General comments

This manuscript investigated diurnal variations of BrC in a megacity in Northeast China. The studied region is distinct, because it has quite different meteorological conditions and emission sources compared to well-known hotspots such as Beijing and surrounding regions. So far, however, aerosols in this region remained poorly understood with limited studies, e.g., regarding their chemical compositions, physical properties, sources and impacts. In this context, the authors conducted field measurements during two distinct seasons in a "largely unexplored" megacity in Northeast China, and traced the diurnal variations of BrC back to the changes of aerosol sources. The unique light absorption spectra of BrC observed during open burning episodes are especially interesting. Therefore, I think this manuscript represents a valuable contribution to better understanding of haze pollution in Northeast China, and my overall assessment is that it could be considered for publication in ACP after addressing the comments below.

Major point

MAE, which can be converted to the imaginary part of the complex refractive index of BrC, is an important parameter for climate models. In addition to a summary of the observational results, implications of this study should also be involved in the Conclusions section. To my understanding, although the winter is much colder in Northeast China compared to Beijing, MAE did not show apparent difference between these two regions. This is a potentially important point for the spatial distribution of MAE, but was completely ignored by the authors. In addition, the authors should make clear recommendations regarding whether the diurnal variations of MAE need to be considered in climate models.

Our responses: We agree with the reviewer that implications of our results should be clearly stated. Actually, this point was also raised by the handling editor. Thus a new section entitled "Implications" was incorporated into the revised manuscript **(see lines 472-490)**:

MAE_{365} and AAE are key parameters for simulating climate effects of brown carbon. In winter, although Harbin experiences low temperatures rarely seen in other Chinese megacities, the observed MAE_{365} and AAE were largely comparable with the typical results from other regions in Northern China (e.g., Beijing; Cheng et al., 2016). In addition, BrC's optical properties were indeed different between daytime and

nighttime samples, which were likely associated with increased HDDT emissions at night. However, the diurnal variations (~10% higher at night for both MAE_{365} and AAE) appeared negligible compared to uncertainties in simulating the mass concentration of BrC, i.e., organic aerosol. Thus for typical winter conditions in Northern China (without open burning), it may be practical to use fixed MAE_{365} and AAE values for estimating the wavelength-resolved absorption by organic aerosol in climate models.

The spring campaign suggested another scenario, that the agricultural fires exhibited strong influences on optical properties of brown carbon, as highlighted by the ~365 nm peak in BrC's absorption spectra. The distinct peak on one hand effectively elevated MAE_{365}, and on the other hand complicated the determination of AAE. In addition, the peak became less significant during the day, indicating that the organic compounds at play were likely subject to photo-bleaching. BrC emitted by the fires remained difficult to constrain, partially due to the variable combustion efficiencies. This in turn resulted in challenges for simulating climate effects of the open burning aerosols. Given the massive agricultural sector in Northeast China, more studies are necessary to understand the emissions, transformation and impacts of the fire-induced pollutants.

Specific Points

(1) Lines 34-35. Suggest towing down this statement.

Our responses: The sentence was changed to "*The presence of the ~365 nm peak complicated the determination of absorption Ångström exponents for the agricultural fire-impacted samples*" **(see lines 35-38)**.

(2) Line 75. Typical temperatures during winter in Beijing should also be given for comparison.

Our responses: Typical wintertime temperature was provided for Beijing (~0 ℃) as suggested **(see line 77)**.

(3) Line 96. Why did the daytime and nighttime samples have different sampling durations?

Our responses: The daytime and nighttime samples would have the same sampling duration (i.e., 8 hours) if the collection of the daytime samples was stopped at 17:00. However, the sunset time was typically between 16:00 and 17:00 during January in Harbin. Thus for the daytime samples, the sampling was ended at 16:00 and

consequently, their sampling duration (7 hours) was 1 hour shorter compared to the nighttime ones. Considerations in determining the starting and ending time were briefly explained in the revised manuscript: "*To avoid rush hours and considering the relatively early sunset time in winter (~16:00–17:00), daytime and nighttime samples were collected from 9:00 to 16:00 and from 21:00 to 5:00 of the next day, respectively*" **(see lines 97-98)**.

(4) Line 112. I think sonication could increase the extraction efficiency of BrC.

Our responses: We found that ultrasonicated and un-ultrasonicated methanol extracts showed comparable BrC absorption coefficients for ambient samples collected in Beijing. The comparison results were shown in Cheng et al. (2016), and we have provided this reference in the text for the extraction procedures **(see lines 113-114)**. To minimize the loss of insoluble carbon (e.g., EC), sonication was not applied in the present study when extracting the samples by methanol.

(5) Line 185. Suggest adding "compared to residential burning of crop residues" after "levels".

Our responses: The change was made as suggested **(see line 192)**.

(6) Line 277. Suggest adding "robustly" before "unfold".

Our responses: The change was made as suggested **(see line 285)**.

(7) Line 348. Is it necessary to introduce another indicator for open burning episodes?

Our responses: Here we introduced LG/OC because it could be used to not only identify open burning episodes but also estimate the strength of biomass burning impact. In other words, compared to LG/K^+, LG/OC is more suitable to be used to link the observed BrC optical properties with the influences of agricultural fires **(see lines 357-358)**.

(8) Line 363. This sentence is unclear, rewrite it.

Our responses: This sentence was rewritten as "*A benefit of using the new term was that a* $\ln(ATN_\lambda)^*$ *value of zero corresponded to* $ATN_\lambda = ATN_{LOD}$ *and thus* $\ln(ATN_\lambda)^*$ *could be considered "real" absorption by chromophores in solutions.*" **(see lines 373-377)**.

(9) Lines 405-407. I would like to see a scatter plot showing the dependence of *r* on *F* or *K*.

Our responses: The plot was provided as suggested **(see Figure S5 and line 417)**.

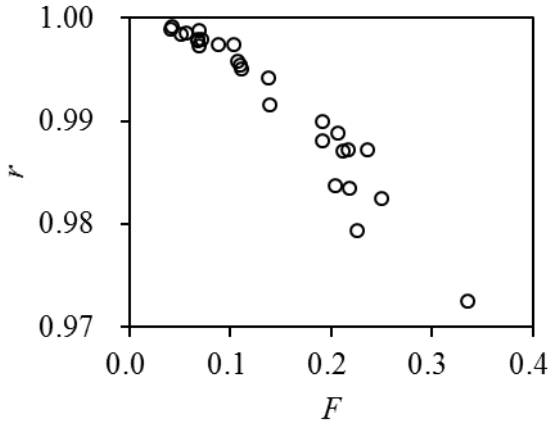

Figure R1. Dependence of *r* on *F* during agricultural fire episodes in spring. *r* was determined by regressing $\ln(ATN_\lambda)^*$ on $\ln(\lambda)$, while *F* was a measure of the significance of the ~365 nm absorption peak. *r* showed a clear decreasing trend with the increase of *F*. The same trend was observed when plotting *r* against *K*, another indicator for the significance of the ~365 nm absorption peak. This figure was presented as Figure S5 in the revised manuscript.

(10) Line 738. Suggest changing "another" to "the other".

Our responses: The sentence was re-written as "*the HC metropolitan area has two central cities as marked by the blue circles*" in the revised manuscript **(see lines 771-772)**.

(11) Line 752. Suggest changing "the same" to "a common".

Our responses: The change was made as suggested **(see line 786)**.

(12) Line 761. I think it is better to clarify again that LG/OC involved in the equation was on a basis of carbon mass and was in %.

Our responses: The change was made as suggested **(see line 795)**.

(13) Table S2. Re-write the note as: AAE were not provided due to the frequent occurrences of agricultural fires, which could result in distinct peak at ~365 nm for the light absorption spectra of BrC.

Our responses: The change was made as suggested **(see Table S3)**.