Anonymous Referee #1:

In this study, the authors examined light absorption of black carbon (BC) under clean and polluted conditions based on observations. They found that we found that the aging degree and light absorption capability of BC containing particles increased by 26-73% and 13-44% respectively, due to more coating materials on the BC surface. This work is interesting and merits publication after following comments addressed.

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

1. General Comments

The authors reported a large amount of BC was originated from sources outside Beijing based on effective emission intensity. It is true in this analysis. But the authors need to caution that they were comparing the contributions from a small region (Beijing) and a large region (outside Beijing or adjacent regions). In addition, the authors evaluated the contribution of local photochemical production by the changes of O3 concentrations in the atmosphere. They found that the O3 concentrations showed a different temporal trend. It only means weak photochemical production of O3 due to high aerosol concentrations blocking sunlight. It does not mean the local aging of BC is weak because high concentrations of aerosols may compensate the adverse conditions for BC aging. Anyway, the authors should provide uncertainty values for the numbers.

Response: We thanks the reviewer for raising these questions and we hope the reviewer will be clear after our detailed clarification below.

(1) In terms of comparing the contributions of BC from a small region (Beijing) and a large region (outside Beijing or adjacent regions), we agree with the reviewer that we need to caution. Noted that we are not paying attention to the difference between the contributions of BC from local Beijing and other regions (For example, the contributions of BC from local Beijing (~37%) during polluted period was smaller than that from other regions (~63%), partly due to comparing the contributions from a small region (Beijing) and a large region (outside Beijing)). In this study, we focus on comparing the contributions of BC from outside Beijing (considered as regional origins in this study) among different pollution levels (i.e., clean, slight polluted and polluted period). During polluted period, we found that the BC amount from regional origins (i.e., other regions not including local Beijing) accounted for ~21%, ~39% and ~63% of total BC amount in the site during the clean, slightly polluted and polluted periods, respectively. This revealed that the regional contribution to BC over Beijing increased as the air pollution levels increased.

To make this point clear, the related discussion has been revised in the manuscript, as "*In this study, the spatial origin of total BC in the site was classified into local Beijing and other regions (i.e., adjacent areas, considered as regional origins in this study). Noted that the local region (i.e., Beijing) defined in this study is smaller than areas outside Beijing (e.g., Hebei, Tianjin, Shanxi and Inner Mongolia (Fig. S1)). Table 1 lists the contribution of BC from regional origins (i.e.,* $EEI_{ousiede}/EEI_{total}$ *ratio). During polluted period, the contributions of BC from regional origins was ~63%, larger than that from local Beijing (~37%). This was partly due to comparing the contributions from a small region (Beijing) and a large region (outside Beijing). In this study, we focus on comparing the contributions of BC from outside Beijing (considered as regional origins in this study) among different pollution levels (i.e., clean, slight polluted and polluted period). The BC amount from regional origins (i.e., outside Beijing) accounted for ~21%, ~39% and ~63% of total BC amount in*

the site during the clean, slightly polluted and polluted periods, respectively. This revealed that the regional contribution to BC over Beijing increased as the air pollution levels increased."

(2) In terms of evaluating the contribution of local photochemical production by the changes of O_3 concentrations in the atmosphere, we have revised the related discussion, as "*When PM_1 concentrations were higher than ~120 μg m^{-3}, O_3 concentrations decreased to ~2 ppb. Zheng et al. (2015) has demonstrated the weakened importance of photochemistry in the production and aging of secondary aerosols in Beijing under polluted conditions due to decrease of oxidant concentrations. This indicated that the photochemical processing in BC aging may be weakened under higher polluted levels (i.e., PM_1>120μg m^{-3}). Noted that photochemical processing is not the only possible pathway in BC aging process and other pathways were not discussed in this study. The local aging process of BC might be enhanced by other pathways.* For example, *high concentrations of aerosols under polluted environment may compensate the adverse photochemical conditions for BC aging.* "

2. Specific comments

(1) Page 1 Line 14: It 'is' well known.

Response: Many thanks. We have revised it.

(2) Page 2 Line 22: What is the 'lens effect'?

Response: Thanks for the comment. We have stated/defined the "lens effect" in the revised manuscript according to the literatures (Bond et al., 2006; Fuller et al., 1999; Jacobson, 2001; Lack and Cappa, 2010): "*The non-BC species (i.e., coating materials) on the surface of BC can enhance BC light absorption via the lens effect (namely, the coating materials act as a lens to focus more photons on BC, Bond et al., 2006; Fuller*

et al., 1999; Jacobson, 2001; Lack and Cappa, 2010)"

(3) Page 7 Line 27: Missing ')'.

Response: Thanks. We apologize for the typo and have revised it.

(4) Page 8 Line 21: How many samples are there for different PM1 conditions?

Response: Thanks for the comments. In order to obtainthe evolution of D_p/D_c ratio and E_{ab} of BC-containing particles with size-resolved rBC cores with pollution development (shown in Fig. 2a), we used 28 different PM1 conditions.

To make this point clear, we revised the statement in the caption of Fig. 2a in the revised manuscript, as "*Figure 2. (a) The aging degree (D_p/D_c ratio) and light absorption capability (E_{ab}) of BC-containing particles with size-resolved rBC cores (D_c) under different PM_1 concentration (28 samples).*"

(5) Page 9 Line 27: It should be 'Figure 4'. The unit of EEI is 't/grid/year' shown in the figure. What does the 't' stand for?

Response: Thanks. We apologize for the typo and have changed "Figure 5" in P9/L27 into "*Figure 4*". The "t" stand for "ton", the unit of amount of air pollutant emission. We have changed "t/grid/year" into "*ton/grid/year*".

(6) Page 9 Line 30: 'account for' what? Does that mean the rest of the contribution is from Beijing's own emissions or emissions from other non-adjacent regions? As I understand, there are three emission source regions, emissions from Beijing, adjacent regions, and other regions. Please clarify.

Response: Thanks to the reviewer to point this out. The "account for" here represents the proportion of BC amount from adjacent regions in total BC amount in the site. In this study, the spatial origin of total BC in the site was classified into local Beijing and other regions (e.g., Hebei, Tianjin, Shanxi and Inner Mongolia).

To make it clear, we have revised the manuscript, as "*In this study, the spatial origin of total BC in the site was classified into local Beijing and other regions (i.e., adjacent areas, considered as regional origins in this study). Noted that the local region (i.e., Beijing) defined in this study is smaller than areas outside Beijing (e.g., Hebei, Tianjin, Shanxi and Inner Mongolia (Fig. S1)). Table 1 lists the contribution of BC from regional origins (i.e., $EEI_{ousiede}/EEI_{total}$ ratio). During polluted period, the contributions of BC from regional origins was ~63%, larger than that from local Beijing (~37%). This was partly due to comparing the contributions from a small region (Beijing) and a large region (outside Beijing). In this study, we focus on comparing the contributions of BC from outside Beijing (considered as regional origins in this study) among different pollution levels (i.e., clean, slight polluted and polluted period). The BC amount from regional origins (i.e., outside Beijing) accounted for ~21%, ~39% and ~63% of total BC amount in the site during the clean, slightly polluted and polluted periods, respectively. This revealed that the regional contribution to BC over Beijing increased as the air pollution levels increased.*"

(7) Page 10 Line 1: Does EEItotal include the contribution from Beijing's own emissions?

Response: Thanks and yes. The EEI_{total} includes the contribution from Beijing's own emissions, calculated by Eq. (8) in the manuscript.

(8) Page 10 Line 3: I am confused that EEItotal increased by a factor of 4.6, but after that, the authors said BC from adjacent area. Should it be EEIadjacent? In addition, I think it needs a supplement to the conclusion that the increased BC is due to transport of polluted air mass, not the adverse local meteorology. It is true for a very small region, based on the analysis of this study, because the authors were comparing the contributions from a small region and a large region. Also, polluted events always occur over a larger region, even spread the whole eastern China. They are definitely caused by adverse meteorology. The more transport of pollutants into Beijing is

probably a consequence of increased pollutants due to adverse meteorology in other regions. For example, Yang et al. (2017) analyzed the source-receptor relationship of BC in China and found that, during polluted days in winter, the increases in BC over the North China Plain (including Beijing) is dominated by its local emissions instead of regions outside North China Plain. The weakening of winds can explain it.

Response: Thanks for the comments. The EEI given here is the total EEI (EEI_{total}, calculated by Eqs. (7) and (8) in the manuscript) including BC from Beijing and other regions. The EEI_{total} can be used to characterize the total BC amount (unit of ton/year, not the BC concentrations) transported to the site. The EEI_{total} strongly depends on BC emission of source origins (including local Beijing and other regions) and dry/wet deposition during atmospheric transport. Considering the change of BC emission from local Bejing under different pollution levels was slight, the variations in EEI_{total} was dominated by BC from different regional origins (i.e., higher EEI_{total} due to BC from regional origins with higher emission (e.g., south of Hebei) and lower EEI_{total} due to BC from regional origins with lower emission (e.g., Mongolia)). On the other hand, the effect of local meteorology on EEI_{total} is slight. However, BC concentration in the site strongly depends on both total BC amount (transported from local Beijing and other regions, characterized by EEI_{total} in this study) and local meteorology. In this study, we found that the EEI_{total} and BC concentrations from the clean period to the polluted period increase by ~4.6 times and ~7.4 times, respectively, revealing that the increase of EEI_{total} account for ~62% of the increase in BC mass concentration. This indicated that the adverse local meteorology contributed ~38% of the increase in BC mass concentration in the site from the clean period to the polluted period.

We agreed with the reviewer that less effect of adverse local meteorology is due to that local Bejing is smaller than other regions (e.g., Hebei, Tianjin, Shanxi and Inner Mongolia). Polluted events always occur over a larger region and are definitely caused by adverse meteorology. For our case, the adverse meteorology during polluted days in the whole large region including Beijing and other adjacent areas can lead to the increase of pollutants and then more transport of pollutants into Beijing.

Following the reviewer's suggestion, we have revised the statement to assess the effect of regional transport and adverse local meteorology on BC increase under polluted conditions, as "*Table 1 shows that the EEI_{total} was 4.6 times higher during the polluted period than during the clean period, revealing that polluted air mass brought more BC to Beijing. BC concentration in the site strongly depends on both total BC amount (transported from local Beijing and other regions, characterized by EEI_{total} in this study) and local meteorology. Table 1 shows that the BC concentrations from the clean period to the polluted period increase by ~7.4 times. The increase of EEI_{total} (~4.6 times) accounted for ~62% the increase in BC mass concentrations (~7.4 times). This indicated that the adverse local meteorology contributed ~38% of the increase in BC mass concentration in the site from the clean period to the polluted period. Compared with regional transport, less effect of adverse local meteorology might be attributed to relatively small areas defined as the local region (i.e., Beijing) in this study. Polluted events in China always occur over a large region, e.g., North China Plain (Yang et al., 2017; Zheng et al., 2015). For our case, the adverse meteorology during polluted days in the whole large region including Beijing and other areas can lead to the increase of pollutants and then more transport of pollutants into Beijing. Yang et al. (2017) found that the increases in BC concentration under polluted conditions over the North China Plain (including Beijing and other adjacent areas) is dominated by its local emissions due to adverse meteorology.* "

(9) Page 11 Line 1: What does the normalized EEI mean? Is it a percent value or some index?

Response: Thanks to the reviewer to point this out. In this study the EEI_{total} was normalized by scaling by a factor of 10^{-3}, namely $EEI_{total,normalized} = EEI_{total}/1000$. To make it clear, we have added the statement in the caption in Fig.6 in the revised manuscript, as "*The normalized EEI_{total} ($EEI_{total,normalized}$) was calculated by scaling by a factor of 10^{-3}, namely $EEI_{total,normalized} = EEI_{total}/1000$.* "

(10) Page 11 Line 19: I see the author calculated DRF by scaling the average DRF (0.32 W m-2) of externally mixed BC with an average MAC of 7.5 m2 g-1 from various climate models (Bond et al., 2013). Is the DRF value global average with a fixed BC climatology? The author should make it clear, or the readers may think the value is the DRF over Beijing during the analyzed clean and polluted days.

Response: We thank the reviewer for raising the important issue. In this study, the DRF (0.31 W m^{-2}) of externally mixed BC was the global averages from the global climate models listed in Table R1 in the response (Table S1 in the revised manuscript). The calculated DRF of BC-containing particles (shown in the Fig.7 in the revised manuscript) was obtained by scaling the average *DRF* (0.31 W m^{-2}) of externally mixed BC from various global climate models (Bond et al. 2013) with a scaling factor of E_{ab} under different PM_1 concentrations. Noted that the DRF values calculated here did not consider the change of BC amount under different pollution levels. In this study, we focused on investigating the effect of BC light-absorption capability on DRF. Therefore, the increase in DRF of BC with increasing pollution levels just considered the change in light-absorption capability of BC.

Table R1 (Table S1 in the revised manuscript) The DRF of externally mixed BC from global climate models. The modeled values were taken from Bond et al. (2013).

Global climate Model	Mixing state	Modeled MAC (m^2 g^{-1})	Modeled DRF (W m^{-2})	Reference
AeroCom models				
GISS	External	8.4	0.22	Schulz et al. (2006)
LOA	External	8.0	0.32	Schulz et al. (2006)
LSCE	External	4.4	0.30	Schulz et al. (2006)
SPRINTARS	External	9.8	0.32	Schulz et al. (2006)
UIO-CTM	External	7.2	0.22	Schulz et al. (2006)
UMI	External	6.8	0.25	Schulz et al. (2006)
Other models				
BCC_AGCM	External	4.3	0.10	Zhang et al. (2012)
CAM3 ECA	External	10.6	0.57	Kim et al. (2008)
GISS-GCM II	External	7.8	0.51	Chung and Seinfeld (2002)
Average values		7.5	0.31	

To make it clear, we added the Table S1 and the related discussion in the revised manuscript, as "*The DRF values for BC-containing particles at different pollution levels were obtained by scaling the average DRF (0.31 W* m^{-2}*) of externally mixed BC from various climate models (Bond et al. 2013) with a scaling factor of the calculated* E_{ab} *under different* PM_1 *concentrations (Fig. 2b). The DRF (0.31 W* m^{-2}*) of externally mixed BC was the global averages from the global climate models listed in Table S1. In order to point out the effect of BC light-absorption capability on DRF under different PM1 concentrations, we did not consider the changes of BC amount for DRF calculation.*"

(11) Page 12 Line 15: Delete 'by'.

Response: Thanks. We have revised it.

(12) Page 13 Line 19: It was defined as transport-controlled 'period'.

Response: Thanks. We have changed "region" into "period".

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