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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aircraft measurements of black carbon in the boundary layer over the North China Plain" *by* Delong Zhao et al.

Anonymous Referee #2

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This paper describes airborne measurements of BC aerosol over the North China Plain near Beijing. They examine seasonal differences in concentrations, vertical distributions, size distributions and coating state and include a bit of meteorological analysis and run back trajectories. There is interesting data to present in this manuscript but the analysis should be re-worked to better support useful conclusions and the discussion includes too many statements that are pure conjecture rather than robust findings. I cannot recommend publication in ACP in its present form. In addition, significant editing is needed for English language and grammar. Below I include some comments which may help the authors re-focus and amend the analysis though this is not necessarily a comprehensive list of issues.

1. There is a lot of discussion of size distributions throughout but one is never actually

shown. I would recommend including at least a representative measured mass size distribution for BC. Also with respect to the size distributions, it does not look to me like the winter and summer PBL MMDs are significantly or systematically different. There is quite a bit of spread in the late spring values with a few flights showing markedly smaller MMDs than the others and seemingly bringing the whole average down. This probably is due to different sources as posited by the authors but I don't think there's any evidence that that is a seasonal behavior and not simply variability in transport. And transport may be more variable in the spring than in the winter but I don't really see how that is useful information. I don't think it's at all surprising that there are seasonal variations in BC sources, the question is what that means or what it can be used for.

2. All discussion of the coated fraction is rather a mess. First of all, the authors never explicitly discuss what they do with the SP2 data to derive F_coating. They often refer to it as the "evaporation time" of coatings which, if I understand what they did, is not at all the case. I believe they are looking at the time delay between the peaks in the scattering and incandescence signals in the SP2 (which is what was done in the papers referenced). This is always necessarily a bimodal distribution and the time delay has no bearing on the coating material or thickness. In the case of a bare BC particle, the peak in scattering will occur just prior to the onset of incandescence; prior to incandescence the signal is increasing because the particle is entering an ever more powerful section of the laser beam, upon incandescence the signal decreases because the particle evaporates. In the case of a coated BC particle, the scattering peak will have an initial maximum prior to the evaporation of coatings and a second maximum at the onset of incandescence. If coatings are thick enough the initial peak will be higher than the second peak and you will find a "time-delay" between the scattering and incandescence signals. In the case of thin coatings you will likely still have a local maximum at the point of coating evaporation but it will not be large enough to exceed the second scattering peak arising from the BC core and you will have no time-delay reported by the analysis software. The initial peak corresponding to coating

evaporation will happen at the same time relative to incandescence for every particle independent of coating material or thickness because the beam profile of the laser is very steep and the temporal resolution of the signal is not adequate to resolve tiny differences in coating evaporation rate. The exact coating thickness where the initial scattering peak will be larger than the peak at incandescence is a function of numerous factors including laser power, beam-width and BC core size so I am not convinced that the coated fractions reported for winter and spring are comparable (i.e. particles with 20nm of coating might be classified as "coated" in one phase while 40 nm of coating might be required to count as "coated" in the other phase such that a different F_coating value would be reported for identical aerosol populations depending on instrument alignment). The variations in F_coating with altitude within a given set of flights are probably usable although, again, I'm not sure that this is particularly useful information. Basically it seems like there is high variability in F_coating in the PBL, BC in the free troposphere is mostly uncoated and the entrainment zone is a mixture between the two. This is in contrast with previous findings where background BC typically has substantial coatings unless there has been a lot of wet removal (see a paper by Y. Kondo's group for example). The authors talk about "removal" but they don't make clear whether they actually saw evidence of wet removal. Can the back trajectories speak to this? Or are there nearby measurements of rainfall? I would be surprised if rain were frequent enough in both seasons that the background FT air had always recently undergone wet removal in all flights. It seems more likely that the variability in F_coating is driven by variations in sources and that the air identified as "Free troposphere" is actually also relatively recently impacted by emissions but from a different source. In the US, Europe and Japan traffic emissions are often thinly coated while biomass burning emissions are thickly coated almost immediately, perhaps what you have here is a local source dominated by biomass combustion mixing into a regional background consisting of more traffic and industrial emissions. In any case, in the absence of further evidence of what's driving these trends, I'm not sure I see the utility.

3. On a related note, with regard to the the trends in F_coating with RH: First, the au-

thors should clarify what is happening with RH during sampling. Is the sample stream dried prior to measurement with the SP2? If not there is likely some ram heating and associated evaporation during sampling but probably not enough to fully remove all the water. Some of the F_coating increase at high RH might be due to water uptake by the coatings. If the stream was fully dried prior to sampling then it seems most likely that the more coated sources are associated with higher RH than that the high RH drives formation of coatings.

4. Did the authors calibrate the optical sizing of the SP2 for either project? Please include that in the methods and discuss the effects of variability in the laser power and alignment.

5. Considerable space is dedicated to the determination of the different layers in the profiles (i.e. PBL, entrainment zone, free troposphere) but it's not clear to me what any of the reported trends mean. I can understand separating data into PBL and free tropospheric sampling but I think the "entrainment zone" is a bit of a distraction in the absence of significantly more detail w.r.t. cloud effects and precipitation. The authors calculate and discuss "removal efficiency" defined as the difference between BC in the PBL and the EZ but it seems that most of these differences are simply dilution. And while dilution does reduce concentrations it is certainly not the same as removal. I would recommend removing this section.

6. Can the authors compare these observed loadings in the PBL and FT to any previously published results? Many parts of the discussion are lacking in context. Similarly, these measurements seem to imply that BC size distributions (and possibly coating state) from Chinese transportation sources are very different than those from the US, Europe and Japan, could the authors definitively make this case and, if so, discuss the larger implications of this finding?

In summary, I think it is valuable to show BC concentrations, size distributions and coating state from an airborne platform over China, I just don't think this particular analysis

presents the data in a very usable format. As a first outcome of this study, it would be interesting to see the variability of these BC parameters as a function of location (or back trajectory origin) and as a rough function of altitude (separated into PBL - i.e. likely reflective of local sources - and free troposphere - reflective of background conditions). Starting from those basic observations it would be interesting if the authors could comment on larger issues such as the effects on radiation, the agreement of these observations with emissions estimates and/or a regional model, the columnar load of BC, etc. As presented there are simply a lot of correlations of different BC-properties with each other and it is never clear whether these relationships are mechanistic or coincidental.

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