon and absorption (babs) and scattering efficient (bsca), the light-coloured lines denote the data with 1-min resolution. (c) The coating thickness (Dp/Dc) and mass median diameter (MMD) of BC, the light-coloured lines denote the data with 1-min resolution. (d) The mixing ratio of O3 and NOx. (e)(f) The meteorological conditions during

the observation period.

3) We have mentioned how much does 0-240 m take up in the boundary layer in the result section. (Line 149-152)

The BLH ranged between 50-2000 m (Fig. 1f) during the observation. The variation of BLH followed a clear diurnal pattern (Fig. S1) with an average value of 400-500 m at night and 1000-1200 m at noon. The observation height (0-240 m) was generally in the mixing layer. However, occasionally at night and in the morning, the BLH could be below 240 m, and the moveable container could detect the BC properties in the residual layer in such cases.

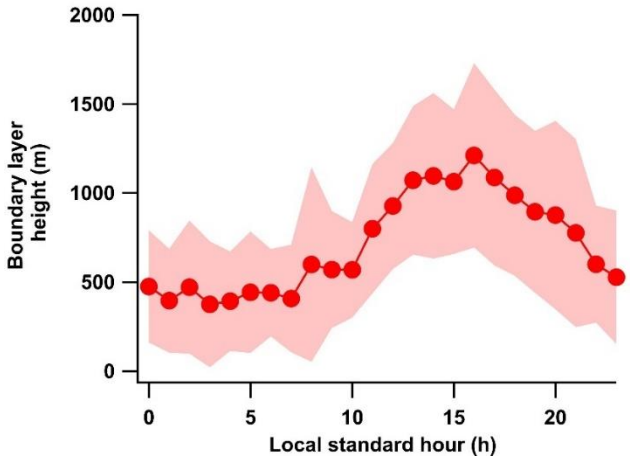

Figure. S1 Diurnal variation of BLH during the observation.

4) We have referred Fig. S1 in the discussion of the diurnal change of vertical profile type. (Line 256-257)

The diurnal variation in the boundary layer (Fig. S1) leads to such vertical profile type changes.

5) The measured BLH was also used to discuss the sudden decrease type. (Line 212-220)

Lei et al. (2021) found that in the stratified type, the BLH measured by ceilometer was relatively low (~110-250 m), which is consistent with the findings of this study. The BLH measured by ceilometer in the stratified type ranged between 100-300 m, which is lower than the average value of ~400-500 m. On the night of 27th June, the BLH measured by ceilometer even reached 80 m. It is known that the absolute value of BLH determined from different methods can vary significantly. Wang et al. (2018a) found that in the stratified type, the BLH determined from ceilometer would always be higher than the sudden concentration transition point and suggested that it is more precise to determine the BLH by using the sudden concentration transition point in such cases. In this study, the top of the BLH was regarded as the sudden concentration transition point in the sharp decrease type, and the profiles were used to investigate the differences in pollutants between the night boundary layer and residual layer in the following section.

6) The cloud data was used to discuss the gradual decrease type during the daytime. (Line 261-264)

In fact, several gradual decrease cases observed during the daytime, such as 11:00-12:00 on 2nd July (Fig. S13) and 19:00 on 27th June (Fig. S8), generally occurred on cloudy days when the radiation was relatively weak. As shown in Fig.1, the total amounts of clouds were nearly 100% for the two cases and the radiation was relatively low compared with that of the entire observation except several rainy days.

Specific comments:

1. Line 37: The expression of "the area" is not good. "the surrounding atmosphere" may be better.

Reply: Thanks for the reviewer. We have followed the advice. (Line 38)

Such depressing of the boundary layer is called the "dome effect" in which the BC in the upper boundary layer could heat the surrounding atmosphere, increasing the stability of the boundary layer.

2. Line 42: Change "study" to "studies".

Reply: Thanks for the reviewer. We have followed the advice. (Line 44)

The present studies mainly focused on the vertical distribution of BC mass concentrations (Wang et al., 2018a;Samad et al., 2020;Wu et al., 2021a;Guan et al., 2022).

3. Line 50: "Shiraiwa et al. (2010) found that BC's absorption ability after coating could reach two times greater than BC without coating." The relationship between BC's absorption ability and coating depends on many factors, the description here is not appropriate since the atmospheric conditions or particle properties are missing.

Reply: We have changed the sentence to make it clearer. (Line 51-53)

Through laboratory study, Shiraiwa et al. (2010) found that BC's absorption ability after coating could reach two times greater than BC without coating. Such absorption enhancement of BC due to coatings was also observed in the ambient with typical factors of 1.0-1.5 (Liu et al., 2017).

4. Line 59: What properties of BC has been measured? Concentration? Please describe clearly.

Reply: This sentence has been extended (See the reply for major comments). (Line 62-68)

5. Line 88: Please describe clearly again. The sentence should be "The concentration of pollutant gases (…), the concentration and mixing state of BC, and optical properties (…) of aerosol were measured…".

Reply: Thanks. The sentence has been changed. (Line 95-96)

The concentration of pollutant gases (NO2, O3, CO), the concentration and mixing state of BC and optical properties (light extinction and absorption) of aerosol were measured

6. Section 2.2: Eabs is calculated at a wavelength of 550 nm, but the Dp used in the calculation of Eabs is calculated at a wavelength of 1064 nm. Does this lead to higher uncertainty in the calculation of Eabs? Please describe it with more details.

Reply: The wavelength SP2's laser is 1064 nm. Thus, the D_p is derived at a wavelength of 1064 nm. We think the uncertainty in Eabs calculation at 550 nm is mainly caused by two factors:

1) The refractive indices

   The refractive indices of BC are wavelength dependent. In the determination of D_p, we used the 2.26-1.26i at wavelength of 1064 nm recommended by (Moteki and Kondo, 2008). In the

calculation of Eabs at 550 nm, we used the 1.95+0.79i recommended by (Bond and Bergstrom, 2006). We have mentioned the chose of refractive indices in the method section. (Line 111 and Line 140-141)

2) The morphology

Since the wavelength of 1064 nm is much larger than BC's diameter, the optical properties of BC is less sensitive to particles' morphology. Liu et al. (2015) found the bias of core-shell assumption in D_p/D_c determination would be less than 6%. However, for the calculation of Eabs at 550 nm, the morphology may play a more important role in BC's optical properties.

We have added the discussion of uncertainties from morphology. (Line 112-113 and Line 141-144)

7. It is mentioned that the core-shell assumption will lead to overestimation of light absorption. Is the light absorption shown in the result section corrected?

Reply: Since we didn't obtain the morphology information data of BC during this observation and the uncertainties of BC optical properties due to morphology may differ among different cases, we didn't correct the E_{abs} in result section. However, we have informed the readers about the extent of this overestimation by citing references. We would clarify it in the method section. (Line 141-144)

The Eabs may be overestimated in this study due to the core-shell assumption in the calculation (Liu et al., 2017;Wu et al., 2018;Liu et al., 2019b) because of the complex morphology of BC (Wang et al., 2017;Hu et al., 2021).

8. Line 139: Reword to "…, which is consistent with…".

Reply: Thanks for the advice, we have changed the expression. (Line 159)

The mass size distribution of the BC core followed a lognormal distribution with a mass median diameter (MMD) of 173 nm and a geometric standard deviation (GSD) of 1.57, as shown in Fig. S2, which is consistent with the MMD value (171 nm) observed in Beijing in the summer of 2018 (Liu et al., 2020b).

9. Lines 148-150: As mentioned above, June 25 is an ozone pollution day with a maximum hourly mixing ratio of 124 ppb. From figure 1, the ozone mixing ratio is about 40 ppb on July 5. Is this the ozone pollution day?

Reply: Thanks for pointing our mistake, it's a typo. The ozone pollution day is June 25, the D_p/D_c increased from 1.1 to 1.4 at that day. We have corrected it. (Line 169)

It is also noted that the Dp/Dc value could increase quickly from 1.1 to 1.4 during the ozone pollution day (25th June).

10. Lines 174-175: From figure S12, the concentration of O3 increase with increasing height in the vertical profile at 18:00 on July 2, which is not consistent with the description here.

Reply: It's another typo and also pointed by another reviewer. It should be 'increase' but not

'decrease', we have changed it. (Line 198-199)
The concentration of O3 increased with increasing height at a sharper slope than that in the uniform case.

11. Line 178: Including a reference here would be helpful.

Reply: Thanks, we have added the references. (Line 202)

This type is also referred to as the 'stratified type' in other studies (Guimaraes et al., 2019;Lei et al., 2021).

12. Lines 203-206: Please check carefully. The figure does not match with the text here. Figure S9 is not the vertical distribution on July 9. Moreover, the vertical distribution at 11:00 on July 9 is not sudden increase type in concentration of BC like the vertical profile at 11:00 on July 11. And, how do you figure out that the sudden increase in this case is due to solid fuel burning? Is there any other evidence here?

Reply: The vertical profile at 11:00 on July 9th is regarded as the sudden increase type in this study. It should be Fig. S16, we have corrected the text.

At 11:00, the BC concentration near the surface (0-50m) was significantly higher than at mid-air, and was characterized by an obviously thin coating and large MMD (denoted by the red circle). This suggests that the vertical profile at 11:00 was influenced by a fresh plume. Because there is no significant NO_x concentration increase and MMD increased instead of decreasing in this case. We thought the plume may be originated from solid fuel burning.

We have changed the expression to make it clearer. (Line 233-236)

The sudden increase that occurred at 11:00 on the 9th of July, as shown in Fig. S16, was accompanied by an increase in MMD, a decrease in Dp/Dc and no significant variation in NOx concentration. The sudden increase in this case may be influenced by the emission from solid fuel burning, since the MMD is generally higher for BC from solid fuel burning than that of fossil fuel burning (Schwarz et al., 2008;Pan et al., 2017;Liu et al., 2019a).

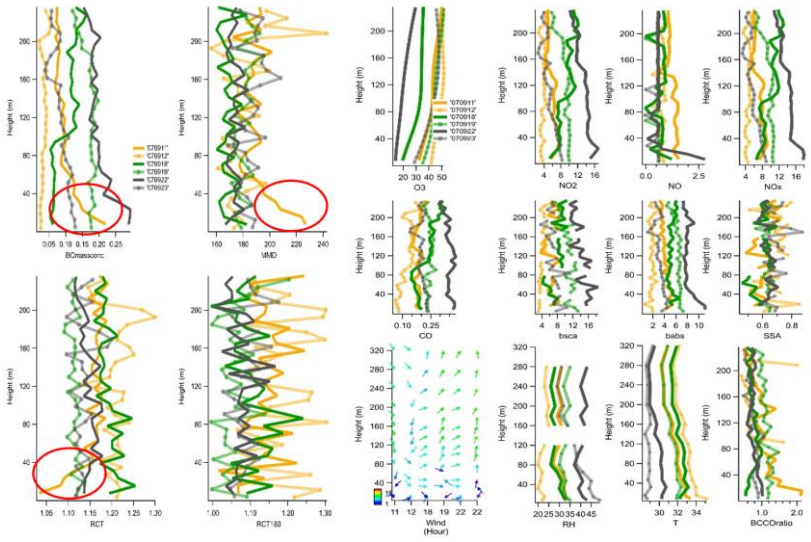

Figure S16 Vertical profiles during 9th July

13. Lines 208-209: Reword to "…the vertical profiles of BC, including its concentration and mixing state,…".

Reply: Thanks, we have modified the expression. (Line 239)

Fig. S7 shows the profiles on the ozone pollution day (maximum hourly ozone mixing ratio reached 124 ppb), the vertical profiles of BC, including its concentration, mixing state and ozone presented in a uniform type.

14. Lines 250-251: The figure does not match with the text again. From figure 4, the vertical profile of Dp/Dc at 23:00 slightly increase with height above 160 m. So the following explanation might be incorrect?

Reply: It's a typo, we have corrected it. Many thanks for the reviewer. (Line 283)

The Dp/Dc is nearly uniform below 160 m and slightly increases with height above 160 m.

15. Line 262: Reword to "…than that in the residual layer."

Reply: It has been reworded. (Line 295)

The BC properties, O3 and NOx at the 240 m height and ground are counted for the 5 sharp decrease profiles to study the pollutant difference between the ground and residual layers. For BC aerosols, the concentration is much higher on the ground than that in the residual layer.

16. Line 263: It is better to change "more coatings" to "thicker coating".

Reply: Thanks, we have followed the advice. (Line 296)

The BC concentration on the ground was 84.6% higher than that in the residual layer at 23:00 on the 8th of July. BC in the residual layer generally had thicker coatings, indicating a higher aging degree.

17. Lines 268-269: Note the preciseness of the data here. The MMD described here is inconsistent with that shown in the figure S7.

Reply: Thanks for the advice. We have changed it. (Line 301-302)

The MMD values were relatively stable (~170-190 nm) on the ground, while the MMD in the residual layer was more variable, ranging between 210 nm and 165 nm.

18. Lines 275-276 and 279: It should be "at 23:00 on June 27,…".

Reply: We have corrected it. (Line 309-312)

19. Line 304: Reword to "…at the top of the boundary layer during the daytime…".

Reply: We have changed the expression. (Line 336)

the pollutants from the residual layer firstly influence at the top of boundary layer during the daytime denoted by the large difference of O3 and Dp/Dc between 240 m and the surface at 6:00.

20. Line 343: Dp/Dc represents the relative thickness of the coating of BC-containing particle. But, Dp/Dc is the diameter ratio of BC-containing particle to BC core, which is not shell to core.

21. The conclusion section is really tedious, mostly repeating the results. I suggest the authors focus on new findings.

Reply: Thanks for the reviewer. We have rewritten the conclusion section. (Line 378-411)

References

Bond, T. C., and Bergstrom, R. W.: Light absorption by carbonaceous particles: An investigative review, Aerosol Science and Technology, 40, 27-67, 10.1080/02786820500421521, 2006.

Liu, D. T., Taylor, J. W., Young, D. E., Flynn, M. J., Coe, H., and Allan, J. D.: The effect of complex black carbon microphysics on the determination of the optical properties of brown carbon, Geophysical Research Letters, 42, 613-619, 10.1002/2014gl062443, 2015.

Moteki, N., and Kondo, Y.: Method to measure time-dependent scattering cross sections of particles evaporating in a laser beam, J Aerosol Sci, 39, 348-364, 2008.