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Interactive comment on "Size distribution and source of black carbon aerosol in urban Beijing during winter haze episodes" *by* Yunfei Wu et al.

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We appreciate the criticisms from the reviewer, although we cannot agree completely as explained below. We also appreciate the several useful comments which helped us improve the paper.

1. The reviewer felt that the scientific novelty value of our results was poor and mentioned that there was a similar dataset published by Wang et al. (2016). Moreover, the reviewer pointed out that there had been already several other articles containing SP2 measurement results published from Beijing and urban areas in Asia with longer data time series and much larger set of instruments, and results from these relevant studies have include the main results of our work. A longer dataset with data collected during different seasons and a larger set of instruments was recommended.

Response:

We are fully aware of the article by Wang et al. (2016a) in the preparation of our manuscript. Wang et al. (2016a) presented the SP2-rBC measurement that was collected right before our campaign in Beijing. The study only revealed the mass concentration of rBC derived from the SP2 measurement, and focused on the contribution of regional transport to the BC mass concentration in Beijing on the basis of WRF-BC model analysis combined with the SP2 measurement. The size distribution and mixing state of rBC particles, which are important knowledge gaps on rBC related sciences, were discussed in our study but not mentioned in their study. Thus, our study and their study have completely different focuses, although both focused on the same city. We'd also like to mention that there are two common coauthors in both papers whose contribution to our work was to provide guidance on instrument operation and data analysis. Our study is independent of that presented by Wang et al. (2016a) and we present more specific and advanced information of rBC that SP2 can provide, including size distribution and mixing state. We have reviewed many published SP2-related studies made in the urban areas in Asia, especially those in China. Studies on the size distribution of rBC, are relatively limited in China, e.g., Pearl River Delta Region (Huang et al., 2011, 2012), Yangtze River Delta (Gong et al., 2016), Xi'an in West China (Wang et al., 2015), Qinghai-Tibetan Plateau (Wang et al., 2014), and no similar study using SP2 was available in urban Beijing, or the whole North China Plain (NCP) where Beijing is located. Since rBC size distribution varied substantially from region to region, as reviewed in Huang et al. (2012), conducting such a study as presented in our manuscript is needed and filled the knowledge gap on this important scientific topic. As for the length of the campaign the reviewer concerned, we'd like to mention that measuring the rBC for a lengthy period with SP2 is challenging mainly due to its high cost, as well as complexity in operation and maintenance. Most in-situ SP2 measurements were performed in a period shorter than one-month (e.g., Liu et al., 2014; Huang et al., 2011; Wang et al., 2014; Gong et al., 2016; ...). Sometimes a number of instruments were operated accompanied with SP2 to show richer aerosol

characteristics. One of our previous work used a photoacoustic extinctiometer (PAX, DMT, USA) operating parallel to the SP2 to investigate the effect of mixing state on the mass absorption efficiency of rBC (Wu et al., 2016). The related in-situ experiment of the previous work was made at the same site in January 2013 when the heavy haze events occurred frequently in Beijing. However, our current research aimed to reveal the rBC size characteristic of rBC in urban Beijing. The following discussion on the sources of rBC in this study was mainly based on the measured rBC sizes. Actually, the size distributions of rBC is very similar in our two campaigns (the previous one was in the winter of 2013 and the current one was in the winter of 2014), because both measurements were made at the same site. They have been presented in Fig. S2 in the revised supplemental files. The slightly larger rBC sizes in the winter of 2013 is likely to relate with the higher rBC concentrations, which indicate more influence of larger rBC from sources other than local traffic, e.g., regional transport. In order to reveal the size distribution of rBC in urban Beijing and the causes of its variation, we focus on the campaign in the winter of 2014 in our current manuscript, during which the PM2.5 samples were collected twice per day and chemical compositions of each sample (including water-soluble ions, metal elements, carbonaceous matters) were comprehensively analyzed. In the discussion section of our current manuscript (Fig.4), we had already employed the dataset acquired in the winter of 2013 to verify the relationship between the number fraction of thickly coated rBC (NFcoated) and the average volume equivalent diameter of rBC (VEDrBC). Since the severe air pollution in Beijing is mainly occurred in wintertime, the rBC characteristics based on SP2 measurement in other seasons were seldom paid attention in our previous work. We will certainly consider the reviewer's suggestion in our future studies.

2. The reviewers mentioned that the develop method for analyzing the contribution of local traffic is poorly described and verified. Meanwhile, it is hard to estimate if it works. Proving the developed method works by comparing results of our method to other independent measurement data is recommended. The reviewer also suggests that it is necessary to estimate uncertainty and validate the method. Moreover, the

reviewer is also concerned with universality of this developed method.

Response:

We have described the method in more detail in the revised manuscript, according to the review's comments (Lines 398–400 in the revision). Generally, the method employed in our study is based on the extrapolative size of local traffic source from the linear relationship between the number fraction of thickly coated rBC (NFcoated) and the average volume equivalent diameter of rBC (VEDrBC). This diameter cannot be verified at current stage, because the directly measurement to rBC size of certain rBC source, e.g., traffic exhaust, is still lacking. However, as mentioned in section 4.1, through the analysis of the data acquired at the same site in the winter of 2013, we found that this relationship between NFcoated and VEDrBC and the inferred traffic-rBC diameter could be repeated. Moreover, several references associated with size-segregated aerosol samples were added in the revised manuscript (Lines 334–345 in the revision), because a similar BC size with diameter of $\sim$150 nm was also presented in these studies (Yu and Yu, 2009; Yu et al., 2010). Thus, we considered that this diameter actually existed in urban Beijing. Of cause, further work should be done to verify this result, especially the measurement nearby the emission sources and in other seasons. Actually, it is very difficult to distinguish the source of ambient rBC, as the rBC was easily to be internally mixed with other components during a short period in the ambient (Peng et al., 2016). Considering the chemical inertness of BC and invariance of the mass-equivalent size of the individual rBC particle in ambient atmosphere, we developed this simple method to estimate the contribution of local traffic to rBC. A more accurate source apportionment of rBC was presented by Liu et al. (2014) also using the difference in the size distribution of traffic-related rBC and solid fuel-related rBC. Their method is much more complicated. Based on the combination of PMF analysis, Wang et al. (2016b) developed a method to distinguish the rBC from traffic, coal combustion and biomass burning sources. A large amount of trace elements should be analyzed if the method was employed. Moreover, large uncertainties should also

be related in their method. The method developed in our study is relatively simple and requires only the SP2 measurement itself. We admit that large uncertainties existed in the resolved contribution of different sources to BC on the basis of our method, as explained in the revised manuscript. Several other methods, such as radiocarbon analysis, are generally used to determine the source of BC. However, the 14C analysis is mainly used to distinguish the contribution of biomass burning and fossil-fuel combustion to BC (Zhang et al., 2015; Liu et al., 2016). It is difficult to distinguish the traffic-related source from the fossil-fuel combustion based on the 14C analysis (see the response to the second major comment in RC1 for detail). Actually, the method developed in our study to resolve the rBC sources is more qualitative than quantitative, as several rough assumptions were employed in this method. However, results from this method should well reflect the variation in the traffic contribution. According to the results resolved from this method, it was much clearer that a significant increase in the contribution of sources other than local traffic, e.g., regional transport, was observed during the haze period. Based on many assumptions, uncertainty in the resolved traffic contribution should be very large and cannon be simply quantized here. As the sources of rBC are discrepant in different regions, we are not very sure this method is suitable in other studies. However, considering the rBC freshly emitted from the local traffic source is mostly non/thinly coated, the extrapolation of local traffic-rBC size by assuming it equal to the VEDrBC of totally non/thinly coated rBC should be tried in the typical urban regions at least. The rBC source apportionment method developed in our study can also be tried in the local traffic dominated urban areas where other rBC sources, e.g., coal combustion and biomass burning, are rare.

3. Why the diurnal cycles of the BC from both traffic and other sources are highest during nighttime?

Response:

As mention in section 4.2 in the initial manuscript, the higher BC concentrations during nighttime were likely to be attributed to the lower mixing layer height. Obvious decrease

in the mixing layer height suppressed the diffusion of air pollutants, resulting the higher BC from both traffic and other sources during nighttime. Another important cause of the higher traffic-BC during nighttime might be the increase in the flow of heavy-duty diesel vehicles. These vehicles have much higher emission factors of BC and are permitted to travel in urban Beijing only during the night time from 23:00 to 06:00 Local Time (Song et al., 2013). We have added such information in the revised manuscript with relevant references (Lines 432–436 in the revision).

4. Uncertainty estimation for BC results and especially for the larger mode would be important addition. The large BC mode (with maximum at around 500nm) is very small and very close to the upper particle size limit of the instrument, so it would be important to estimate how real this BC mode is.

Response:

Thanks for this recommendation. As mentioned in the methodology section, the overall uncertainty in the rBC mass determination was $\sim$25%, including the uncertainties inherented in the mass calibration, flow measurement and estimation of BC masses beyond the SP2 detection range. Indeed, the large BC mode is very small and very close to the upper size limit of the SP2. At the initial stage of analyzing this dataset, we also suspected the reliability of the large rBC mode. We then first consulted a large amount of relevant references and found that the large mode was also observed in several previous studies as mentioned in the first paragraph of section 3.1 in our manuscript (Huang et al., 2011; Wang et al., 2014). We still make reservations on whether the large rBC mode (only accounting for $\sim$6% of the SP2-determined rBC masses) was related to the inherent measurement bias, as it is close to the upper size limit of the SP2. Thus, we chose not to focus on the large mode in our manuscript discussion, and leave this to future more accurate measurements. Since there is no explicit interpretation to this large mode in literature, we simply present the observational facts from the dataset without much discussions.

5. The title and aim of this article are slightly conflicting. Title suggest that this article focuses on BC size distributions and sources, whereas the aim states that the aim was to develop a novel approach to evaluate the contribution of local traffic to the rBC concentration. Might be useful to include the method development to the title, if that is the main goal.

Response:

Actually, the major goal of this study is to reveal the size distribution of rBC and analyze its variation in urban Beijing based on the SP2 measurement. The source of rBC is a second goal of the study. We have modified the last paragraph of the introduction section to make the subject of this study clearer according to the suggestion (Lines 125–133 in the revision).

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-1096/acp-2016-1096-AC2-supplement.pdf

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