Anonymous Referee #1

The manuscript by Wang et al. focused on the source apportionment and mixing state of black carbon in the North China Plain. The objectives of the study were to (a) determine the contributions of different sources and regions to the BC mass, (b) identify the chemical composition of BC coatings, and (c) evaluate the impacts of BC on regional radiative forcing. The authors used various methods to achieve these objectives. The major methods include a multi-wavelength optical approach combined with the source-based absorption Ångström exponent, WRF-Chem model, ART-2a, and SBDART model. The authors found that (a) the contribution of traffic emission was dominant to BC mass in the study region; (b) BC-containing particles existed in six classes to the mixing state of organic and inorganic substances; (c) the estimated BC forcing was positive with +18.0 W m^{-2} and a heating rate of 0.5 K day^{-1} in the study region. The designed experiments were comprehensive, and the results were robust. The new data generated from this study should be valuable to understand the present status of the regional air quality and radiative forcing affected by BC in the North China Plain under the background of emission reduction. Therefore, I would suggest the manuscript for publication after the authors address the following specific comments.

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

(1) Although a photoacoustic instrument was used to correct the impact of filter matrix scattering of AE33 aethalometer, the PAX was operated at a single wavelength of 532 nm, which may bring uncertainty for other wavelengths of AE33 absorption. This should be pointed out in the study.

Response: We agree with the reviewer. In the revised manuscript, we added a sentence to address the this uncertainty. It now reads as follows:

"As shown in Fig. S2, a 520 nm wavelength of AE33 absorption was strongly correlated with the PAX absorption ($R^2 = 0.97$, $p < 0.01$). The slope of 2.57 was then used to correct the AE33 data. However, a single-wavelength-based correction method may result in underestimation at $\lambda = 370$ and 470 nm, and overestimation at $\lambda = 590$, 660, and 880 nm (Kim et al., 2019)."

(2) A Nafion dryer was used for ACSM. How about the AE33, PAX, and SPAMS? High ambient humidity can also influence the measurements of these instruments. A schematic of the measurement system should be used to show the setup of each instrument.

Response: The collected ambient aerosols were dried before measurements by all instruments. We followed the reviewer's suggestion and added a schematic of the instrumental setups. Please see Figure R1 below (also see Figure S1 in the revised supplemental material).

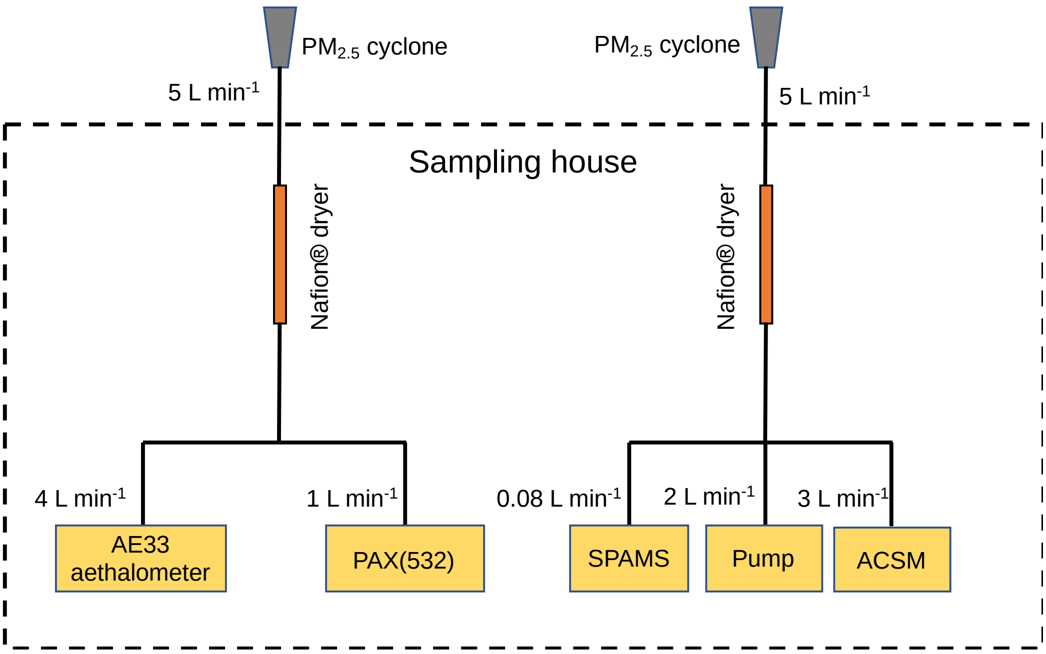

Figure R1. Schematic presentation of the instrumental setups of the ambient aerosol measurements.

(3) The authors should provide more information on bench tests, as the vehicle emission is a crucial component of the BC sources; relevant references should be cited.

Response: Following the reviewer's suggestion, we added more description about the bench tests in the revised manuscript. It now reads as follows:

> "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."

(4) Insufficient detail is given regarding the radiative forcing calculations. The radiative forcing is related to the vertical information of the atmosphere. The authors used the OPAC model to retrieve the vertical optical parameters used in the SBDART model, but the brief description makes the calculation unclear. More information about the OPAC model should be added in the supporting information.

Response: Following the reviewer's suggestion, we added more description of the OPAC model in the revised supplemental material (Text S3). It reads as follows:

> "The aerosol optical depth (AOD), single scattering albedo (SSA), and asymmetric parameter (AP) are important parameters used in the SBDART model to estimate aerosol radiative effect. In this study, these optical parameters were derived by the OPAC model. A detailed description of the software package of OPAC has been documented by Hess et al. (1998). The measured mass concentrations of OC, EC, and water-soluble ions as well as the estimated mineral dust (= [Fe]/0.035) were input in the OPAC model to estimate the AOD, SSA, and AP. The measurements of these chemical species are described in Text

S1. The number concentration of BC in the OPAC model was constrained by the measured EC mass concentration. Although several water-soluble ions and mineral dust were obtained, they did not cover all water-soluble and insoluble materials. Therefore, the number concentrations of water-soluble and insoluble materials were tuned for OPAC model based on the measured data. This was done by comparing the OPAC-derived light scattering, light absorption, and SSA with the corresponding PAX-measured ones until the differences were within 5% for each parameter. After the aerosol light extinction coefficient (sum of light scattering and absorption) was obtained, the AOD was then estimated as follows (Hess et al., 1998):

$$\mathrm{AOD} = \sum_j \int_{H_{j,min}}^{H_{j,max}} \sigma_{e,j}(h)dh = \sum_j \sigma_{e,j}^1 N_j(0) \int_{H_{j,min}}^{H_{j,max}} e^{-\frac{h}{Z_j}} dh \qquad \text{(S1)}$$

where $H_{j,max}$ and $H_{j,min}$ were the upper and lower boundary in layer j; $\sigma_{e,j}$ was the surface aerosol light extinction coefficient in layer j; h was the layer height; $\sigma_{e,j}^1$ represented the aerosol light extinction coefficient that was normalized to 1 particle cm^{-3}; N_j was the number concentration in layer j; and Z was the scale height. The OPAC-derived AODs were tuned to match the satellite-derived AODs (https://giovanni.gsfc.nasa.gov/giovanni) by altering the scale height in OPAC until the difference between them was within 5%. Owing to the closure with AOD and anchoring of the chemical composition, the assumptions in the OPAC model did not exhibit a significant impact on the estimation of radiative effect using SBDART model (Satheesh and Srinivasan 2006)."

(5) One finding of this study is the change of BC source in winter NCP through comparison with previous studies in this region. The authors considered this as a successful example of coal-to-gas switching to reduce pollutants under the new regulations by the Chinese State Council released in 2013. It would be useful to see a thorough comparison between the results of this study and those conducted before 2013 in this region. There should be relevant data from previous measurements by

others in this region.

Response: Following the reviewer's suggestion, we added BC source apportionment studies conducted before 2013 in the NCP region. This information was updated in Figure 4 in the revised manuscript (also see Figure R2 below).

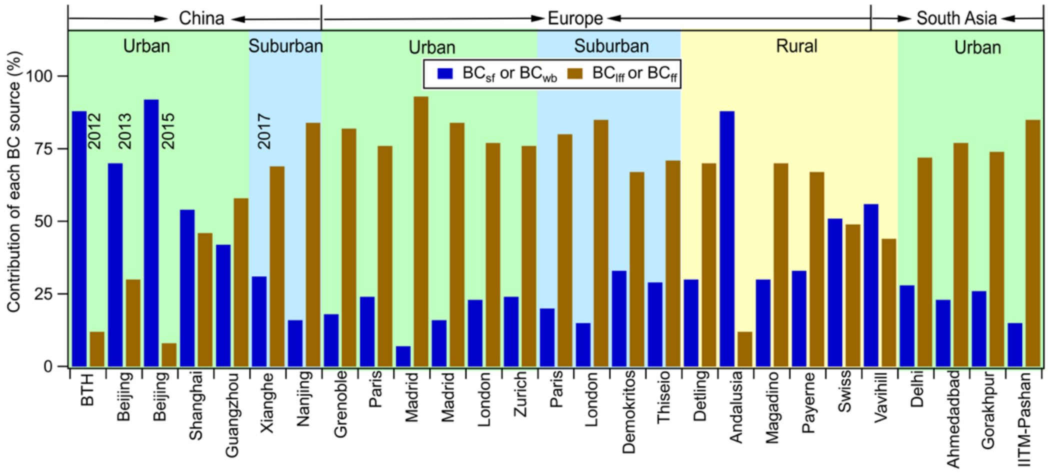

Figure R2. Comparisons of the different sources of black carbon (BC) in urban, suburban, and rural areas in China and Europe. BC_{sf} and BC_{wb} describe BC from solid fuel sources and wood burning, respectively. BC_{lff} and BC_{ff} represent BC from liquid fossil fuel and solid fossil fuel sources, respectively. Detailed information of the data is summarized in Table S1.

(6) The term 'Aethalometer model' gets misused in places. This refers to the data analysis technique, not the instrument itself.

Response: To make it clearer, we changed the "Aethalometer model" to "multi-wavelength optical method" in the revised manuscript.

(7) The map of Fig. S6 does not make much sense to someone unfamiliar with Chinese geography. Suggest showing a zoomed-out version showing the wider region.

Response: Following the reviewer's suggestion, we modified the map of Fig. S6 in the revised manuscript. Please see Figure R3 below (also see Figure S8 in the revised supplemental material).

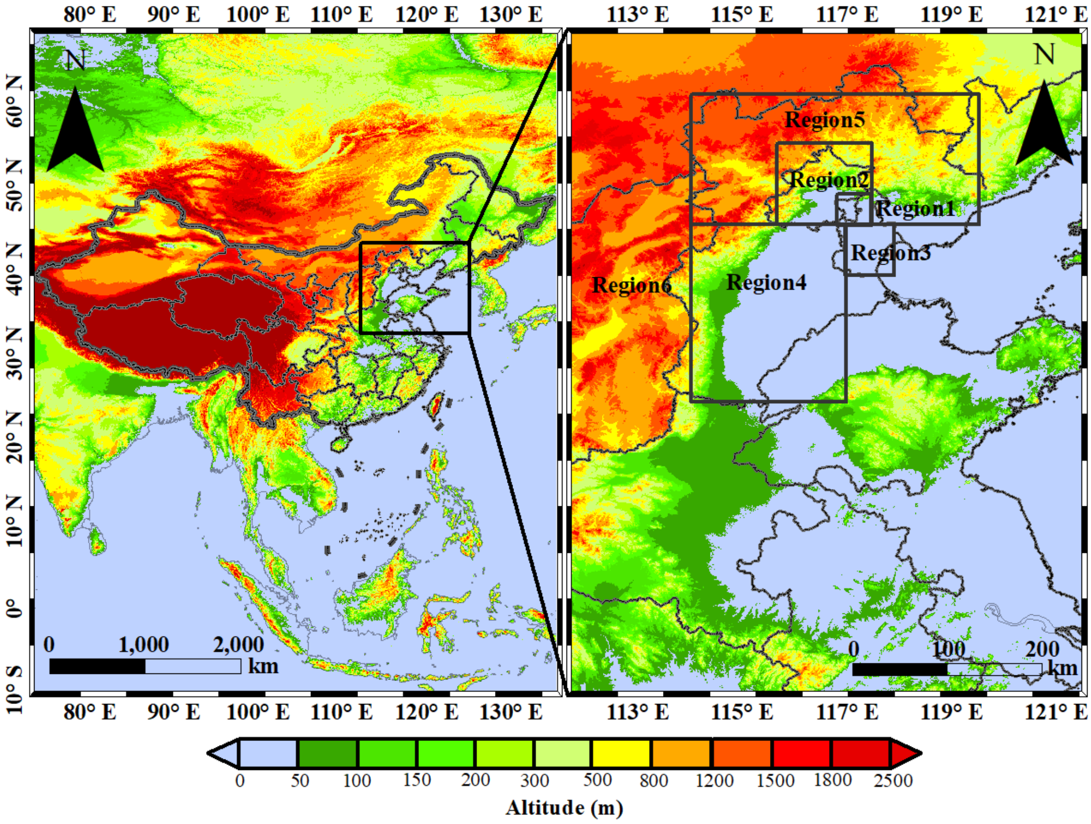

Figure R3. Division of different regions in the Weather Research and Forecasting model coupled with chemistry (WRF-Chem).

(8). The authors may improve the manuscript content by avoiding grammar and spelling errors throughout the text. I'm not going to list them, but the manuscript should be checked carefully and polished by a native English speaker.

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