

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

Anonymous Referee #1

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The paper by Wu et al. is focused around BC bearing particle properties measured by state of the art instrument SP2. The authors did a reasonable job in trying to apportion BC to traffic and coal/biomass combustion sources, however, due to number of assumptions used the result is quite uncertain and unfortunately no validation is available. The paper can be considered for publication after addressing the comments below. Overall, the paper is fairly well written and executed, although a better job is expected regarding the uncertainties.

Major comments

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

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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.

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.

Minor comments

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?

Line 129. Introduce assumption(s) briefly here.

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

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Line 167. “were” instead fo “was”.

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

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

Line 195. The relatively similar. . . .

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

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

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

Line 242. Availability of ammonia is most likely responsible for such pattern, 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.

Line 252. How could secondary AN have any effect on the core size of rBC when formation processes of the two are completely different?

Line 274. Reference London study.

Line 315. “which has little coating. . . .”

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)?

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 con-

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tribution 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).

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.

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

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

Fig.1. Why there is a gap in the spectrum?

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.

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