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Atmospheric Chemistry and Physics Discussions

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 greatly appreciate the reviewer for providing very constructive comments which have helped us improve the paper. We have considered the comments carefully and revised the manuscript accordingly, as detailed below in our point-to-point responses to the specific comments.

Major comments 1. It looks like the whole source apportionment is centered on the regression analysis of VED (rBC) versus thickly coated particles, resulting in traffic related rBC particles of 150nm in size. This result is very central to the main findings of the study – source contribution of traffic related BC particles. However, no justification whatsoever is provided what defines thickly coated particle and, consequently, what impact it would have if criterion of thickly coated particle is varied (I believe the criterion

is the ratio of equivalent diameters of the core and the particle). Overall, the method based on regression analysis is neglecting the fact that particles are rarely externally mixed, except very close to the source which in case of traffic is a car tailpipe. Further away from the source, spatially and temporarily, particles become internally mixed and sources combined (through coagulation, secondary processes and deposition) making it very difficult to justify whether 150 nm particle is indeed traffic related or some of them advected from a population of coal combustion particles (sub-population of smaller coal combustion particles). There is no firm justification that 150 nm particles are indeed originating from traffic only (why not e.g. 120 nm) and indirect evidence provided is not sufficient.

Response:

(1) The reviewer is concerned with the definition of 'thickly coated rBC-containing particles', and suspects the definition might have impact on the results presented in our manuscript. In our initial manuscript, we simply introduced the method to define the 'thickly coated rBC-containing particles' according to the delay time between SP2 detected incandescent and scattering signal peaks of individual particle. Since this method was described in detail in literature (Schwarz et al., 2006; Moteki and Kondo, 2007) and has been widely used in SP2 related studies (e.g., Wang et al., 2014; Wu et al., 2016; ...), there is no need for a detailed description in our study. We have added a short description in the revised manuscript to make the definition of 'thickly coated rBC-containing particles' clearer, which reads: "The rBC-containing particles were defined as either thickly coated or uncoated/thinly coated according to the distribution of detected lag times, which was bimodal and had a local minimum at 2 μs (Fig. S1 in the supplemental files). We defined the rBC particles as thickly coated if the lag times were longer than 2 μs" (Lines 162–166 in the revision). In addition, the method to distinguish the mixing status of rBC-containing particles used in this study is traditional and qualitative, which don't need the assumptions on the particle's physical and chemical properties (e.g., morphology and refractive indices). This method cannot give

the exact diameter of the particles, which is different from the method according to the equivalent diameter of rBC core and optical size of the entire particle. The delay time method can only estimate whether the particle is thickly coated and cannot provide how thick the coating is. Although the definition of 'thickly coated' based on the delay time method is related to the ratio of equivalent diameters of the core and the particle, it is not entirely decided by the absolute ratio of the diameters. Moteki and Kondo (2007) indicated that at least a coating thickness of 70% was needed to reach the delay time threshold for 'thickly coated particles'. Laborde et al. (2012) also illustrated the delay time threshold was expected to occur somewhere in the range of ∼70% coating volume fraction. The reviewer mentioned that particles are rarely externally mixed, except very close to the source which in case of traffic is a car tailpipe. We also illustrated that the rBC particles freshly emitted from traffic can be quickly coated (Line 70 in our initial manuscript). It took ∼4.6 hours for the particles thickly coated by other component under the ambient conditions in Beijing (Peng et al., 2016). In this study, we are concerned about the sizes of rBC which are relatively stable in the atmosphere. The mixing states of rBC particles provide an auxiliary description in the discussion on the sources of rBC. We don't focus on the mixing state of rBC particles and how thick the coating is. Thus, the qualitative method on the basis of delay time between incandescent and scattering signal peaks was employed. (2) As the reviewer proposed, there is indeed no further firm justification to prove the typical mass equivalent diameter of rBC is ∼150 nm for traffic source at current stage of our study. We tried to investigate the sizes of rBC directly measured using the similar method at typical sources, such as traffic, coal/biomass combustion from existing literatures. Unfortunately, to the best of our knowledge, we cannot find available supporting materials. However, the mass size distribution of elemental carbon (EC) from the size-segregated sample revealed a mode with peak at 150 nm in aerodynamic diameter for freshly emitted EC (Yu and Yu, 2009; Yu et al., 2010) in PRD region. Huang et al. (2006) reported a relatively smaller accumulate EC mode with peak at ∼120 nm. However, in these studies, a much larger traffic-related EC size was also observed with peak diameter at ∼400 nm,

which was seldom observed in the urban area and tunnel in developed countries. Yu et al. (2010) considered this characteristic of EC size distributions is related to the result of high engine loads and low combustion efficiencies in Chinese vehicles. We don't know whether the size distribution of EC in Beijing is similar to that in PRD region as there were few studies measured the EC in such small sizes (i.e., with diameter < ∼400 nm). Considering that the urban location of our site and the stringent fuel and vehicle emission standards implemented in urban Beijing, relatively higher combustion efficiencies can be expected, resulting in a smaller EC mode. On this basis, we considered the smaller EC mode with diameter of ∼150 nm should be dominated at our site, although further direct measurement is needed. In the revised manuscript, we have added the citation of these relevant literatures (Lines 334–345 in the revision). In our study, this typical local traffic-related VED with diameter of ∼150 nm was extrapolated from the linear regression of VEDrBC against NFcoated. The physical meaning can be expected from the extrapolation as the freshly traffic-emitted rBC are rarely thickly coated. During the analysis of experimental data, we also suspected the robustness of this linear regression and the deductive VED of traffic-emitted rBC. Thus, we analyzed the data obtained in another independent campaign conducted in winter 2013. It was very interesting to find there was a similar linear relationship between VEDrBC and NFcoated. Meanwhile, the slope and y-intercept values of the linear regression in the two independent campaigns were almost the same (Fig. 4). On this basis, we considered that the similar linear regression might be commonly existed, although more observations are needed to further confirm this result. Meanwhile, the direct measurement of rBC sizes at the traffic emission source is also needed to give a more reliable evidence. At the current knowledge status, the assumption made in this study should be acceptable.

2. Second major problem is the absence of method validation. One obvious validation would be radiocarbon analysis if that was considered at the start of the study. As it stands, the authors should at least do a thorough analysis of the studies (primarily but not exclusively in China) where radiocarbon analysis has been done and BC has been

isotopically apportioned. If the SP2 measurements were combined with radiocarbon analysis it would constitute a significant advancement.

Response: This comment proposed by the reviewer is meaningful and farsighted. It provides a direction for our further studies on BC sizes and apportionment. Unfortunately, we didn't perform the radiocarbon analysis in this study. Although the aerosol samples were collected in the campaign, there were not enough carbon amount for radiocarbon analysis in the laboratory as a low-volume sampler operated at a nominal flow rate of 16.7 L/min was utilized. According to the reviewer's suggestion, we have reviewed literature associated with BC apportionment based on radiocarbon analysis. The 14C analysis was usually used to distinguish the fossil-fuel combustion and biomass burning elemental carbon (EC). Zhang et al. (2015) revealed 76% EC was attributed to fossil-fuel combustion in urban Beijing in the extreme winter haze episode of 2013 on the basis of 14C analysis. The 14C analysis for the samplers collected at a suburban site of Beijing before/during/after the Asia-Pacific Economic Cooperation (APEC) summit held in November 2014 revealed a relatively higher contribution of biomass burning to EC (36% / 46% / 33%). The significant decrease in the contribution of fossil-fuel combustion during APEC was related to the control of industry and traffic emissions (Liu et al., 2016). However, it is difficult to distinguish the traffic-related source from the fossil-fuel combustion based on the 14C analysis. Stable carbon isotopes of traffic source showed a distinguished range from coal combustion (Widory, 2006; Cao et al., 2011), it was also very difficult to identify the exact contribution of traffic to EC due to the wide range of stable carbon isotopes. Generally, the radiocarbon analysis is advantage to distinguish the fossil-fuel and biomass burning source, but incapable of separating traffic-related sources from the fossil-fuel combustion. On the basis of SP2 measurement, the contribution of fossil-fuel combustion (traffic and coal burning) contributed $\sim$80% of rBC in Xi'an (Wang et al., 2016), close to the apportionment result based on the 14C analysis (78%, Zhang et al., 2015). The apportionment results were comparable, even the completely different approaches were employed. The traffic related rBC was determined in Wang et al. (2016), with a mean contribution

of 46%. It was not much different from our result where the local traffic contributed 59% to rBC on average. We will try to combine the two methods (i.e., SP2 and radiocarbon analysis) in future studies to give a more specific and reliable apportionment of BC.

Minor comments 1. Line 32. Why exactly VED would vary little once rBC is emitted into atmosphere when the time scale of secondary processes is many hours or as long as the particle is airborne and particles undergo cloud processing and dry/wet deposition?

Response: We meant that the VED of an individual rBC particle varied little during its lifetime in the atmosphere because the rBC is chemically inert under ambient condition. Although cloud processing will change the physical/optical size of the rBC-containing particle through coating of other components (e.g., organic matters, sulfate, ...), the mass-size (i.e., the VED) of the rBC core varies little. Moreover, the typical lifetime of BC is $\sim$1 week in the atmosphere (Bond et al., 2013), and ultimately removed from the atmosphere through wet (e.g., in precipitation) and dry deposition to the Earth's surface. Because there is no industrial rBC emission in urban Beijing, traffic should be one of the most important local sources of rBC. The emission intensity of vehicles is relative stable, resulting a constant VED of rBC measured in our study if other sources effect little. Thus, the variation in VED of rBC should be greatly interpreted by the alteration in rBC sources.

2. Line 129. Introduce assumption(s) briefly here.

Response: We have illustrated the assumptions in our revised manuscript according the comment, which reads: "a novel approach was employed to evaluate the contribution of local traffic to the rBC concentration based on the measured rBC sizes and reasonable assumptions including a deductive mean diameter of rBC from local traffic and relatively stable rBC sizes in the air masses transported over certain regions." (Lines 129–133 in the revision)

3. Line 162. Provide a number for defining thickly-coated rBC particles. What constitutes "thickly"?

Response: As we mention in the response to the first major comment, the 'thickly coated rBC particle' defined here was a qualitative conception. It was distinguished using the delay time between SP2 detected incandescent and scattering signal peaks of an individual particle, instead of directly characterize the size of rBC and its coating. This method is relatively easy to operate and doesn't rely on the assumptions of particle physical/chemical properties, which has been widely used in SP2-related studies (e.g., Moteki and Kondo, 2007; Wang et al., 2016). Previous studies revealed that at least a coating thickness of 70% was needed to reach the delay time threshold for 'thickly coated particles' (Moteki and Kondo, 2007; Laborde et al., 2012). As the observation site is not close to the emission sources (e.g., traffic road), naked rBC without coating should be seldom observed. Due to the quick coating processes of freshly emitted rBC from traffic, there should be a great part of thinly coated rBC under traffic dominant condition. These thinly coated rBC particles were classified together with the naked ones, significantly distinguished from the thickly coated ones which were underwent sufficient coating processes in the atmosphere or from sources such as coal combustion/biomass burning. Once the regional transport dominated, a great fraction of thickly coated rBC should be observed because the rBC particles were initially thickly coated from coal combustion/biomass burning sources and suffered from coating processes during their transport to the observation site. We don't concern about how thick the coating is in this study.

4. Line 167. "were" instead of "was".

Response: Corrected.

5. Line 180. When something mentioned for the first time, spell it out. PRD = Pearl River Delta?

Response: Corrected. We have also checked throughout the manuscript for other acronyms.

6. Line 188. What was the reason of choosing particular density?

Response: The rBC density with value of 1.8 g/cm3 utilized in this study is referred to relevant literature (Bond and Bergstrom, 2006). This density was also widely used in SP2-related studies (e.g., Schwarz et al., 2013; Gong et al., 2016). There are also many studies chose the value of 2 g/cm3 for the rBC density (e.g., Wang et al., 2016). We have considered the difference in rBC density when compared to the size distributions shown in other studies.

7. Line 195. The relatively similar. . .

Response: We have modified the statement there in the revision according to the comment.

8. Line 207. This is clearly the most likely reason for different VED values. Consider also cloud processing ($\sim$15min time scale) initiating wet deposition.

Response: Actually, we also consider the dry/wet deposition of large rBC particles result in the small VED values of rBC measured at the remote regions where local emissions are rare. In the manuscript, we just cited the initial interpretation for the small rBC presented in the corresponding reference in which they considered the small rBC was result from the source. Please note that we are referring to the rBC core inside an aerosol particle while the reviewer frequently referred to the ambient aerosol. The rBC cores can be coated by other components through aging processes in the atmosphere regardless of the initial size of rBC cores. Coating alters the physical/chemical of the initial rBC particle, including the geometric/optical size and the hydrophilicity of the particle in which the rBC core is embedded. Thus, the coated rBC particles should all undergo the cloud processing no matter the initial rBC cores are large or small.

9. Line 214. It is not much smaller, only smaller by 10%.

Response: The size distribution of rBC cores was presented in this study rather than the ambient aerosol particle. In this sense, 10% is significant. Aging processing changed the size of rBC-containing particles and had less impact on the mass equiv-

alent size of rBC cores. The difference in peak diameter of SP2 derived rBC during polluted and clean days was significant, although the relative deviation was ∼10% (221 nm on polluted day vs. 199 nm on clean day).Wang et al. (2016) also showed a significant difference in rBC peak diameter on coal combustion (215 nm) and traffic (189 nm) dominant days. In the revised the manuscript, we have used 'obviously' instead of the initial 'much' to make the statement here more rigorous.

10. Line 230. Generally, VED(rBC) were positively correlating with AS/EC and AN/EC ratios....

Response: We have modified the statement there in the revision according to the comment.

11. Line 242. Availability of ammonia is most likely responsible for such patter, because sulphuric acid is neutralized first and only then nitric acid (acid strength effect) if there is enough ammonia. Neutralisation with ammonia is a passive process. Try looking at differences in the degree of neutralisation.

Response: Generally, there was enough ammonia to neutralize sulphuric acid and nitiric acid in most samples except in several severely polluted samples (Fig. 1). Even in the severely polluted samples, the amount of ammonia was enough to neutralize sulphuric acid. In this study, we simply assumed there was enough ammonia to neutralize both sulphuric acid and nitiric acid. The mass concentrations of ammonia sulfate were calculated from those of sulfate multiplying by a factor of 1.375, and those of ammonia nitrate were from nitrate multiplying by a factor of 1.29, according to the molecular weight. The AS/EC ratios should be constant even the neutralization was considered because there was enough ammonia to neutralize sulphuric acid first. Although the AN/EC ratios in severely samples might decrease because there was not enough residual ammonia to neutralize nitric acid entirely, it had little effect on the results and conclusion there (Figure 2b and 2d in the revision).

12. Line 252. How could secondary AN have any effect on the core size of rBC when

formation processes of the two are completely different?

Response: The statement in the initial manuscript was indeed confusing. Actually, we intended to express that the secondary formation of AN played an important role in the coating processes of rBC while had negligible effect on the core size of rBC. The positive correlation between VED values of rBC and AN/EC ratios was resulted from the simultaneous effect of source alteration. Regional transport brought larger rBC particles as well as higher AN/EC. Secondary formation itself should not affect the core sizes of rBC. We have revised in the revised manuscript to this: "The secondary formation of AN might play an important role in the coating processes of rBC but have a negligible effect on the core size of the rBC."

13. Line 274. Reference London study.

Response: We have added the reference in the revision.

14. Line 315. "which has little coating. . ."

Response: Revised.

15. Line 316. Why is this surprising as large VED(rBC) would be formed in the presence of copious amounts of gaseous precursors contributing to thick coating during atmospheric processing (secondary formation)?

Response: The VED of rBC presented in this study indicated the mass equivalent size of rBC cores. Thus, even coating can increase the size of an entire particle in which rBC core was embedded, the mass size of rBC core should not vary. If only the coating processing had effect, the VEDrBC should vary little while NFcoated gradually increase. The linear relationship between VEDrBC and NFcoated should relate to the rBC source alteration. We are surprised that almost the same linear relationship was observed in another winter campaign, having similar slope and y-intercept values. Thus, we suspected the linear relationship between VEDrBC and NFcoated might be a common result at our observation site.

16. Line 351. The method ignores the fact that local traffic VED is including contribution of other sources unless the authors have access to specific experiments proving the traffic VED. Considering contribution of other sources to VED makes local traffic contribution biased high. The method works for the estimating the upper limit, but the real contribution can be very different without proper measurement of traffic emitted VED (rBC).

Response: We quite agree with the reviewer's comments here. Actually, the VED of rBC from local traffic was inferred from the observed linear relationship between VEDrBC and NFcoated, by assuming that the VEDrBC of non/thickly coated (i.e., non or thinly coated) rBC particles was from local traffic. As the freshly emitted rBC was gradually coated through aging processes in the atmosphere, the completely non/thinly coated rBC was seldom observed at an ambient site. The observed minimum NFcoated with value of $\sim$10% at 5 minutes resolution (Figure 4) also indicated that there was no absolutely non/thinly coated rBC unless measured close to the traffic source (i.e., car tailpipe). This VED presented in this study is an inferred value and cannot be directly proved at this stage because of the limitation in the direct measurement of traffic VED. As mentioned in the response to the first major comment, size-segregated EC distributions revealed a typical traffic emitted EC mode with mass aerodynamic diameter of $\sim$150 nm in China, although another traffic emitted EC mode with diameter of $\sim$400 nm was also reported which was seldom observed in urban areas of developed countries (Kleeman et al., 2000; Allen et al., 2001; Huang et al., 2006, 2008; Yu and Yu, 2009; Yu et al., 2010). Further direct measurement to the VED of rBC from certain source is needed. However, the interpretation in our study is scientifically sound. Coal combustion, biomass burning and traffic exhaust should be the most major sources of the airborne rBC. Due to the combustion condition and fuel type, the rBC particles from coal combustion and biomass burning are most thickly coated by organic or inorganic components. In contrast, rBC particles freshly emitted from traffic source are usually naked and become thinly coated in a short time after emitted to the atmosphere. They should take several hours or longer to get thick coating. Thus, on this basis, we consid-

ered the VED of completely non/thinly coated rBC corresponded to traffic induced rBC. It was further confirmed by a very similar result observed in another independent campaign. As we illustrated in the conclusion section in our revised manuscript, "despite potential large uncertainties in the estimated contribution from the local traffic to rBC, due to the many assumptions employed, its relative variation is clearly demonstrated." Further research measuring sizes of rBC directly from various sources, including coal combustion, biomass burning and traffic exhaust, is needed to validate the findings presented in this study.

17. Line 357. This is a very bold assumption when BC sources are not localized, but instead present in every region. Differences in VED in different regions depend on relative contribution of sources, but never fixed. Also consider heterogeneity of traffic sources as car fleet in major cities is very different from rural ones.

Response: Indeed, the assumption here is very bold. However, it is very difficult to identify the rBC sources when they are not localized. Carbon isotope method is suitable to identify whether rBC from fossil fuel or biomass burning, however, it is also difficult to distinguish the traffic source from coal or other fossil oil combustion. A simple method based on SP2 measurement was thus proposed in this study, although with uncertainties. We considered that the contribution of different rBC sources (coal combustion/biomass burning) in air masses from a certain direction should vary little since the emission factors should be spatially and temporally stable during a short period (e.g., several days). We didn't focus on how much the coal combustion/biomass burning contributed. As there were no sources of local coal combustion/biomass burning in urban Beijing, we treated the coal combustion/biomass burning as an ensemble source from regional transport which was distinguishable from local traffic according the rBC sizes. The rBC sizes from such ensemble source were mainly determined by where these rBC particles were from. Thus, the assumption here is reasonable to some degree. We presented the analysis of local traffic contribution as a discussion section, indicating there were inevitable uncertainties during the analysis. We cannot give a

definite conclusion at current stage. In addition, considering the urban location of our observation site, the local traffic source referred in particular to the traffic emissions on the roads in urban Beijing in this study.

18. Line 370. Five typical VEDs were identified in VED range of 5% only? This is unreasonable.

Response: Considering the relative difference of rBC peak diameter on polluted and clean days is only 10%, the VED range of 5% is acceptable. The sizes of rBC core are generally small and relative stable. Due to their chemical inertia, the rBC sizes are mainly determined by sources and depositions. The five typical VEDs estimated in this study represent the rBC sizes from sources other than local traffic. The rBC sizes from these sources should be relatively stable and influenced by the contribution of coal combustion/biomass burning as well as the deposition processing. We cannot distinguish the contribution of coal combustion and biomass burning from these sources other than local traffic. During our twenty days campaign, the emission inventory should vary little and significant wet scavenging wasn't observed. Thus, the difference with relative value of 5% can be expected.

19. Line 435. Those values must correlate, because they are methodologically related.

Response: Although the size and mass of rBC are methodologically related, the negative correlation between them is not inevitably. If the increase in mass concentration of rBC only induced by the meteorological condition (e.g., decrease in mixing layer height), the relative contribution of traffic to rBC should varied little. Change a way of thinking, if we used the mass concentration of PM2.5 measured using a real-time PM2.5 monitor instead of rBC, the contribution of traffic should also negatively correlated with the mass concentration of PM2.5 because the mass concentrations of PM2.5 and rBC are well positively correlated (Wu et al., 2016).

20. Why there is a gap in the spectrum?

Response: The gap is related to the methodology. The SP2 used in this study is revision C*. It has eight channels. The incandescence signals are simultaneously captured at high and low gain channels. The high gain channel provides a much sensitive measurement of small rBC particles while the low gain extend the up limitation of SP2. There is a merging of dataset recorded by the two channels, resulting in abnormal values at the junction. The gap in the spectrum is resulted from the artificial elimination of those abnormal values before curve fitting.

21. Fig.5 (and respective text). Sources are poorly separated, because of high traffic contribution around midnight. That is unreasonable and points at overestimated traffic contribution.

Response: The high rBC concentration of traffic in night time is greatly induced by the depressing of mixing height and suppressing of turbulence. Besides, although the vehicle number on the road decreases in night time, heavy-duty diesel vehicles are permitted to travel in the urban area of Beijing during the period of 23:00 to 06:00 (local time) under Beijing's traffic regulation (Song et al., 2012). They have much higher emission factors than light-duty gasoline vehicles, also resulting in the high traffic BC concentration around midnight (Song et al., 2013).

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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-AC1-supplement.pdf

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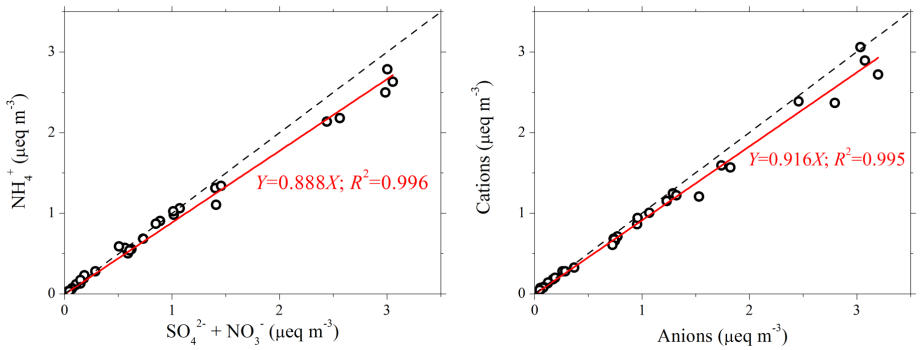

Fig. 1. Correlations between cations and anions: (a) NH4+ versus SO42-+NO3-; (b) Cations versus Anions