Anonymous Referee #2

Summary:

This paper discusses measurements of black carbon (BC) made with an AE33 aethalometer and a SP-AMS in the North China Plain (NCP) during winter in 2017-2018. These observations are discussed in the context of recent emission reduction policies. Initial laboratory experiments were made of different fuel sources in an environmental chamber to develop a model to interpret the aethalometer measurements. This model was then applied to 2 months of measurements at a ground site, and the results are interpreted in the context of WRF-Chem modeling results to estimate relative contributions from regional sources and the BC radiative effect. The relative contributions of liquid fossil fuel sources and solid fuel sources were evaluated, and these relative sources were discussed in the context of their contribution to the regional BC radiative effect.

The observations and the analysis presented here are a valuable contribution to the literature, given the importance of BC as a climate forcer and the NCP as a significant anthropogenic source region for these aerosols. The conclusion that focusing on a reduction of BC from solid fuel sources could lead to greater gains than liquid fuels is an important conclusion for policy makers.

While it's clear that a significant amount of work went into the acquisition of measurements and the analysis of results presented in this study, the discussion is often challenging to follow. While each separate part of the measurements, analysis, and modeling were described in Section 2, it is often not clear which measurement or model contributed to the results discussed in Section 3. The paper would also benefit from a more clear discussion of the BC observations in light of best practices for discussing observations from different instruments established in the recent literature. In addition, the radiative forcing calculation is not clearly described.

Response: The authors appreciate the reviewer's valuable suggestions, and we believe that the revised manuscript has been significantly improved after addressing the comments. Below are the point-to-point responses, and the modifications to the

manuscript are marked.

General Comments:

(1) Observations of BC with the different instruments used in the study should be more clearly distinguished (eBC, rBC) as discussed in Lack et al. 2014. This is an important issue when comparing observations between different instruments, as BC is operationally defined, and this point should be made clearly in the discussion. This is particularly confusing in the discussion of the source-specific AAE model, as it seems both observations from the aethalometer and the SP-AMS are used to determine the source-specific AAE values.

Response: We followed the reviewer's suggestion and clarified the term of BC measured by different instruments. According to the review studies of BC by Lack et al. (2014) and Petzold et al. (2013), we used "equivalent BC (eBC)" for AE33 aethalometer measurements and "elemental carbon (EC)-containing particles" for SPAMS measurements. The source-specific AAE was calculated based on the aerosol light absorption which was measured by AE33 aethalometer. We have clarified this in the revised manuscript. It now reads as follows:

> "Table 1 summarizes the average AAEs obtained from the sources of liquid fossil fuels and solid fuels. These source-specific AAEs were calculated using $b_{abs}(370)$ and $b_{abs}(880)$ (Eqs. 3 and 4)."

(2) It is also important in the context of the BC radiative effect calculation, as observations of BC mass loadings can differ by a factor of 2 or more when comparing different measurement techniques.

Response: We agree with the reviewer that different BC instruments induce uncertainties in BC measurements in studies. In the revised manuscript, we clarified this point, and it reads as follows:

> "Compared to previous DRE obtained from the SBDART model, the atmospheric DRE derived by eBC values in this study (+18.0 ± 9.6 W m^{-2}) was

comparable to that of South China (+17.0 W m^{-2}, Huang et al., 2011) but was lower than that of Northwest China (+16.6 to +108.8 W m^{-2}, Zhao et al., 2019). In addition to the varying BC burden in different areas, the BC measurement techniques used in different studies may also contribute to the differences in BC DRE calculations."

(3) The discussion of the radiative forcing is not very clear, and I had a hard time following how this calculation was performed. More details need to be given. In addition, this appears to be a calculation of the BC radiative effect (rather than radiative forcing), as described in Heald et al. 2014.

Response: After carefully read the study of Heald et al. (2014), we changed the original used 'radiative forcing' to 'radiative effect' in the revised manuscript. Additionally, we added more detailed description about the calculation of BC radiative effect. It now reads as follows:

"Aerosol direct radiative effect (DRE) (Heald et al. 2014) at the top of the atmosphere (TOA) or at the Earth's surface (ES) is the difference between the incoming (↓) and outgoing (↑) solar fluxes (F) with and without aerosols:

$$\mathrm{DRE} = (\mathrm{F}\downarrow - \mathrm{F}\uparrow)_{\text{with aerosol}} - (\mathrm{F}\downarrow - \mathrm{F}\uparrow)_{\text{without aerosol}} \quad (8)$$

The aerosol DRE in the atmosphere was calculated by subtracting the DRE at the Earths' surface from the DRE at the top of the atmosphere.

In this study, the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model that was developed by Ricchiazzi et al. (1998) was used to perform the radiative transfer calculations in the shortwave spectral region of 0.25–4.0 μm. The SBDART model is a widely used tool for estimating aerosol DRE in the atmosphere (e.g., Zhang et al., 2017; Rajesh and Ramachandran, 2018; Boiyo et al., 2019). A detailed description of this model can be found in Ricchiazzi et al. (1998). The aerosols' optical depth, single scattering albedo, and asymmetric parameters are essential input factors in the SBDART model. These

optical parameters were estimated using the Optical Properties of Aerosols and Clouds (OPAC) model (Hess et al., 1998). Detailed calculations are shown in Text S3. Moreover, the surface albedo, solar zenith angle, and atmospheric parameter profiles are also important input factors in the SBDART model. The surface albedo was derived from the Moderate Resolution Imaging Spectroradiometer (https://modis-atmos.gsfc.nasa.gov/ALBEDO/index.html, last access: November 2019). The solar zenith angle was estimated using the latitude, longitude, and sampling time of the location. The atmospheric vertical profiles (including vertical distributions of temperature, pressure, water vapor, and ozone density) of mid-latitude winter embedded in the SBDART model were used."

(4) Overall, the paper could benefit from English language editing.

Response: The revised manuscript was polished by an English language editing agency.

Specific Comments:

(5) The title is quite long.

Response: We shortened the title as follows:

"Measurement report: Source and mixing state of black carbon aerosol in the North China Plain: Implications for radiative effect"

(6) Page 1, lines 23-25. It is unclear here how the "local emissions" differ from the "emissions in the NCP". If this is meant to differentiate local emissions relative to regional emissions this could be more clearly stated.

Response: Yes, it means to differentiate local emissions from regional emissions. To make it clearer, we revised this sentence in the manuscript. It now reads as follows:

"The air quality model indicated that local emissions were the dominant contributors to eBC at the measurement site. However, regional emissions from NCP were a critical factor for high eBC pollution."

(7) A schematic of the instrument setup during the environmental chamber experiments and the measurement campaign could be a helpful addition. Additionally the map showing the location of the measurement site (S1) would be helpful in the main text.

Response: Following the reviewer's suggestion, we have added the schematic of instrumental setups of our ambient measurements (see Figure R1 below and Figure S1) and source experiments (see Figure R2 below and Figure S4) in the revised supplementary material. Moreover, map of the sampling location was put in the main text (see Figure 1 in the revised main text).

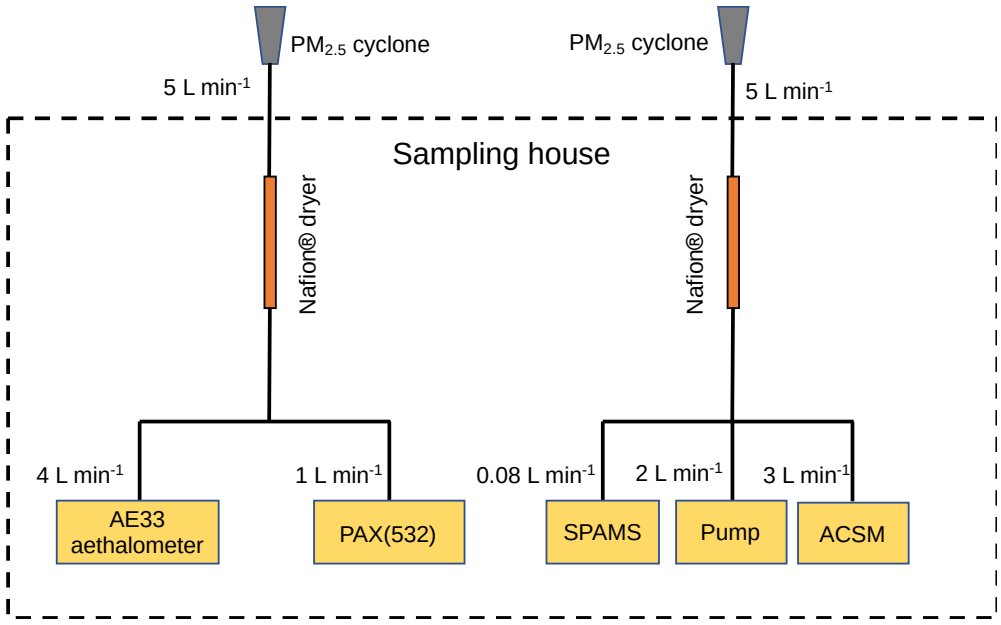

Figure R1. Schematic presentation of the instrumental setups of the ambient aerosol measurements.

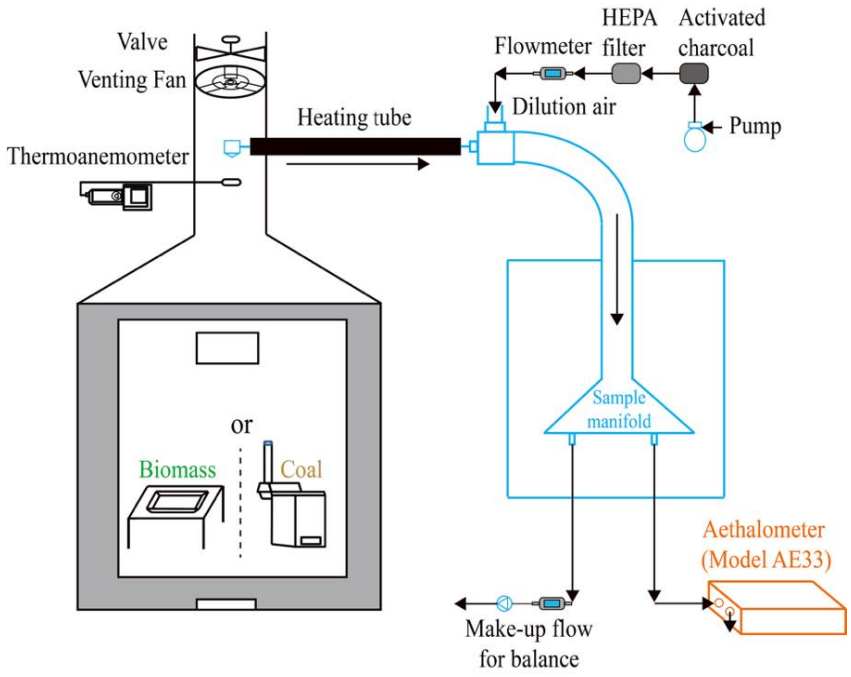

Figure R2. Schematic presentation of the instrumental setups of source experiments of biomass burning and coal combustion.

(8) Some more details would be useful for the observations from the source emission experiments in order that these could be more easily compared with other similar experiments. At what point during the burns was the emitted smoke measured?

Response: We thank the reviewer's insightful suggestion. In the revised manuscript, we added more description of the source emission experiments, which includes the information concerned by the reviewer. It now reads as follows:

"A custom-made passivated aluminum chamber (~8 m^3) was used to characterize the emission of solid fuels (i.e., biomass and coal) (Fig. S4). Performance evaluation of this chamber was done by Tian et al. (2015). Several types of biomass residues (wheat straw, rice straw, and corn stalk, cotton stalk, sesame stalk, soybean straw, and firewood) and coal (bituminous coal and honeycomb briquet) were used to represent biomass burning and coal combustion that occurs in the NCP. Each weighted sample was burned on a platform or in a stove that was placed inside the combustion chamber. For

biomass burning, the chamber background $b_{abs}(\lambda)$ was measured by AE33 aethalometer before ignition. When the background $b_{abs}(\lambda)$ was close to zero and stable, a propane torch was used to ignite the biomass on the platform. For coal combustion, a burned-out honeycomb coal in the stove was used as the igniter after the background $b_{abs}(\lambda)$ was small and stable in the chamber. The emitted smokes of each burn test were first diluted by a Model 18 dilution sampler (Baldwin Environmental Inc., Reno, NV, USA) before AE33 aethalometer measurements (Fig. S4). The $b_{abs}(\lambda)$ used to estimate the AAE was averaged over the entire period of each burn from ignition to $b_{abs}(\lambda)$ back to the background.

The motor vehicle exhaust emissions were performed using a LDWJ6/135 detection system of loading and speed reduction on the light duty diesel vehicle (Shenzhen Huiyin Industrial Development Co., Ltd, Shenzhen, China). This system contains two different sizes of expansion cylinders that are used to carry the driving wheels of the vehicles. Fig. S5 shows the schematic presentation of the instrumental setup of motor vehicle exhaust emissions. Gasoline and diesel cars at idle and at different driving speeds (i.e., 20 and 40 km h^{-1}) were tested. The automobile exhaust smoke particles were collected using a particle sampling probe in the exhaust pipe. The particles were dried by a silica gel dryer before AE33 aethalometer measurement. The measured $b_{abs}(\lambda)$ used to estimate the AAE was averaged over the period that the driving speed was relatively stable."

(9) Do the values in Table 1 represent average values of the aerosol optical properties during the entire period of the burn? What does the "test number" in Table 1 refer to? Is this the number of experiments performed? This should be made clear in the caption.

Response: Yes, the aerosol optical properties in Table 1 were calculated using the average light absorption of the entire period of each burn. The test number denotes the number of performed experiments. We added notes in the revised Table 1 (also see Table R1 below).

Table 1. Summary of aerosol absorption Ångström exponent (AAE) obtained from source experiment.

	Solid fuel				Liquid fuel	
	Crop residues	Firewood	Bituminous coal	Honeycomb briquet	Gasoline	Diesel
Maximum	3.3	3.2	1.4	5.2	1.5	1.3
Minimum	1.6	2.7	1.0	2.2	1.4	1.0
Average[a]	2.4	2.9	1.1	4.0	1.5	1.2
S.D.[b]	0.4	0.2	0.2	0.9	0.1	0.1
Test number[c]	30	4	4	16	3	7

[a]The average value was calculated using the light absorption of the entire period of each burn.
[b]S.D. represents standard deviation.
[c]Test number denotes the number of performed experiments.

(10) I found the discussion in 3.1.1 and Figure 1 to be unclear. Given the range of values for AAE for the different fuel types shown in Table 1, the limitations of the aethalometer model, which assumes a single value for AAE for liquid fuels and solid fuels, should be more clearly discussed in the text.

Response: From the computational equations, the limitation of 'aethalometer model' is mainly from the applied source-specific AAE. Due to the high variation in the solid fuel AAE (e.g., 1.1–4.0 in this study), the selection of different value in the 'aethalometer model' can cause uncertainties in estimation of contribution from each source (e.g., solid fuels and liquid fuels) to total BC mass. Most of the current studies used source-specific AAEs from previous publication (e.g., Healy et al., 2017; Rajesh and Ramachandran, 2018; Zheng et al., 2019). Some other studies obtained the source-specific AAE through comparison with results from external source apportionment methods, such as ACSM-based organic aerosol (OA) sources (Ealo et al., 2016) and ^{14}C technique (Martinsson et al., 2017; Zotter et al., 2017). In this study, we conducted the source experiments to obtain the possible suitable source-specific AAEs. In addition, we performed sensitivity analyses by comparing results of BC source apportionment with different OA sources (i.e., HOA and BBOA+CCOA) to verify the rationality of the applied AAE values. In the revised manuscript, we added

the limitation of the 'aethalometer model'. And based on the comment from Reviewer 1, we changed the term "Aethalometer model" to "multi-wavelength optical method" in the revised manuscript. It now reads as follows:

"From Eqs. 1–4 of the multi-wavelength optical method, its limitation is attributed to the choice of source-specific AAE. Since AAE exhibited high variations (e.g., 1.1–4.0 in this study), different AAE selections may lead to uncertainties when estimating the contributions of solid fuels and liquid fuels to eBC mass. In this study, the obtained average AAE_{lff} (1.3) and AAE_{sf} (2.8) were applied in the multi-wavelength optical method to obtain eBC source apportionment. A sensitivity test for each eBC source and organic aerosol (OA) subtype was further performed to verify the rationality of the used AAEs."

References:

Ealo, M., Alastuey, A., Ripoll, A., Pérez, N., Minguillón, M. C., Querol, X. and Pandolfi, M.: Detection of Saharan dust and biomass burning events using near-real-time intensive aerosol optical properties in the north-western Mediterranean, Atmos. Chem. Phys., 16(19), 12567–12586, https://doi.org/10.5194/acp-16-12567-2016, 2016.

Healy, R. M., Sofowote, U., Su, Y., Debosz, J., Noble, M., Jeong, C.-H., Wang, J. M., Hilker, N., Evans, G. J., Doerksen, G., Jones, K. and Munoz, A.: Ambient measurements and source apportionment of fossil fuel and biomass burning black carbon in Ontario, Atmos. Environ., 161, 34–47, https://doi.org/10.1016/j.atmosenv.2017.04.034, 2017.

Martinsson, J., Abdul Azeem, H., Sporre, M. K., Bergström, R., Ahlberg, E., Öström, E., Kristensson, A., Swietlicki, E. and Eriksson Stenström, K.: Carbonaceous aerosol source apportionment using the Aethalometer model – evaluation by radiocarbon and levoglucosan analysis at a rural background site in southern Sweden, Atmos. Chem. Phys., 17(6), 4265–4281, https://doi.org/10.5194/acp-17-4265-2017, 2017.

Rajesh, T. A. and Ramachandran, S.: Black carbon aerosols over urban and high altitude remote regions: Characteristics and radiative implications, Atmos.

Environ., 194, 110–122, https://doi.org/10.1016/j.atmosenv.2018.09.023, 2018.

Zheng, H., Kong, S., Wu, F., Cheng, Y., Niu, Z., Zheng, S., Yang, G., Yao, L., Yan, Q., Wu, J., Zheng, M., Chen, N., Xu, K., Yan, Y., Liu, D., Zhao, D., Zhao, T., Bai, Y., Li, S. and Qi, S.: Intra-regional transport of black carbon between the south edge of the North China Plain and central China during winter haze episodes, Atmos. Chem. Phys., 19(7), 4499–4516, https://doi.org/10.5194/acp-19-4499-2019, 2019.

Zotter, P., Herich, H., Gysel, M., El-Haddad, I., Zhang, Y., Močnik, G., Hüglin, C., Baltensperger, U., Szidat, S. and Prévôt, A. S. H.: Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer-based source apportionment using radiocarbon measurements of ambient aerosol, Atmos. Chem. Phys., 17(6), 4229–4249, https://doi.org/10.5194/acp-17-4229-2017, 2017.

(11) Does Table 2 give observations from the SP-AMS? As this discusses the number of particles, that would seem to be the case but this should be clearly stated (e.g. by referring to rBC rather than BC).

Response: Yes, Table 2 shows the observations from SPAMS. As replied in comment (1) above, the 'BC' in table was revised to 'EC' as shown in Table R2 (also see Table 2 in the revised manuscript).

Table R2. Summary of names, numbers, and fractions of six types of elemental carbon (EC)-containing particles determined by a single particle aerosol mass spectrometer.

Group	Number of particles	Fraction of particles (%)
EC internally mixed with OC and sulphate (EC-OCSOx)	235874	51.9
EC internally mixed with Na and K (EC-NaK)	107272	23.6
EC internally mixed with K, sulphate, and nitrate (EC-KSOxNOx)	75227	16.6
EC from biomass burning (EC-BB)	26307	5.8
Pure-EC	5083	1.1
Unidentified EC (EC-others)	4670	1
Total EC-containing	454433	100

(12) Page 11, lines 13. How was this percent contribution determined? Is this from the WRF-Chem model or does this use the observed BC? Is the BC concentration shown in Figure 5 from the model results or from the observations?

Response: The percent contributions of local emissions and regional transport were obtained from WRF-Chem model. The BC concentration shown in Figure 5 was the observation values. We clarified in the manuscript, and it now reads as follows:

"As shown in Fig. S8, six source regions were identified in the WRF-Chem model to quantify the contributions of local emissions and regional transport to observed eBC mass. The information on each source region is summarized in Table S2, and their contributions to observed eBC mass are shown in Fig. 5."

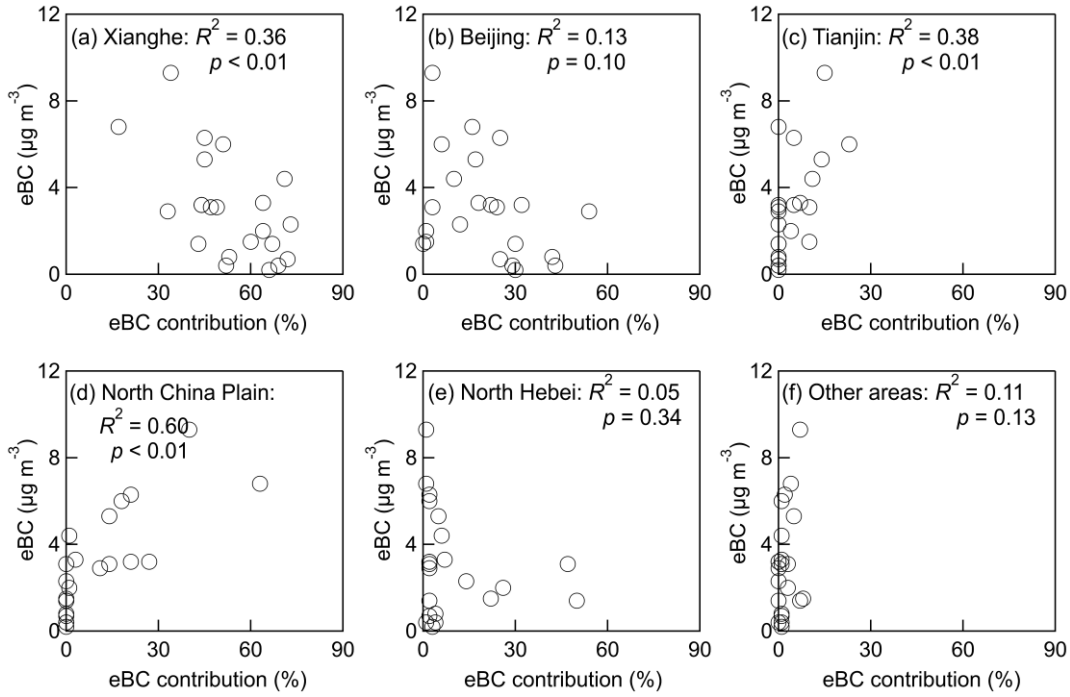

Figure R3. Scatter plots of the measured mass concentrations of equivalent black carbon (eBC) versus the eBC contributions of different source regions obtained by WRF-Chem model.

(13) Figure 6. It would be useful to replace the Region 1-6 labels shown in the figure with the names of the regions used in the text (or include this information in the caption).

Response: Following the reviewer's suggestion, we added this information in the figure caption. Please see the Figure R4 below (also see Figure 7 in the revised manuscript).

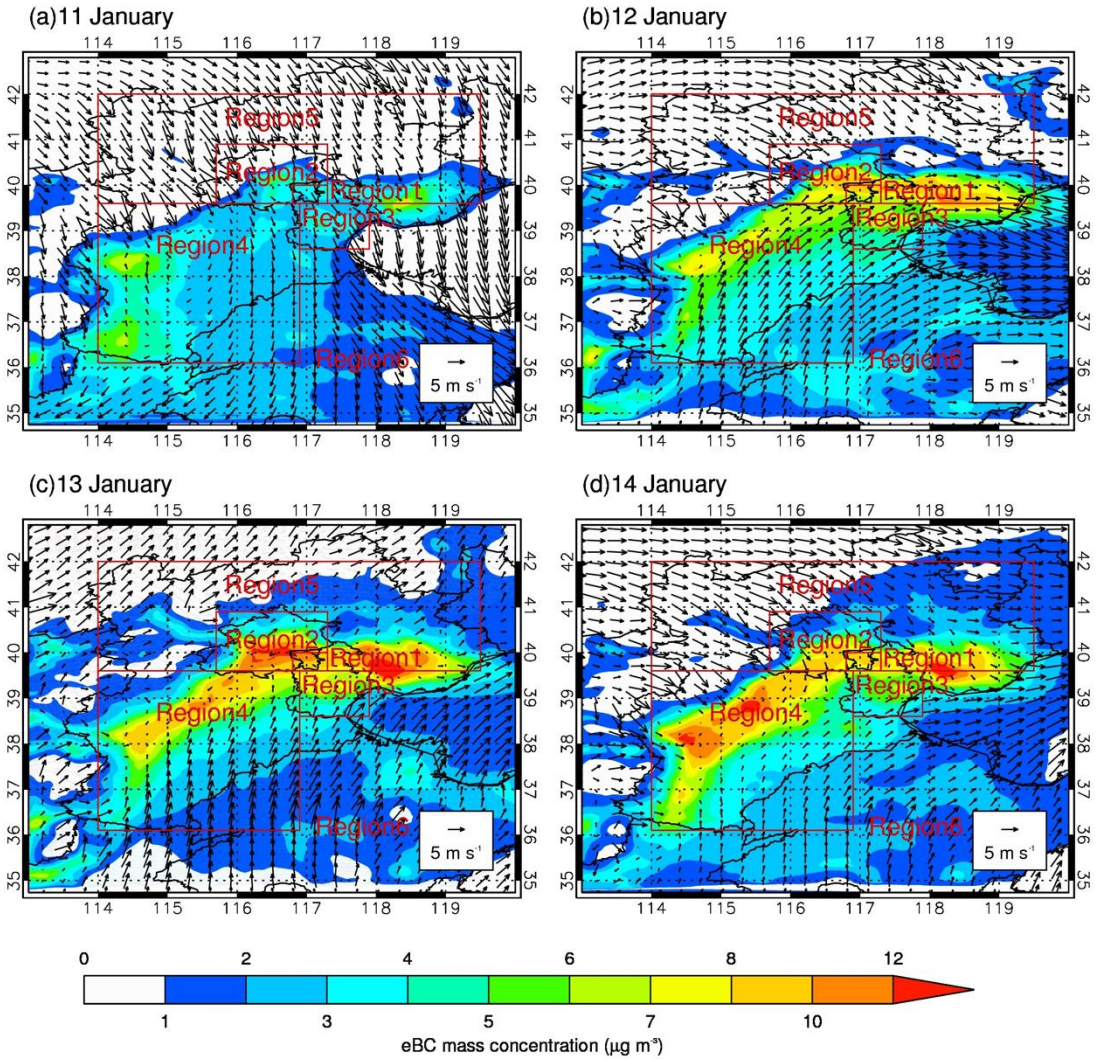

Figure R4. Distributions of the daily average mass concentrations of equivalent black carbon (eBC) (μg m^{-3}, represented by the color bar) at Xianghe and surrounding areas from 11th to 14th January, 2018 simulated by WRF-Chem model. The arrow denotes the wind speed. The red rectangles represent different source regions, which Region 1 is Xianghe, Region 2 is Beijing, Region 3 is Tianjin, Region 4 is North China Plain, Region 5 is North Hebei Province, and Region 6 is other areas except Region 1–5.

(14) Figure 8. It is unclear what the percentile range on the x-axis refers to. Is this with respect to the BC mass loading?

Response: Yes, the percentile range is the BC mass loading of different sources. We revised this figure to make it clearer. Please see Figure R5 below (also see Figure 9 in the revised manuscript).

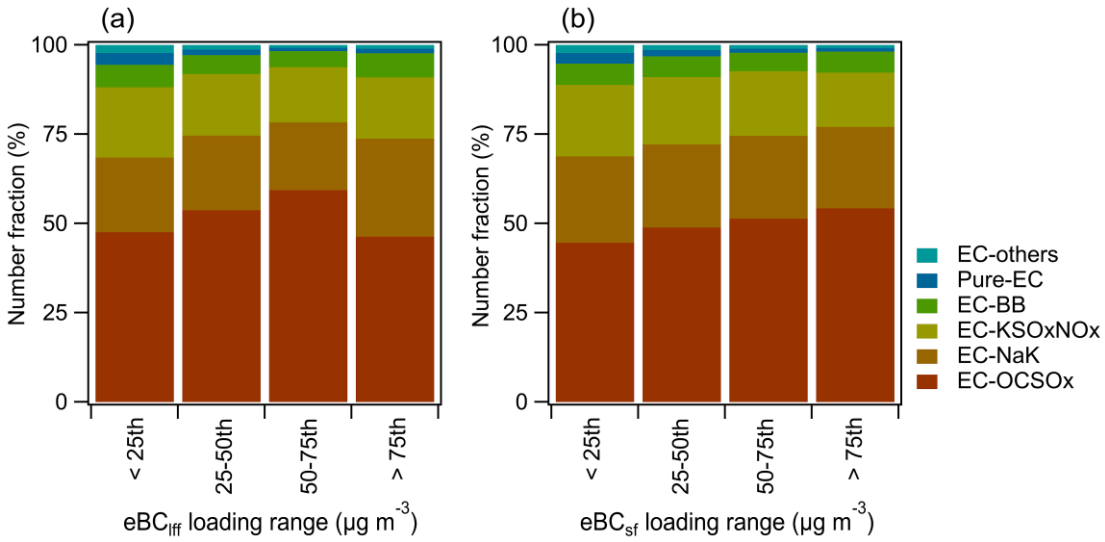

Figure R5. Number fractions of elemental carbon (EC)-containing particle classes at different loading ranges of equivalent black carbon (eBC) from sources of (a) liquid fossil fuels (eBC_{lff}) and (b) solid fuels (BC_{sf}). The 25th, 50th, and 75th denote the 25%, 50%, and 75% percentiles, respectively.

(15) It would be useful to discuss the results in Section 3.3 in the context of the different BC source emissions discussed in Section 3.1

Response: We thank the reviewer's suggestion. As shown in Figure R5 above, we actually discussed the characteristics of chemical composition of EC-containing particles in the context of the different eBC sources in the original manuscript. We revised the manuscript in this part to make the discussion clearer. It now reads as follows:

"Fig. 9 shows the number of fractions of each class of EC-containing particles at different ranges of eBC_{lff} and eBC_{sf}. The EC-OCSOx number fraction increased as eBC_{sf} increased. In contrast, it dropped when eBC_{lff} was higher than the value of the 75th percentile of BC_{lff}. This indicated a greater impact of solid fuel source on EC-OCSOx at a high eBC loading environment compared to the liquid fossil fuel source."

"The number fraction of EC-NaK increased with an increase in BC_{lff} but kept stable with BC_{sf} as shown in Fig. 9. These results demonstrate that EC-NaK was

likely associated with fresh traffic emissions than from solid fuels."

(16) Are the results in Section 3.4 from the WRF-Chem model? It is not clearly stated in the text.

Response: The results of radiative effect in Section 3.4 were estimated from the SBDART model. We added a sentence at the beginning of this paragraph to clarify this in the revised manuscript:

"Fig. 11 shows the eBC DRE variations as estimated by the SBDART model."

(17) Also it would be useful to provide some context for this estimation of the BC radiative effect in terms of previous calculations in the literature for the NCP region.

Response: We followed the reviewer's suggestion and added some comparisons of BC radiative effect from previous studies. It now reads as follows:

"In contrast, eBC exhibited a DRE range of +0.6 to +20.8 W m^{-2} with an average of +4.4 ± 3.0 W m^{-2} at the TOA, indicating a net energy gain and warm effect. This was attributed to the strong BC light absorption property that can impede the back scattered radiation reaching the TOA. The eBC DRE at the TOA in this study was comparable to the value over the NCP region (+6 to +8 W m^{-2}, Li et al., 2016)."

"Compared to previous DRE obtained from the SBDART model, the atmospheric DRE derived by eBC values in this study (+18.0 ± 9.6 W m^{-2}) was comparable to that of South China (+17.0 W m^{-2}, Huang et al., 2011) but was lower than that of Northwest China (+16.6 to +108.8 W m^{-2}, Zhao et al., 2019)."

References:

Heald, C.L., Ridley, D.A., Kroll, J.H., Barrett, S.R.H., Cady-Pereira, K.E., Alvarado, M.J. and Holmes, C.D., 2014. Contrasting the direct radiative effect and direct radiative forcing of aerosols.

Lack, D.A., Moosmüller, H., McMeeking, G.R., Chakrabarty, R.K. and Baumgardner, D., 2014. Characterizing elemental, equivalent black, and refractory black carbon

aerosol particles: a review of techniques, their limitations and uncertainties. Analytical and bioanalytical chemistry, 406(1), pp.99-122.

Response: We thank the reviewer providing relevant references, and we have added them in the revised manuscript.